

DESCRIPTION

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The invention relates to a positive-working light-sensitive mixture which, as the light-sensitive compound, contains a 1,2-naphthoquinone-2-diazide-sulfonic acid ester of a polyhydric phenol containing keto groups and
10 is intended in particular for the preparation of planographic printing plates.

Light-sensitive compounds of the generic type indicated are known from German Patent Specification 938,233 and U.S. Patent Specification 3,802,885.
15 These are di- or tri-hydroxybenzophenones which are partially or completely esterified with naphthoquinone-diazide-sulfonic acids. Amongst these compounds, the completely esterified representatives which no longer contain any free phenolic OH groups have been
20 preferred more recently. These compounds have a relatively high light-sensitivity and give long print runs, but they have the disadvantage of a solubility, which is unduly low for some purposes, in the organic solvents customarily used for the coating of printing
25 plates, such as partial ethers of glycol or carboxylic acid alkyl esters, such as butyl acetate. Moreover, in combination with the preferred binders which are insoluble in water and soluble in aqueous-alkaline

solvents, for example novolaks, they give layers which, in the unexposed state, are not sufficiently resistant to the usual alkaline developer solutions. These layers are also attacked to a certain extent during printing in offset printing presses, when fountain solutions containing alcohol are used.

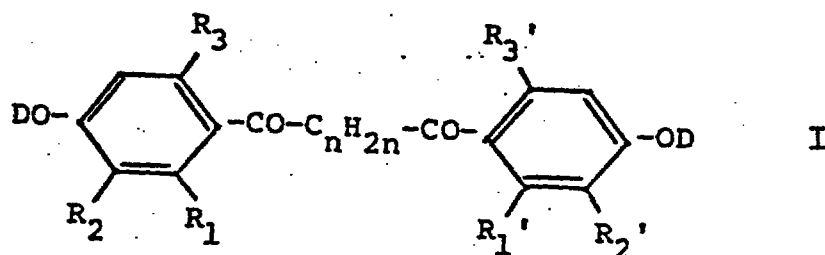
German Offenlegungsschrift 2,828,037 has disclosed mixtures which contain naphthoquinone-diazide-sulfonic acid esters of certain bisphenol derivatives in which two benzene rings are linked via an aliphatically substituted methylene bridge. These compounds have a high solubility in organic solvents. However, they do not yet have optimum resistance to alkaline developer solutions and to fountain solutions containing alcohol.

It was the object of the invention to propose a light-sensitive, positive-working mixture which, as the light-sensitive compound, contains a novel 1,2-naphthoquinone-2-diazide-sulfonic acid ester and which, in its properties with respect to printing technology and copying technology, is at least comparable with the best hitherto known mixtures of this type, but which has a higher resistance to aqueous-alkaline developer solutions and to the alcohol-containing fountain solutions used in offset printing, a hard gradation, a high solubility in organic solvents and a satisfactory oleophilic character and can be prepared in a simple manner in a chemically homogeneous form.

The subject of the invention is a light-sensitive

mixture which, as the light-sensitive compound, contains a 1,2-naphthoquinone-2-diazide-4- or -5-sulfonic acid ester of a polyhydric phenol containing keto groups.

5 In the mixture according to the invention, the naphthoquinone-diazide-sulfonic acid ester corresponds to the general formula I



10 in which D is a 1,2-naphthoquinone-2-diazide-5-sulfonyl or -4-sulfonyl radical, preferably the -5-sulfonyl radical, R_1 , R_2 , R_3 , R_1' , R_2' and R_3' are hydrogen atoms or halogen atoms, alkyl groups having 1 - 4 carbon atoms or radicals of the formula DO and n denotes a number from 2 to 18, preferably from 6 to 14, no more than
15 three DO radicals in total being present on one benzene ring.

According to the invention, a light-sensitive copying material composed of a support and a light-sensitive layer is also proposed, which layer contains
20 a 1,2-naphthoquinone-2-diazide-sulfonic acid ester of a polyhydric phenol containing keto groups, and comprises a naphthoquinone-diazide-sulfonic acid ester corresponding to the formula I defined above.

