

Australian Patent Declaration Form

Forms 7 and 8

COMMONWEALTH OF AUSTRALIA

Patents Act 1952

DECLARATION IN SUPPORT OF A CONVENTION OR NON-CONVENTION APPLICATION FOR A PATENT OR A PATENT OF ADDITION

In support of the application No. 63707/86 made by BASF LACKE & FARBEN AKTIENGESELLSCHAFT for a patent for an invention entitled:- HARDENABLE COMPOSITION

I/We, Dr. Wilfried Hoselmann of BASF Lacke + Farben AG with its address at Max-Winkelmann-Straße 80, 4400 Münster, Federal Republic of Germany

do solemnly and sincerely declare as follows:-

1. I am/we are the applicant(s) for the patent, or am/are authorised by the abovementioned applicant(s) to make this declaration on its behalf.
2. The basic application(s) as defined by Section 141 of the Act was/were made in the following country or countries on the following date(s) by the following applicant(s) namely:-
in Federal Republic of Germany on 30th September 1985
by BASF Lacke + Farben Aktiengesellschaft
3. The said basic application(s) was/were the first application(s) made in a Convention country in respect of the invention the subject of the application.
4. The actual inventor(s) of the said invention is/are
Werner Jung, Uhrwerkerstrasse 65, D-4715 Ascheberg, Germany
5. The facts upon which the applicant(s) is/are entitled to make this application are as follows:-

The said applicant is the assignee of the actual inventor.

DECLARED at Münster this 4th day of May

1988

[Handwritten signatures]

Dr. Hoselmann Dr. Dobbelstein
Prokurist Prokurist

PLEASE ADD NAME AND TITLE OF PERSON SIGNING, LEGIBLY.



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HARDENABLE COMPOSITION

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(56) Prior Art Documents
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(57) Claim

1. A curable composition containing a soluble hydroxyl-containing acrylate copolymer, a compound possessing at least two cyclic carboxylic anhydride groups per molecule and a compound possessing at least two epoxide groups per molecule, wherein the hydroxyl-containing acrylate copolymer is obtained from

- a1) from 10 to 60% by weight, of hydroxyl-containing esters of acrylic acid and/or methacrylic acid, where the alkyl radical is of two to 14 carbon atoms,
- a2) more than 3 to 30% by weight, of monomers having at least two polymerizable, olefinically unsaturated double bonds and
- a3) from 15 to 82% by weight, of further polymerizable monomers having an olefinically unsaturated double bond, the sum of components a1, a2 and a3 being 100% by weight.

29. A process for the preparation of a coating composition based on the curable composition according to any one of claims 1 to 28, wherein, in order to prepare the hydroxyl-containing acrylate copolymer

a1) from 10 to 60% by weight, of hydroxyl-containing esters of acrylic acid and/or methacrylic acid, where the alkyl radical is of 2 to 14 carbon atoms,

a2) more than 3 to 30% by weight, of monomers possessing at least two polymerizable, olefinically unsaturated double bonds and

a3) from 15 to 82% by weight, of further polymerizable monomers having an olefinically unsaturated double bond, the sum of components

a1, a2 and a3 being 100% by weight, are copolymerized in an organic solvent at from 80 to 130°C, using at least 0.5% by weight, based on the total weight of the monomers, of a polymerization regulator and using polymerization initiators, and the resulting acrylate solution, together with organic solvents, if appropriate pigments, fillers, conventional assistants, additives, the compound having at least two cyclic carboxylic anhydride groups per molecule, the compound having at least two epoxide groups per molecule and, if appropriate, a crosslinking catalyst, is processed to a coating composition by mixing and, if required, dispersing.

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Form 10

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Complete Specification for the invention entitled:
HARDENABLE COMPOSITION

The following statement is a full description of this invention including the best method of performing it known to me:-



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PCT

WELTORGANISATION FÜR GEISTIGES EIGENTUM
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A. D. J. P. 2 8 MAY 1987

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This document contains the amendments made under Section 49 and is correct for printing.

(54) Title: HARDENABLE COMPOSITION
 (54) Bezeichnung: HÄRTBARE ZUSAMMENSETZUNG
 (57) Abstract
 The above-described composition comprises a compound with at least 2 cyclic carboxylic acid anhydride groups per molecule, a compound with at least 2 epoxide groups per molecule and a hydroxyle group containing acrylate copolymerizate. The composition is obtained from a1) 10 to 60% by weight, preferably 15 to 60% by weight of acrylate and/or methacrylate resins with 2 to 14 carbon atoms in the alkyl residue, a2) more than 3 to 30% by weight, preferably 5 to 25% by weight of monomers with at least 2 polymerizable olefinic-unsaturated double bonds and a3) 15 to 82% by weight, preferably 40 to 70% by weight of other polymerisable monomers with an olefinic-unsaturated double bond, whereby the sum of the components a1, a2 and a3 amounts to a 100% by weight.
 (57) Zusammenfassung
 Eine härtbare Zusammensetzung, enthaltend eine Verbindung mit mindestens 2 cyclischen Carbonsäureanhydridgruppen pro Molekül, eine Verbindung mit mindestens 2 Epoxidgruppen pro Molekül und ein Hydroxylgruppenhaltiges Acrylatcopolymerisat, erhältlich aus a1) 10 bis 60 Gewichts-%, vorzugsweise 15 bis 60 Gewichts-%, Hydroxylgruppenhaltigen Estern der Acrylsäure und/oder Methacrylsäure mit zwei bis 14 Kohlenstoffatomen im Alkylrest, a2) mehr als 3 bis 30 Gewichts-%, vorzugsweise 5 bis 25 Gewichts-%, Monomeren mit mindestens zwei polymerisierbaren, olefinisch ungesättigten Doppelbindungen und a3) 15 bis 82 Gewichts-%, vorzugsweise 40 bis 70 Gewichts-%, weiteren polymerisierbaren Monomeren mit einer olefinisch ungesättigten Doppelbindung, wobei die Summe der Komponenten a1, a2 und a3 100 Gewichts-% beträgt.