Amongst the compounds of the formula I, those
25 are preferred in which at least two DO groups are

present in each benzene ring. Derivatives derived from resorcinol, that is to say the compounds in which R_1 and R_1' are DO radicals and R_2 , R_3 , R_2' and R_3' are hydrogen atoms, are particularly preferred.

- 5 The preferred alkyl substituents are methyl and ethyl groups, in particular methyl groups. The halogen atoms are preferably chlorine atoms or bromine atoms.

- 10 The quinone-diazides contained in the mixture according to the invention are novel. They are prepared, analogously to known processes, by esterifying the phenolic OH groups with o-quinone-diazide-sulfonic acids or reactive derivatives thereof, for example the sulfonic acid chlorides.

- 15 The corresponding precursors, namely the diketophenols, are obtained, for example, by reacting corresponding phenols with dicarboxylic acids in the melt at about 140°C in the presence of anhydrous zinc chloride. The reaction time is here 2 to 5 hours.

- 20 Some of the diketophenols, listed in Table 1, prepared by the reaction of higher paraffin-dicarboxylic acids with resorcinol have already been described by J. v. Braun, Anton and Meyer in Chem. Ber. 74 (1941), 1772.

- 25 In some cases, the yield of diketophenol can be substantially increased if an excess of the phenol component (up to five times the stoichiometric quantity) is employed. In place of zinc chloride, boron trifluoride can also be used advantageously for this reaction. For this purpose, the reaction melt at 80 to

120°C is saturated with boron trifluoride for a period of 2 to 3 hours. For working up, the still liquid reaction mixture is poured into ice water, with good stirring. The reaction product which has precipitated is treated with N/10 sodium carbonate solution or ammonia solution, if necessary. Keto acid which may have formed and unconverted dicarboxylic acid go into solution, whilst the diketone compound remains.

Some of the diketophenols prepared from the corresponding phenols and dicarboxylic acids are listed in the table which follows. The diketophenols are colorless to yellow. Column 3 gives the melting points which were obtained after recrystallization from the solvents indicated in brackets.

Table 1

	Phenol component	Dicarboxylic acid	Melting point of diketophenol (0°(sic)C)
5	Resorcinol	Succinic acid	305 (pyridine)
	Resorcinol	Adipic acid	285 (methanol)
	Resorcinol	Suberic acid	187 (water/ acetone)
	Phenol	Sebacic acid	195 (ethanol)
10	Resorcinol	Sebacic acid	167 (water/ ethanol)
	Pyrogallol	Sebacic acid	184 (ethanol)
	Resorcinol	1,10-Decane- dicarboxylic acid	152 (toluene)
	Resorcinol	1,12-Dodecane- dicarboxylic acid	162 (toluene)
15	Resorcinol	1,14-Tetradecane- dicarboxylic acid	161 (glacial acetic acid)
	Resorcinol	1,16-Hexadecane- dicarboxylic acid	139 (glacial acetic acid)
	Resorcinol	1,11-Undecane- dicarboxylic acid	146 (glacial acetic acid)
	Pyrogallol	Succinic acid	> 300 (ethanol)
20	Phenol	Succinic acid	191 (glacial acetic acid)
	m-Cresol	Adipic acid	123 (ethanol)
	Resorcinol	Azelaic acid	142 (ethanol)
	Phloroglucinol	Sebacic acid	270 (glacial acetic acid)

The concentration of the novel naphthoquinone-diazide-sulfonic acid esters in the light-sensitive layer can vary within relatively wide limits. In general, the proportion is 3 to 50 % by weight, preferably between 7 and 25 % by weight, relative to the content of non-volatile constituents of the light-sensitive mixture.

Preferably, the light-sensitive mixtures according to the invention also contain a polymeric, water-insoluble resinous binder which dissolves in the solvents used for the mixture according to the invention and is also soluble or at least swellable in aqueous alkalis.