19.09.1985

- 1A -

BASF Farben + Fasern Aktiengesellschaft, Hamburg

Curable Composition

The invention relates to a curable composition containing a soluble hydroxyl-containing acrylate copolymer, a compound possessing at least two cyclic carboxylic anhydride groups per molecule and a compound possessing at
5 least two epoxide groups per molecule.

Coating compositions which contain, as the essential binder, a hydroxyl-containing acrylate copolymer are well known. Aminoplast resins, for example alkylated
10 melamine/formaldehyde resins, are frequently used as cross-linking agents for these hydroxyl-containing acrylate copolymers. Compositions of this type cure at temperatures above 80°C. In many cases, a strong acid, for example p-toluenesulfonic acid, is employed as a catalyst in these
15 systems.

For many intended uses, however, it is advantageous if coating compositions cure at room temperature or slightly elevated temperatures, for example in automotive
refinishing.

EP-B-64 338 discloses a hydroxyl-containing acrylate copolymer which is cured with an aminoplast resin, using a special catalyst mixture. This coating composition can be cured at room temperature but has the disadvantage that the coatings based on this coating composition
20 have insufficient resistance to water and steam.
25



Systems which are based on epoxy/carboxyl cross-linking and cure at room temperature are also known. EP-A-123 793 describes compositions which cure at as low as room temperature, consist of polyepoxides and polymers
5 containing carboxyl groups and tertiary amino groups, and are obtainable by reacting vinyl polymers containing anhydride and carboxyl groups with compounds which contain at least one active hydrogen which is capable of reacting with the anhydrides and at least one tertiary amino group,
10 such as, for example, tertiary amino alcohols. The compositions described have the advantage that they cure at as low as room temperature and possess good resistance to gasoline, water and alkalis, and no undesirable discoloration attributable to tertiary amino compounds occurs.

15 EP-A-134 691 discloses curable compositions which contain a compound having at least two hydroxyl groups per molecule, a compound having at least two cyclic carboxylic anhydride groups per molecule and a compound having at least two epoxide groups per molecule. Both condensates and polymers are mentioned as suitable hydroxyl-containing polymers. According to this patent application, hydroxyl-containing acrylate copolymers are mixed with bis- and polycarboxylic anhydrides and bis- and polyepoxides, if appropriate with the concomitant use of a
20 catalyst, and the mixture is processed together with solvents and additives to give a coating composition which cures at as low as room temperature.

It is the object of the present invention to improve the properties of coatings based on epoxy/carboxyl



crosslinking from the point of view of resistance to chemicals, solvents, resistance to gasoline and resistance to water and steam and from the point of view of flexibility and corrosion resistance. It is also desirable for the coating compositions to cure, where appropriate, at room temperature so that they can be employed, for example, in automotive refinishing.

This object is achieved, surprisingly, if an acrylate copolymer having a high content of copolymerized polyethylenically unsaturated monomers is employed as the hydroxyl-containing acrylate copolymer. Low viscosities coupled with relatively high solids content can be achieved by means of the acrylate copolymers employed in the curable composition; furthermore, as a result of the highly branched structure of the copolymers, high reactivity of the hydroxyl groups toward the other functional groups of the curable compositions is achieved.

The invention relates to the curable composition of the type stated at the outset, wherein the hydroxyl-containing acrylate copolymer is obtainable from

- a1) from 10 to 60% by weight, preferably from 15 to 60% by weight, of hydroxyl-containing esters of acrylic acid and/or methacrylic acid, where the alkyl radical is of 2 to 14 carbon atoms,
- a2) more than 3 to 30% by weight, preferably from 5 to 25% by weight, of monomers having at least two polymerizable, olefinically unsaturated double bonds and
- a3) from 15 to 82% by weight, preferably from 40 to 70% by weight, of further polymerizable monomers having



an olefinically unsaturated double bond,
the sum of components a1), a2) and a3) being 100% by weight.

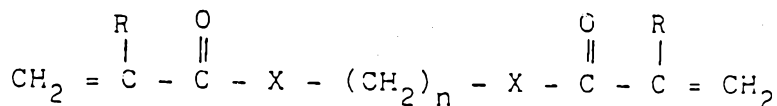
The choice of the further polymerizable monomers
of component a3 is not particularly critical. They may
5 be selected from a group consisting of styrene, vinyltol-
uene, acrylic acid, methacrylic acid, crotonic acid, ita-
conic acid, alkyl esters of acrylic and methacrylic acid,
alkoxyethyl acrylates and aryloxyethyl acrylates and the
corresponding methacrylates, and esters of maleic and
10 fumaric acid. Examples are methyl acrylate, ethyl acry-
late, propyl acrylate, butyl acrylate, isopropyl acrylate,
isobutyl acrylate, pentyl acrylate, isoamyl acrylate,
hexyl acrylate, 2-ethylhexyl acrylate, octyl acrylate,
3,5,5-trimethylhexyl acrylate, decyl acrylate, dodecyl
15 acrylate, hexadecyl acrylate, octadecyl acrylate, octa-
decenyl acrylate, pentyl methacrylate, isoamyl methacry-
late, hexyl methacrylate, 2-ethylbutyl methacrylate, octyl
methacrylate, 3,5,5-trimethylhexyl methacrylate, decyl
methacrylate, dodecyl methacrylate, hexadecyl methacrylate,
20 octadecyl methacrylate, butoxyethyl acrylate, butoxyethyl
methacrylate, methyl methacrylate, ethyl methacrylate,
propyl methacrylate, isopropyl methacrylate, butyl meth-
acrylate, cyclohexyl acrylate, cyclohexyl methacrylate,
acrylonitrile, methacrylonitrile, vinyl acetate, vinyl
25 chloride and phenoxyethyl acrylate. Other monomers can
be used provided that they do not result in the copolymer
having undesirable properties. The choice of component
a3 depends substantially on the desired properties of the
curable composition in respect of flexibility, hardness,



compatibility and polarity.

Among the compounds which can advantageously be used as component a3 are, from 0.1 to 20% by weight, preferably from 1 to 14% by weight, based on the total weight of all monomers, of tertiary amines having a polymerizable, olefinically unsaturated double bond, for example N,N'-dimethylaminoethyl methacrylate, N,N-diethylaminoethyl methacrylate, 2-vinylpyridine, 4-vinylpyridine, vinylpyrrolidine, vinylquinoline, vinylisoquinoline, N,N'-dimethylaminoethyl vinyl ether and 2-methyl-5-vinylpyridine. The advantage of these tertiary amino groups incorporated in the acrylate copolymer is that they catalyze subsequent carboxyl/epoxy crosslinking.

Compounds of the general formula



in which R denotes H or CH₃, X denotes O, NR', where R' is H or CH₃, or S, and n denotes 2 to 8, can advantageously be used as component a2.

Examples of such compounds are hexanediol diacrylate, hexanediol dimethacrylate, glycol diacrylate, glycol dimethacrylate, butanediol diacrylate, butanediol dimethacrylate, trimethylolpropane triacrylate, trimethylolpropane trimethacrylate and similar compounds. It is of course also possible to employ combinations of the polyunsaturated monomers. Divinylbenzene may furthermore be mentioned as a suitable component a2.

It should be noted that, when, inter alia, tertiary



amines possessing an olefinically unsaturated double bond are used as component a3, di- and polyesters of di- and polyols with acrylic acid are not used as component a2, since in this case gelling of the copolymer occurs.

5 Component a2 may advantageously furthermore be a reaction product of a carboxylic acid having a polymerizable, olefinically unsaturated double bond and glycidyl acrylate and/or glycidyl methacrylate or a polycarboxylic acid or monocarboxylic acid esterified with an unsaturated alcohol. If tertiary amino groups are incorporated in the
10 acrylate copolymer, reaction products of a carboxylic acid having an ethylenically unsaturated double bond, with the exception of acrylic acid, with glycidyl methacrylate, or with a polycarboxylic acid or unsaturated monocarboxylic acid esterified with an unsaturated alcohol, with the
15 exception of derivatives of acrylic acid, are suitable.

A reaction product of a polyisocyanate and an unsaturated alcohol or amine can also advantageously be used as component a2. The reaction product of one mole of
20 hexamethylene diisocyanate and two moles of allyl alcohol may be mentioned as an example of this.

Another advantageous component a2 is a diester of polyethylene glycol and/or polypropylene glycol having a mean molecular weight of less than 1500, preferably of
25 less than 1000, and acrylic acid and/or methacrylic acid. In this case, where monomers possessing tertiary amino groups are copolymerized, acrylic acid derivatives are not used as component a2. Di- or polyvinyl compounds of hydrocarbons, eg. divinylbenzene, may also be used as



component a2.

Particularly suitable components a1 are hydroxy-alkyl esters of acrylic acid and/or methacrylic acid possessing a primary hydroxyl group, for example hydroxyethyl
5 acrylate, hydroxypropyl acrylate, hydroxybutyl acrylate, hydroxyamyl acrylate, hydroxyhexyl acrylate, hydroxyoctyl acrylate and the corresponding methacrylates.

At least some of the components a1 may advantageously be a reaction product of one mole of hydroxyethyl
10 acrylate and/or hydroxyethyl methacrylate and on average 2 moles of ϵ -caprolactone.

Furthermore, component a1 can advantageously consist of up to 75% by weight, particularly preferably up to 50% by weight, based on the total amount of a1, of a
15 hydroxyl-containing ester of acrylic acid and/or methacrylic acid possessing a secondary hydroxyl group, in particular a reaction product of acrylic acid and/or methacrylic acid and the glycidyl ester of a carboxylic acid possessing a tertiary α -carbon atom.

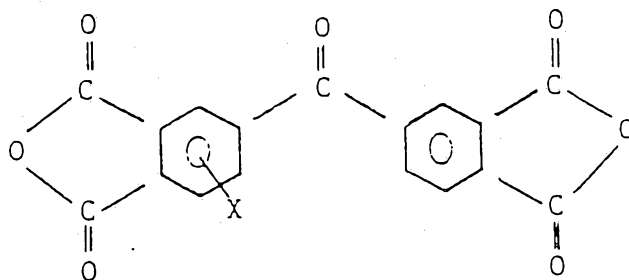
20 2-Hydroxypropyl acrylate, 2-hydroxybutyl acrylate, 3-hydroxybutyl acrylate and the corresponding methacrylates may be mentioned as examples.

Advantageously used compounds having at least two cyclic carboxylic anhydride groups per molecule are add-
25 ucts of trimellitic anhydride and a polyhydric alcohol. Examples of suitable polyhydric alcohols are ethylene glycol, propylene glycol, neopentyl glycol, hexane-1,6-diol, glycerol and trimethylolpropane.

Other suitable polyanhydrides are benzophenone



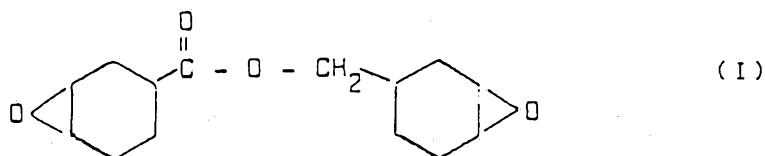
tetracarboxylic dianhydrides of the general formula