The novolak condensation resins, which have proved suitable in many positive-working copying materials based on naphthoquinone-diazides, were found to be particularly useful and advantageous as an additive also in the mixtures according to the invention, which contain the novel naphthoquinone-diazide-sulfonic acid esters. They promote a sharp differentiation between the exposed and unexposed layer areas on developing, and this is particularly true of the more highly condensed resins with substituted phenols, for example cresols, as the partners for the condensation with formaldehyde. Further alkali-soluble binders, or binders which are swellable in alkali, which should be mentioned are natural resins, such as shellac and colophony, and synthetic resins, such as copolymers of styrene and maleic anhydride or copolymers of acrylic acid or methacrylic acid, in particular with acrylates or methacrylates.

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The nature and quantity of the alkali-soluble resin can differ, depending on the intended purpose; proportions of between 95 and 50, particularly preferably 90 - 65, % by weight on total solids are preferred.

5 Additionally, numerous other resins can also be co-used, preferably epoxides and vinyl polymers, such as polyvinyl acetates, polyacrylates, polyvinyl acetals, polyvinyl ethers, polyvinylpyrrolidones and the copolymers of the monomers on which these are based. The most

10 advantageous proportion of these resins depends on the technological requirements and the influence on the development conditions, and in general it is not more than 20% by weight of the alkali-soluble resin. For

15 special requirements, such as flexibility, adhesion, gloss, coloration and color change and the like, the light-sensitive mixture can also contain small quantities of substances such as polyglycols, cellulose derivatives, such as ethylcellulose, wetting agents, dyes, adhesion-promoters and finely divided pigments, and also UV

20 absorbers, if required.

To coat a suitable support, the mixtures are in general dissolved in a solvent. The selection of the solvents should be matched to the intended coating process, the layer thickness and the drying conditions.

25 Ketones, such as methyl ethyl ketone, chlorinated hydrocarbons, such as trichloroethylene and 1,1,1-trichloroethane, alcohols, such as n-propanol, ethers, such as tetrahydrofuran, alcohol-ethers, such as ethylene glycol monoethyl ether, and esters, such as butyl acetate, are

suitable solvents for the mixture according to the invention. It is also possible to use mixtures which additionally, for special purposes, can also contain solvents, such as acetonitrile, dioxane or dimethylformamide. In principle, all those solvents can be used which do not irreversibly react with the components of the layer. Partial ethers of glycols, especially ethylene glycol monomethyl ether, are particularly preferred.

10 In most cases, metals are used as the supports for layer thicknesses of less than about 10 μm . The following can be used for offset printing plates: bright-rolled mechanically or electrochemically roughened and, if appropriate, anodically oxidized aluminum which can also
15 have been additionally pretreated chemically, for example with polyvinylphosphonic acid, silicates, phosphates, hexafluorozirconates or with hydrolyzed tetraethyl orthosilicate, and also multi-metal plates, for example those of Al/Cu/Cr or of brass/chromium. For
20 the preparation of letterpress plates, the mixtures according to the invention can be applied to zinc or magnesium plates, or to commercially available microcrystalline alloys thereof, for single-stage etching processes, and also to etchable plastics, such as poly-
25 oxymethylene. Due to their good adhesion and etch resistance on copper and nickel surfaces, the mixtures according to the invention are suitable for gravure printing forms or screen-printing forms. Likewise, the mixtures according to the invention can be used as

photoresists in the manufacture of printed circuit boards and in chemical milling. In further applications, other supports, such as wood, paper, ceramics, textiles and other metals can also be used.

5 The preferred supports for thick layers of more than 10 μm are plastic films which then serve as temporary supports for transfer layers. For this purpose, and for color-proofing films, polyester films, for example those of polyethylene terephthalate, are preferred.

10 However, polyolefin films, such as polypropylene, are also suitable.

 Coating of the support material is carried out in a known manner by spin-coating, spraying, dipping, rolling, by means of slot dies, blade application or
15 coater application. Finally, the coating of, for example, printed circuit boards, glass or ceramics and silicon discs, can also be effected by layer transfer from a temporary support. The light sources customary in industry are used for exposure. Electron or laser
20 irradiation is also a possibility for providing an image.