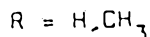
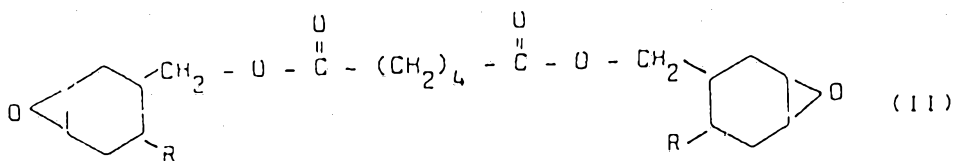


where X is H, halogen, NO₂-COOH or -SO₃H. 3,3',4,4'-Benzo-
phenonetetracarboxylic dianhydride, 2-bromo-3,3',4,4'-
5 benzophenonetetracarboxylic dianhydride and 5-nitro-3,3'-
4,4'-benzophenonetetracarboxylic dianhydride may be men-
tioned as examples. Other suitable compounds having at
least two cyclic carboxylic anhydride groups per molecule
are cyclopentanetetracarboxylic dianhydride, phenoxyphenyl-
10 tetracarboxylic dianhydride, the trianhydride of benzene-
hexacarboxylic acid and of cyclohexanehexacarboxylic acid,
and 1,2,3,4-butantetracarboxylic dianhydride. Preferably
used di- or polyanhydride compounds are copolymers of
maleic anhydride with ethylenically unsaturated compounds.
15 Styrene and vinyl esters of organic acids may be mentioned
as examples of the latter.

Examples of compounds having at least two epoxide
groups per molecule are condensates of epichlorohydrin
and bisphenol A.

20 Cycloaliphatic bisepoxides of the formulae (I) and
(II)





are particularly preferred.

Other preferred di- or polyepoxide compounds are polyglycidyl esters and/or polyglycidyl ethers, such as ethylene glycol diglycidyl ester, glycerol polyglycidyl ether, sorbitol polyglycidyl ether, trimethylolpropane polyglycidyl ether and pentaerythritol polyglycidyl ether. Epoxidized polybutadienes, epoxide-containing novolaks and low molecular weight acrylate resins having oxirane side groups, for example glycidyl methacrylate copolymers, are also suitable.

The invention furthermore relates to a process for the preparation of a coating composition based on the curable composition according to claims 1 - 21, wherein, in order to prepare the hydroxyl-containing acrylate copolymer

- a1) from 10 to 60% by weight, preferably from 15 to 60% by weight, of hydroxyl-containing esters of acrylic acid and/or methacrylic acid, where the alkyl radical is of two to 14 carbon atoms,
- a2) more than 3 to 30% by weight, preferably from 5 to 25% by weight, of monomers having at least two polymerizable, olefinically unsaturated double bonds and
- a3) from 15 to 80% by weight, preferably from 40 to



70% by weight, of further polymerizable monomers having an olefinically unsaturated double bond, the sum of components a1, a2 and a3 being 100% by weight, are copolymerized in an organic solvent at from 80 to 130°C, preferably from 90 to 120°C, using at least 0.5% by weight, preferably 2.5% by weight, based on the total weight of the monomers, of a polymerization regulator and using polymerization initiators, and the resulting acrylate solution, together with organic solvents, if appropriate pigments, fillers, conventional assistants, additives, the compound having at least two cyclic carboxylic anhydride groups per molecule, the compound having at least two epoxide groups per molecule and, if appropriate, a crosslinking catalyst, is processed to a coating composition by mixing and, if required, dispersing. During the preparation of the acrylate copolymer, care should be taken to ensure that a pre-crosslinked but non-gelled copolymer is obtained. By means of suitable polymerization conditions, it is possible, surprisingly, to prepare a clear, transparent, non-gelled solution of a branched copolymer. The use of monomers having at least two ethylenically unsaturated groups produces preliminary crosslinking of the copolymer molecule, which, because of the special reaction conditions according to the invention, nevertheless does not result in gelled products. These special reaction conditions comprise carrying out the polymerization at temperatures of 80 to 130°C, preferably 90 to 120°C. The polymerization is advantageously carried out so that a solution of the polymer having a solids content of from 40 to 65% by



weight results. The choice of initiator depends on the amount of bifunctional monomers employed. Where this amount is low, the initiators conventionally used for such temperatures, such as, for example, peroxy esters, can be used. For larger amounts of bifunctional monomer, initiators such as, for example, azo compounds are preferably employed. After the polymerization, the polymer solution is concentrated to the desired solids content, preferably to solids contents of 60% by weight, by distilling off the solvent. The clear copolymer solutions thus obtained possess a viscosity of from 0.4 to 10 dPa.s when adjusted to a solids content of 50% by weight.

Particularly suitable polymerization regulators are compounds containing mercapto groups, preferably mercaptoethanol.

If appropriate, catalysts for the epoxy/carboxyl crosslinking, for example tertiary amines, quaternary ammonium compounds, and specific chromium and tin compounds, can, if appropriate, be employed in the process according to the invention. Particularly preferably a tertiary amine, in an amount of from 0.5 to 10% by weight, based on the weight of the epoxide component, is used as the catalyst.

The use of an external catalyst is unnecessary in most cases in which acrylate copolymers already contain tertiary amino groups.

The curable compositions according to the invention can be mixed with pigments, solvents and additives shortly before use.



The curable coating agents according to the invention can be applied onto a substrate in the form of a film by spraying, flooding, dipping, roller coating, knife coating or painting, the film then being cured to give a
5 firmly adhering coating.

The coatings according to the invention possess improved properties in respect of resistance to gasoline and resistance to water and steam, in comparison with the coatings described in EP-A-134 691 (see Example 1). With
10 regard to resistance to solvents and to chemicals, too, the coatings according to the invention possess good properties.

The invention is illustrated in detail below with reference to examples:

15 A) Preparation of copolymers according to the invention (binder A)

In the examples below, all percentages are based on weight and all parts are parts by weight, unless stated otherwise. The solids contents were determined in a
20 through-circulation oven after 1 hour at 130°C. The viscosities were determined using a cone-and-plate viscometer.

Acrylate resin I

The following are initially taken in a 3 l stainless steel kettle and heated to 110°C:

25 Initially taken mixture: 386.5 parts of xylene
193.2 parts of 1-methoxyprop-2-yl acetate
310.0 parts of a glycidyl ester of a commercial α, α' -



dialkylalkanemonocarboxy-
lic acid having the empiri-
cal formula $C_{13}H_{24}O_3$
(Cardura E 10).

5 Feed 1 was metered in uniformly in the course of
3 hours.

Feed 1 91 parts of acrylic acid
 86 parts of hydroxyethyl methacrylate
 219 parts of styrene
10 150 parts of butanediol dimethacrylate
 144 parts of methyl methacrylate
 40 parts of mercaptoethanol

Feed 2 is metered in uniformly in the course of
3.5 hours. The two feeds are begun simultaneously.

15 Feed 2 32 parts of azobisisobutyronitrile
 328.2 parts of xylene
 164.1 parts of 1-methoxyprop-2-yl
 acetate

The temperature is kept at $110^{\circ}C$ during the polymeri-
20 zation, after which it is kept at $130^{\circ}C$ for 3.5 hours.
Thereafter, 380 parts of solvent mixture are distilled off
at $100^{\circ}C$ and under 180 mbar.

The acrylate resin solution thus obtained has a
solids content of 58.7%, a viscosity of 14.5 dPa.s and an
25 acid number of 14.5.

Acrylate resin II

The following are initially taken in a 3 l stain-
less steel kettle and heated to $110^{\circ}C$:

Initially taken mixture: 424.8 parts of xylene



212.4 parts of 1-methoxyprop-2-yl
acetate

5 310.0 parts of a glycidyl ester of
a commercial α, α' -dialkylal-
kanemonocarboxylic acid having
the empirical formula $C_{13}H_{24}O_3$
(Cardura E 10)

Feed 1 is metered in uniformly in the course of
3 hours.

10 Feed 1 91 parts of acrylic acid
161 parts of hydroxyethyl methacrylate
219 parts of styrene
150 parts of butanediol dimethacrylate
69 parts of methyl methacrylate
15 40 parts of mercaptoethanol

Feed 2 is metered in uniformly in the course of
3.5 hours. The two feeds are metered in simultaneously.

20 Feed 2 28 parts of azobisisobutyronitrile
287.2 parts of xylene
143.6 parts of 1-methoxyprop-2-yl
acetate

The temperature is kept at $110-112^{\circ}C$ during the
polymerization, after which the solution is kept at $130^{\circ}C$
for 4 hours. 428 parts of solvent mixture are distilled
25 off at $100^{\circ}C$ and under 180 mbar. The acrylate resin
solution thus obtained has a solids content of 61.4% (15
minutes, $180^{\circ}C$), a viscosity of 38.5 dPa.s and an acid
number of 13.6.



Preparation and testing of clear coats containing the acrylate resin solutions I - III

Coating formulation 1

2.90 parts of a bisanhydride obtained from trimellitic anhydride and 1,2-ethanediol
7.80 parts of methyl ethyl ketone and
3.52 parts of 3,4-epoxycyclohexylmethyl 3,4-epoxycyclohexanecarboxylate
are mixed.

10 10.00 parts of the acrylate resin from Example I
and

0.28 part of a metal salt catalyst solution
(accelerator Cordova AMC-2)
are added to this solution.

15 Coating formulation 2

2.9 parts of a bisanhydride obtained from 2 moles of trimellitic anhydride and 1 mole of 1,2-ethanediol,
7.10 parts of methyl ethyl ketone and
3.52 parts of bis-(3,4-epoxycyclohexyl) adipate
20 are mixed with one another.

10.00 parts of the acrylate resin from Example II
and

0.40 part of a metal salt catalyst solution (accelerator Cordova AMC-2)
25 are added to this solution.

Coating formulation 3

2.90 parts of a bisanhydride obtained from 2 moles of trimellitic anhydride and 1 mole of 1,2-ethanediol,
7.20 parts of methyl ethyl ketone and



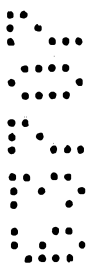
3.52 parts of 3,4-epoxycyclohexyl 3,4-epoxycyclohexanecarboxylate

are mixed with one another.

5 11.5 parts of the acrylate resin from Example III are added to this solution.

Immediately after mixing, 200 μ m films of the coatings are applied onto glass panels, and, after drying for 16 hours at room temperature, the films are tested with regard to hardness, resistance to gasoline and resistance to water.

The pendulum hardness is determined by the König method, in the gasoline test a felt pad impregnated with FAM mineral spirit is covered and left on the film for 1 hour, and in the water test a large drop of water (diameter 5 cm) is left on the film for 2 hours.



15

Coating film from Example	Pendulum hardness (König)	Resistance to gasoline	Resistance to water
1	63''	no softening no marking	no softening no marking
2	45''	no softening no marking	no softening no marking
3	91''	no softening no marking	no softening no marking



20



THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A curable composition containing a soluble hydroxyl-containing acrylate copolymer, a compound possessing at least two cyclic carboxylic anhydride groups per molecule and a compound possessing at least two epoxide groups per molecule, wherein the hydroxyl-containing acrylate copolymer is obtained from

- a1) from 10 to 60% by weight, of hydroxyl-containing esters of acrylic acid and/or methacrylic acid, where the alkyl radical is of two to 14 carbon atoms,
- a2) more than 3 to 30% by weight, of monomers having at least two polymerizable, olefinically unsaturated double bonds and
- a3) from 15 to 82% by weight, of further polymerizable monomers having an olefinically unsaturated double bond, the sum of components a1, a2 and a3 being 100% by weight.

2. A curable composition as claimed in claim 1 in which the amount of component a1 is from 15 to 60% by weight.

3. A curable composition as claimed in claim 1 or 2 in which the amount of component a2 is from 5 to 25% by weight.

4. A curable composition as claimed in any preceding claim in which the amount of component a3 is from 40 to 70% by weight.

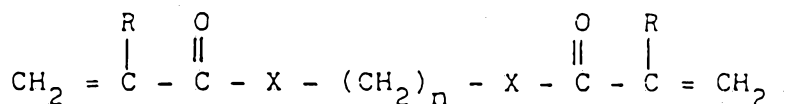
5. A curable composition as claimed in any preceding claim wherein the further polymerizable monomers of component a3 are one or more of styrene, vinyltoluene, acrylic acid, methacrylic acid, crotonic acid, itaconic acid, alkylesters of acrylic and methacrylic acid, alkoxylethyl acrylates and aryloxyethyl acrylates and the corresponding methacrylates, or esters of maleic and fumaric acid.