 The aqueous-alkaline solutions of stepped alkalinity, preferably with a pH in the range of 10-14, which are used for developing and which can also contain minor amounts of organic solvents or wetting agents,
25 remove those areas of the copying layer which have been struck by the light, and thus produce a positive image of the original.

 The preferred use of the light-sensitive mixtures according to the invention is in the preparation

of printing forms, that is to say in particular offset printing forms, autotype gravure printing forms and screen-printing forms, in liquid resists and in dry resists.

5 The printing plates prepared with the use of the novel compounds have a high light sensitivity in practice and an improved resistance to alkaline developers. The preferred compounds themselves are distinguished by very good solubility in the customary solvents, by a
10 good oleophilic character and alcohol resistance and by good compatibility with the other constituents of the copying layer.

 The invention is explained in more detail by reference to the examples which follow and in which parts
15 by weight (p.b.w.) and parts by volume (p.b.v.) have the same relationship as the g to the cm^3 . Unless otherwise stated, percentages are per cent by weight.

Example 1

 An electrolytically roughened and anodically oxidized
20 aluminum plate having an oxide weight of 3.0 g/m^2 was coated with a solution of

 1.10 p.b.w. of the esterification product obtained from
 1 mole of 1,10-bis-(2,4-dihydroxy-phenyl)-
 decane-1,10-dione and 4 moles of 1,2-
25 naphthoquinone-2-diazide-5-sulfonic acid
 chloride,

 5.70 p.b.w. of a cresol/formaldehyde novolak having a softening point of $105 - 120^\circ\text{C}$ according to DIN 53,181,

0.12 p.b.w. of 4-(p-tolylmercapto)-2,5-diethoxy-benzene-diazonium hexafluorophosphate,

0.40 p.b.w. of 2,3,4-trihydroxy-benzophenone and

0.60 p.b.w. of crystal violet in

5 40 p.b.w. of ethylene glycol monomethyl ether and

50 p.b.w. of tetrahydrofuran.

Before the application of the light-sensitive copying layer, the anodically oxidized aluminum support had been treated with an aqueous solution of polyvinylphosphonic acid, as
10 described in German Patent Specification 1,621,478.

The pre-sensitized material thus prepared, having a layer weight of the light-sensitive layer of 2.30 g/m², was exposed imagewise under a transparent positive original and then developed with the following solution:

15 5.3 p.b.w. of sodium metasilicate x 9 water,

3.4 p.b.w. of trisodium phosphate x 12 water and

0.3 p.b.w. of sodium dihydrogen phosphate (anhydrous) in
91.0 p.b.w. of water.

The portions of the copying layer struck by the
20 light were removed by the development, and the unexposed image areas remained on the support. On an offset printing press, approximately 200,000 perfect prints were obtained from the printing form thus prepared. The printing stencil was distinguished by an excellent
25 oleophilic character, alcohol resistance and adhesion to the support.

Whereas the solubility of the above-indicated naphthoquinone-diazide-sulfonic acid ester in ethylene glycol monomethyl ether is 42% by weight at 23°C, it

was possible, under the same experimental conditions, only to prepare a 0.4% solution, as a maximum, in ethylene glycol monomethyl ether from the esterification product of 1 mole of 2,4-dihydroxy-acetophenone and 2 moles of 1,2-naphthoquinone-2-diazide-5-sulfonic acid chloride.

In the examples which follow, further coating solutions are indicated, with which similar results were obtained. Unless specially noted, the preparation and processing of the printing plates thus obtained correspond to the conditions described in Example 1.

Example 2

An electrolytically roughened and anodically oxidized aluminum plate was coated with a solution of 1.10 p.b.w. of the esterification product obtained from 1 mole of 1,14-bis-(2,4-dihydroxy-phenyl)-tetradecane-1,14-dione and 4 moles of 1,2-naphthoquinone-2-diazide-5-sulfonic acid chloride, 6.10 p.b.w. of the novolak indicated in Example 1, 0.14 p.b.w. of 1,2-naphthoquinone-2-diazide-4-sulfonic acid chloride and 0.06 p.b.w. of crystal violet in 30 p.b.w. of ethylene glycol monomethyl ether, 25 40 p.b.w. of tetrahydrofuran and 10 p.b.w. of butyl acetate.

Example 3

An electrolytically roughened and anodically oxidized aluminum plate, which had been post-treated with an

aqueous solution of polyvinylphosphonic acid, was coated with a solution of

- 1.30 p.b.w. of the esterification product obtained from 1 mole of 1,8-bis-(2,4-dihydroxy-phenyl)-octane-1,8-dione and 4 moles of 1,2-naphthoquinone-2-diazide-5-sulfonic acid chloride,
- 5.80 p.b.w. of the novolak indicated in Example 1,
- 0.18 p.b.w. of 1,2-naphthoquinone-2-diazide-4-sulfonic acid chloride and
- 0.07 p.b.w. of crystal violet in
- 30 p.b.w. of ethylene glycol monomethyl ether,
- 40 p.b.w. of tetrahydrofuran and
- 10 p.b.w. of butyl acetate.

Example 4

- 15 An electrolytically roughened and anodically oxidized aluminum plate, which had been post-treated with an aqueous solution of polyvinylphosphonic acid, was coated with a solution of
- 1.10 p.b.w. of the esterification product obtained from
- 20 1 mole of 1,10-bis-(2,3,4-trihydroxy-phenyl)-decane-1,10-dione and 6 moles of 1,2-naphthoquinone-2-diazide-5-sulfonic acid chloride,
- 5.70 p.b.w. of the novolak indicated in Example 1,
- 0.12 p.b.w. of 4-(p-tolylmercapto)-2,5-diethoxy-benzene-
- 25 diazonium hexafluorophosphate,
- 0.40 p.b.w. of 2,3,4-trihydroxy-benzophenone and
- 0.06 p.b.w. of crystal violet in
- 40 p.b.w. of ethylene glycol monomethyl ether and
- 50 p.b.w. of tetrahydrofuran.

Example 5

- An aluminum foil mechanically roughened by means of a steel wire brush was coated with a solution of
- 1.00 p.b.w. of the esterification product obtained from 1 mole of 1,12-bis-(2,4-dihydroxy-phenyl)-dodecane-1,12-dione and 4 moles of 1,2-naphthoquinone-2-diazide-5-sulfonic acid chloride,
 - 6.40 p.b.w. of the novolak indicated in Example 1,
 - 0.20 p.b.w. of a polyvinyl acetal containing about 70% of vinyl acetal units, 24 - 27% of vinyl alcohol units and 1% of vinyl acetate units,
 - 0.12 p.b.w. of 1,2-naphthoquinone-2-diazide-4-sulfonic acid chloride and
 - 0.06 p.b.w. of crystal violet in
 - 30 p.b.w. of ethylene glycol monomethyl ether,
 - 40 p.b.w. of tetrahydrofuran and
 - 10 p.b.w. of butyl acetate.

Example 6

- An electrolytically roughened and anodically oxidized aluminum plate having an oxide weight of 3.0 g/m^2 was coated with a solution of
- 1.20 p.b.w. of the esterification product obtained from 1 mole of 1,10-bis-(2,4-dihydroxy-phenyl)-decane-1,10-dione and 4 moles of 1,2-naphthoquinone-2-diazide-4-sulfonic acid chloride,
 - 6.30 p.b.w. of the novolak indicated in Example 1,
 - 0.12 p.b.w. of 4-(p-tolylmercapto)-2,5-diethoxy-benzene-

diazonium hexafluorophosphate and
0.06 p.b.w. of crystal violet in
40 p.b.w. of ethylene glycol monomethyl ether and
50 p.b.w. of tetrahydrofuran.