6. A curable composition as claimed in any preceding claim wherein, from 0.1 to 20% by weight, based on the total weight of all monomers, of tertiary amines having a polymerizable, olefinically unsaturated double bond are used as component a3.

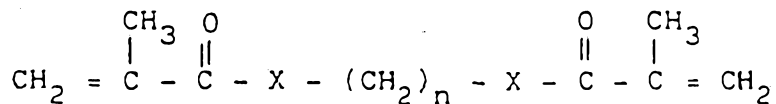
7. A curable composition, as claimed in claim 6 in which the amount of tertiary ^{amines} ~~A means~~ having a polymerizable, olefinically unsaturated double bond used as component a3 is from 1 to 14% by weight.

8. A curable composition as claimed in any preceding claim wherein component a2 is of the formula



in which R denotes H or CH₃, X denotes O, S, or NR¹, where R¹ is H, alkyl or aryl, and n denotes 2 to 8.

9. A curable composition as claimed in any one of claims 1 to 7 wherein component a2 is of the formula



where X denotes O, S or NR, where R is H, alkyl or aryl, and n denotes 2 to 8.



10. A curable composition as claimed in any one of claims 1 to 7 wherein component a2 is a reaction product of a carboxylic acid having a polymerizable, olefinically unsaturated double bond and glycidyl acrylate and/or glycidyl methacrylate.

11. A curable composition as claimed in any one of claims 1 to 7 wherein component a2 is a reaction product of a carboxylic acid having a polymerizable, olefinically unsaturated double bond, with the exception of acrylic acid, and glycidyl methacrylate.

12. A curable composition as claimed in any one of claims 1 to 7 wherein component a2 is a polycarboxylic acid or unsaturated monocarboxylic acid which is esterified with an unsaturated alcohol having a polymerizable double bond.

13. A curable composition as claimed in any one of claims 1 to 7 wherein component a2 is a polycarboxylic acid or unsaturated monocarboxylic acid which is esterified with an unsaturated alcohol having a polymerizable double bond, with the exception of derivatives of acrylic acid.

14. A curable composition as claimed in any one of claims 1 to 7 wherein component a2 can be prepared by reacting a polyisocyanate with amines or alcohols having an unsaturated, polymerizable double bond.

15. A curable composition as claimed in any one of claims 1 to 7, wherein component a2 is a diester of polyethylene glycol and/or polypropylene glycol, having a mean molecular weight of less than 1500, and acrylic acid and/or methacrylic acid.

16. A curable composition as claimed in claim 15 wherein a2 is a diester of polyethylene glycol and/or polypropylene glycol having a mean molecular weight of less than 1000.



17. A curable composition as claimed in any one of claims 1 to 7 wherein component a2 is a diester of polyethylene glycol and/or polypropylene glycol, having a mean molecular weight of less than 1500, and methacrylic acid.

18. A curable composition as claimed in claim 17 wherein a component a2 is a diester of polyethylene glycol and/or polypropylene glycol, having a mean molecular weight of less than 1000.

19. A curable composition as claimed in any one of claims 1 to 18 wherein component a1 is a hydroxyalkyl ester of acrylic acid and/or methacrylic acid possessing a primary hydroxyl group.

20. A curable composition as claimed in any one of claims 1 to 19, wherein component a1 at least partially comprises a reaction product of one mole of a hydroxyethyl acrylate and/or hydroxyethyl methacrylate and on average two moles of ϵ -caprolactone.

21. A curable composition as claimed in any one of claims 1 to 20, wherein component a1 consists of up to 75% by weight, based on the total amount of a1, of a hydroxyl-containing ester of acrylic acid and/or methacrylic acid possessing a secondary hydroxyl group.

22. A curable composition as claimed in claim 21 wherein component a1 consists of up to 50% by weight based on the total weight of a1, of a hydroxyl containing ester of acrylic acid and/or methacrylic acid and/or methacrylic acid having a secondary hydroxyl group.

23. A curable composition as claimed in claim 21 or 22 wherein the hydroxyl-containing ester is a reaction product of acrylic acid and/or methacrylic acid with the



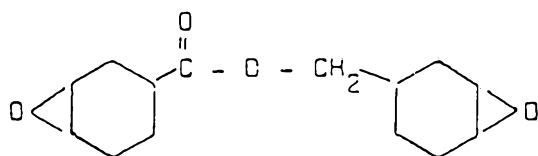
glycidyl ester of a carboxylic acid having a tertiary α -carbon atom.

24. A curable composition as claimed in any one of claims 1 to 23, wherein the compound having at least two cyclic carboxylic anhydride groups per molecule is an adduct of trimellitic anhydride and a polyhydric alcohol.

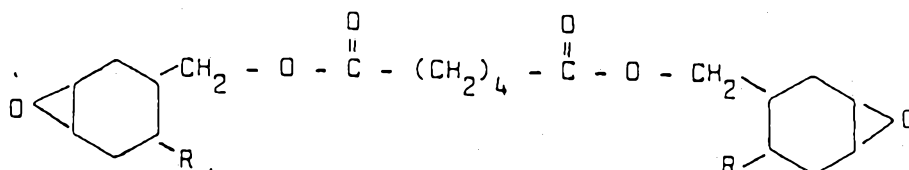
25. A curable composition as claimed in claim 24, wherein the compound having at least two cyclic carboxylic anhydride groups per molecule is an adduct of trimellitic anhydride and ethyleneglycol, propyleneglycol, neopentylglycol, hexane-1,6-diol, glycerol or trimethylolpropane.

26. A curable composition as claimed in any one of claims 1 to 25, wherein the compound having at least 2 cyclic carboxylic anhydride groups is a copolymer of maleic anhydride with ethylenically unsaturated monomers.

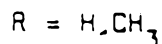
27. A curable composition as claimed in any one of claims 1 to 26, wherein the compound having at least two epoxide groups per molecule is a bisepoxide of the formula (I) or (II)



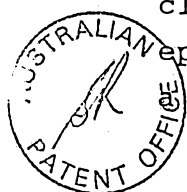
(I)



(II)



28. A curable composition as claimed in any one of claims 1 to 27, wherein the compound having at least 2 epoxide groups per molecule is a polyglycidyl ether and/or polyglycidyl ester.



29. A process for the preparation of a coating composition based on the curable composition according to any one of claims 1 to 28, wherein, in order to prepare the hydroxyl-containing acrylate copolymer

a1) from 10 to 60% by weight, of hydroxyl-containing esters of acrylic acid and/or methacrylic acid, where the alkyl radical is of 2 to 14 carbon atoms,

a2) more than 3 to 30% by weight, of monomers possessing at least two polymerizable, olefinically unsaturated double bonds and

a3) from 15 to 82% by weight, of further polymerizable monomers having an olefinically unsaturated double bond, the sum of components

a1, a2 and a3 being 100% by weight, are copolymerized in an organic solvent at from 80 to 130°C, using at least 0.5% by weight, based on the total weight of the monomers, of a polymerization regulator and using polymerization initiators, and the resulting acrylate solution, together with organic solvents, if appropriate pigments, fillers, conventional assistants, additives, the compound having at least two cyclic carboxylic anhydride groups per molecule, the compound having at least two epoxide groups per molecule and, if appropriate, a crosslinking catalyst, is processed to a coating composition by mixing and, if required, dispersing.

30. A process according to claim 29 in which the amount of a1) is from 15 to 60% by weight.

31. A process according to claim 29 or 30 in which the amount of a2) is from 5 to 25% by weight.

32. A process according to any one of claims 29 to 31 in which the amount of a3) is from 40 to 70% by weight.

33. A process according to any one of claims 29 to 32 in which the components are co-polymerized in an organic solvent at from 90 to 120°C.



34. A process according to any one of claims 29 to 33 in which there is 2.5% by weight, less than the total weight of the monomers of a polymerization regulator.

35. A process as claimed in claim 34, wherein the polymerization regulators used are compounds containing mercapto groups, preferably mercaptoethanol.

36. A process as claimed in any one of claims 29 to 35, wherein the polymerization initiators used are azo compounds and/or peroxy esters.

37. A process as claimed in any one of claims 29 to 36, wherein the polymerization for the preparation of the hydroxyl-containing acrylate copolymer is carried out in such a way that a solution of the polymer having a solids content of 40 to 65% by weight results.

38. A process as claimed in any one of claims 29 to 37, wherein the crosslinking catalyst is a tertiary amine and is used in an amount of from 0.5 to 10% by weight, based on the weight of the epoxide component.

39. A curable composition substantially as hereinbefore described with reference to any one of the foregoing examples.

40. A process for the preparation of a coating composition based on a curable composition substantially as hereinbefore described with reference to any one of the foregoing examples.

Dated this 31st day of August, 1989.

BASE LACKE & FARBEN AKTIENGESELLSCHAFT
By Their Patent Attorneys:

GRIFFITH HACK & CO.
Fellows Institute of Patent
Attorneys of Australia.



INTERNATIONAL SEARCH REPORT

International Application No PCT/EP 86/00522

I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) *				
According to International Patent Classification (IPC) or to both National Classification and IPC C 08 G 59/62; C 08 G 59/42; C 08 L 63/00; C 09 D 3/32 Int. Cl. ⁴ C 09 D 3/81; C 08 L 33/06				
II. FIELDS SEARCHED				
Minimum Documentation Searched ⁷				
Classification System	Classification Symbols			
Int. Cl. ⁴	C 08 G; C 08 L, C 09 D			
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched ⁸				
III. DOCUMENTS CONSIDERED TO BE RELEVANT ⁹				
Category ⁹	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³		
A	EP, A, 0134691 (I.C.I.) 20 March 1985 cited in the application			
			
A	DE, A, 2630011 (SCHERING) 12 January 1978			
			
A	Chemical Abstracts, volume 97, Nr 24, 13 December 1982, Columbus, Ohio (US) see page 36, abstract 199044y & JP, A, 8284843 (MATSUSHITA ELECTRIC WORKS LTD.) 27 May 1982			
			
A	FR, A, 2432542 (TORAY INDUSTRIES, INC.) 29 February 1980			
			
<table style="width: 100%; border: none;"> <tr> <td style="width: 50%; vertical-align: top;"> ¹⁰ Special categories of cited documents: <ul style="list-style-type: none"> "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed </td> <td style="width: 50%; vertical-align: top;"> <ul style="list-style-type: none"> "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art. "Δ" document member of the same patent family </td> </tr> </table>			¹⁰ Special categories of cited documents: <ul style="list-style-type: none"> "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed 	<ul style="list-style-type: none"> "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art. "Δ" document member of the same patent family
¹⁰ Special categories of cited documents: <ul style="list-style-type: none"> "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed 	<ul style="list-style-type: none"> "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art. "Δ" document member of the same patent family 			
IV. CERTIFICATION				
Date of the Actual Completion of the International Search		Date of Mailing of this International Search Report		
12 December 1986 (12.12.86)		28 January 1987 (28.01.87)		
International Searching Authority		Signature of Authorized Officer		
EUROPEAN PATENT OFFICE				

ANNEX TO THE INTERNATIONAL SEARCH REPORT ON

INTERNATIONAL APPLICATION NO. PCT/EP 86/00522 (SA 14475)

This Annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on 13/01/87

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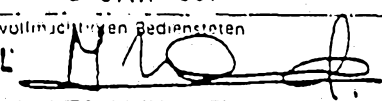
Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A- 0134691	20/03/85	AU-A- 3193984	21/02/85
		GB-A- 2148900	05/06/85
		JP-A- 60106830	12/06/85
DE-A- 2630011	12/01/78	None	
FR-A- 2432542	29/02/80	JP-A- 54162729	24/12/79
		US-A- 4294939	13/10/81
		GB-A, B 2029422	19/03/80
		DE-A- 2833647	21/02/80
		NL-A- 7808088	05/02/80