5 Example 7

 An electrolytically roughened and anodically oxidized aluminum plate having an oxide weight of 2.0 g/m^2 was coated with a solution of

1.20 p.b.w. of the esterification product obtained from
10 1 mole of 1,10-bis-(4-hydroxy-phenyl)-
 decane-1,10-dione and 2 moles of 1,2-
 naphthoquinone-2-diazide-5-sulfonic acid
 chloride,
6.00 p.b.w. of the novolak indicated in Example 1,
15 0.10 p.b.w. of 1,2-naphthoquinone-2-diazide-4-sulfonic
 acid chloride,
0.20 p.b.w. of 2,3,4-trihydroxy-benzophenone and
0.06 p.b.w. of crystal violet in
35 p.b.w. of ethylene glycol monomethyl ether,
20 45 p.b.w. of tetrahydrofuran and
10 p.b.w. of butyl acetate.

Example 8

 An electrolytically roughened and anodically oxidized aluminum plate, which had been post-treated with an
25 aqueous solution of polyvinylphosphonic acid, was coated with a solution of

1.00 p.b.w. of the esterification product obtained from
 1 mole of 1,4-bis-(2,4-dihydroxy-phenyl)-
 butane-1,4-dione and 4 moles of 1,2-naphtho-

- quinone-2-diazide-5-sulfonic acid chloride,
 6.30 p.b.w. of the novolac indicated in Example 1,
 0.11 p.b.w. of 4-(p-tolylmercapto)-2,5-diethoxy-benzene-
 diazonium hexafluorophosphate and
 5 0.06 p.b.w. of crystal violet in
 40 p.b.w. of ethylene glycol monomethyl ether and
 50 p.b.w. of tetrahydrofuran.

Example 9

- This example describes a copying layer solution
 10 which can be used as a positive resist solution for
 various purposes, in particular for the preparation of
 etching stencils, for example in the chemical milling of
 shaped parts and labels, and in the preparation of printed cir-
 cuit images, and in particular also for micro-electronics.
 15 The solution had good stability. It was possible to
 match the type of coating and the layer thickness to the
 particular requirements within wide limits. After
 exposure, the exposed areas stood out with good contrast
 from the unexposed areas. The exposed areas could
 20 readily be removed by means of an alkaline developer (pH
 about 12.8).

- (sic) with a solution of
 1.5 p.b.w. of the esterification product obtained from
 1 mole of 1,10-bis-(2,4-dihydroxy-phenyl)-
 25 decane-1,10-dione and 4 moles of 1,2-naphtho-
 quinone-2-diazide-5-sulfonic acid chloride,
 0.4 p.b.w. of 4-(p-tolylmercapto)-2,5-diethoxy-benzene-
 diazonium hexafluorophosphate,
 0.1 p.b.w. of crystal violet (C.I. 42,555),

10.0 p.b.w. of the novolak indicated in Example 1 and
2.0 p.b.w. of an epoxy resin having an epoxy equivalent weight of 450 - 500 and a melting range of 64 - 74°C in

- 5 40 p.b.v. of ethylene glycol monomethyl ether,
50 p.b.v. of tetrahydrofuran and
10 p.b.v. of butyl acetate.

A plate of insulating material, laminated with a 35 μ m thick copper foil, was coated with this solution.

10 Example 10

A biaxially stretched and thermoset polyethylene terephthalate film was coated with a solution of
1.20 p.b.w. of the esterification product obtained from
1 mole of 1,10-bis-(2,4-dihydroxy-phenyl)-
15 decane-1,10-dione and 4 moles of 1,2-naphthoquinone-2-diazide-5-sulfonic acid chloride,
5.60 p.b.w. of the novolak indicated in Example 1,
0.12 p.b.w. of 4-(p-tolylmercapto)-2,5-diethoxy-benzene-diazonium hexafluorophosphate,

- 20 0.20 p.b.w. of 2,3,4-trihydroxy-benzophenone and
1.00 p.b.w. of crystal violet in
40 p.b.w. of ethylene glycol monomethyl ether,
40 p.b.w. of tetrahydrofuran and
10 p.b.w. of butyl acetate.

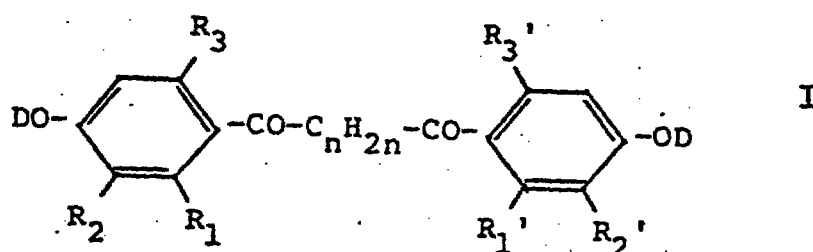
25 The colored film obtained was used for the preparation of make-up copies and register copies for the mounting of films and for mounting checks. Exposure and development were carried out as indicated in Example 1.



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Patent Claims

1. A light-sensitive mixture which contains, as the light-sensitive compound, a 1,2-naphthoquinone-2-diazide-4- or -5-sulfonic acid ester of a polyhydric phenol containing keto groups, characterized in that the naphthoquinone-diazide-sulfonic acid ester corresponds to the general formula I



in which D is a 1,2-naphthoquinone-2-diazide-5-sulfonyl or -4-sulfonyl radical, R_1 , R_2 , R_3 , R_1' , R_2' and R_3' are hydrogen atoms or halogen atoms, alkyl groups having 1 - 4 carbon atoms or radicals of the formula DO and n denotes a number from 2 to 18, no more than three DO radicals in total being present on one benzene ring.

2. A light-sensitive mixture as claimed in claim 1, which additionally contains a binder which is insoluble in water and soluble or at least swellable in aqueous-alkaline solutions.

3. A light-sensitive mixture as claimed in claim 1, which contains 3 to 50% by weight of naphthoquinone-diazide-sulfonic acid ester, relative to its content of non-volatile constituents.

4. A light-sensitive mixture as claimed in claim 1,

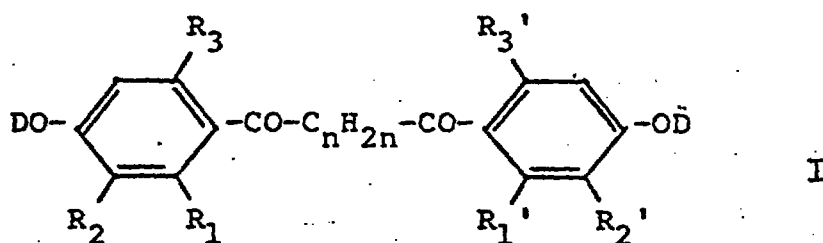
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which contains a compound of the formula I in which in each case at least one of the radicals R_1 , R_2 and R_3 or R_1' , R_2' and R_3' is a DO radical.

5. A light-sensitive mixture as claimed in claim 4, which contains a compound of the formula I in which R_1 and R_1' are DO radicals and R_2 , R_3 , R_2' and R_3' are hydrogen atoms.

6. A light-sensitive mixture as claimed in claim 1, which contains a compound of the formula I in which n is a number from 6 to 14.

7. A light-sensitive copying material composed of a support and a light-sensitive layer which contains a 1,2-naphthoquinone-2-diazide-sulfonic acid ester of a polyhydric phenol containing keto groups, characterized in that the naphthoquinone-diazide-sulfonic acid ester corresponds to the general formula I



in which D is a 1,2-naphthoquinone-2-diazide-5-sulfonyl or -4-sulfonyl radical, R_1 , R_2 , R_3 , R_1' , R_2' and R_3' are hydrogen atoms or halogen atoms, alkyl groups having 1 - 4 carbon atoms or radicals of the formula DO and n denotes a number from 2 to 18, no more than three DO radicals in total being present on one benzene ring.

8. A light-sensitive copying material as claimed in claim 7, wherein the support is composed of aluminium.