For more details about this annex :
see Official Journal of the European Patent Office, No. 12/82

INTERNATIONALER RECHERCHENBERICHT

Internationales Aktenzeichen **PCT/EP 86/00522**

I. KLASSE		
I. KLASSE DES ANMELDUNGSGEGENSTANDS (bei mehreren Klassifikationssymbolen sind alle anzugeben) ⁶		
Nach der internationalen Patentklassifikation (IPC) oder nach der nationalen Klassifikation und der IPC		
Int. Cl. ⁴ C 08 G 59/62; C 08 G 59/42; C 08 L 63/00; C 09 D 3/38; C 09 D 3/81; C 08 L 33/06		
II. RECHERCHIERTE SACHGEBIETE		
Recherchierter Mindestprüfstoff ⁷		
Klassifikationssystem	Klassifikationssymbole	
Int. Cl. ⁴	C 08 G; C 08 L; C 09 D	
Recherchierte nicht zum Mindestprüfstoff gehorende Veröffentlichungen, soweit diese unter die recherchierten Sachgebiete fallen ⁵		
III. EINSCHLÄGIGE VERÖFFENTLICHUNGEN⁹		
Art [*]	Kennzeichnung der Veröffentlichung ¹¹ , soweit erforderlich unter Angabe der maßgeblichen Teile ¹²	Betr. Ansoruch Nr. ¹³
A	EP, A, 0134691 (I.C.I.) 20. März 1985 (In der Anmeldung erwähnt)	
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A	DE, A, 2630011 (SCHERING) 12. Januar 1978	
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A	Chemical Abstracts, Band 97, Nr. 24, 13. Dezember 1982, Columbus, Ohio (US) siehe Seite 36, Zusammenfassung 199044y & JP, A, 8284843 (MATSUSHITA ELECTRIC WORKS LTD.) 27. Mai 1982	
	--	
A	FR, A, 2432542 (TORAY INDUSTRIES, INC.) 29. Februar 1980	

<p>[*] Besondere Kategorien von angegebenen Veröffentlichungen¹⁰</p> <p>"A" Veröffentlichung, die den allgemeinen Stand der Technik definiert, aber nicht als besonders bedeutsam anzusehen ist</p> <p>"E" älteres Dokument, das jedoch erst am oder nach dem internationalen Anmeldedatum veröffentlicht worden ist</p> <p>"L" Veröffentlichung, die geeignet ist, einen Prioritätsanspruch zweifelhaft erscheinen zu lassen oder durch die das Veröffentlichungsdatum einer anderen im Recherchenbericht genannten Veröffentlichung belegt werden soll oder die aus einem anderen besonderen Grund angegeben ist (wie ausgeführt)</p> <p>"O" Veröffentlichung, die sich auf eine mündliche Offenbarung, eine Benutzung, eine Ausstellung oder andere Maßnahmen bezieht</p> <p>"P" Veröffentlichung, die vor dem internationalen Anmeldedatum, aber nach dem beanspruchten Prioritätsdatum veröffentlicht worden ist</p> <p>"T" Spätere Veröffentlichung, die nach dem internationalen Anmeldedatum oder dem Prioritätsdatum veröffentlicht worden ist und mit der Anmeldung nicht kollidiert, sondern nur zum Verständnis des der Erfindung zugrundeliegenden Prinzips oder der ihr zugrundeliegenden Theorie angeeignet ist</p> <p>"X" Veröffentlichung von besonderer Bedeutung; die beanspruchte Erfindung kann nicht als neu oder auf erfinderischer Tätigkeit beruhend betrachtet werden</p> <p>"Y" Veröffentlichung von besonderer Bedeutung; die beanspruchte Erfindung kann nicht als auf erfinderischer Tätigkeit beruhend betrachtet werden, wenn die Veröffentlichung mit einer oder mehreren anderen Veröffentlichungen dieser Kategorie in Verbindung gebracht wird und diese Verbindung für einen Fachmann naheliegend ist</p> <p>"Z" Veröffentlichung, die Mitglied derselben Patentfamilie ist</p>		
IV. BESCHEINIGUNG		
Datum des Abschlusses der internationalen Recherche		Absenddatum des internationalen Recherchenberichts
12. Dezember 1986		28 JAN 1987
Internationale Recherchenbehörde		Unterschrift des bevollmächtigten Bediensteten
Europäisches Patentamt		M. VAN MOL 

ANHANG ZUM INTERNATIONALEN RECHERCHENBERICHT ÜBER DIE

INTERNATIONALE PATENTANMELDUNG NR. PCT/EP 86/00522 (SA 14475)

In diesem Anhang sind die Mitglieder der Patentfamilien der im obengenannten internationalen Recherchenbericht angeführten Patentdokumente angegeben. Die Angaben über die Familienmitglieder entsprechen dem Stand der Datei des Europäischen Patentamts am 13/01/87

Diese Angaben dienen nur zur Unterrichtung und erfolgen ohne Gewähr.

Im Recherchenbericht angeführtes Patentdokument	Datum der Veröffentlichung	Mitglied(er) der Patentfamilie	Datum der Veröffentlichung
EP-A- 0134691	20/03/85	AU-A- 3193984	21/02/85
		GB-A- 2148900	05/06/85
		JP-A- 60106830	12/06/85
DE-A- 2630011	12/01/78	Keine	
FR-A- 2432542	29/02/80	JP-A- 54162729	24/12/79
		US-A- 4294939	13/10/81
		GB-A,B 2029422	19/03/80
		DE-A- 2833647	21/02/80
		NL-A- 7808088	05/02/80

Für nähere Einzelheiten zu diesem Anhang :
siehe Amtsblatt des Europäischen Patentamts, Nr. 12/82