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C62X C628 C65X C658 C80Y C813 C815
U1S S1346 S1409

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DE 001900346 A FR 001527054 A FR 001511011 A
JP 630275545 A US 3536687 A
Tetrahedryn letters 31 (51) 7457-60 (1990)
Chemical abstracts 101 (15):130271d

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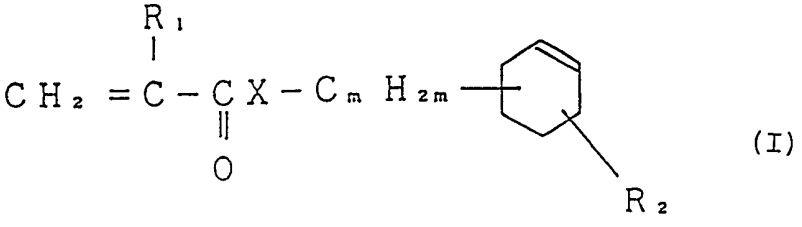
UK CL (Edition K) C2C CBZ CKG
INT CL[5] C07C
Online database: CAS ONLINE

(54) **Cyclohexenyl compounds and photocurable resin compositions prepared therefrom**

(57) Photoreactive compounds of the formula:



(I)

(wherein R_1 is H, CH_3 or C_2H_5; X is -O- or -NH-; R_2 is H, halogen or a C_1-C_4 alkyl or alkoxy group, and m is 1 to 5) may be used in the preparation of *photosensitive composition comprising a polymer of the said compound together with an azide compound* (as photosensitivity-imparting agent).

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FIG. 1

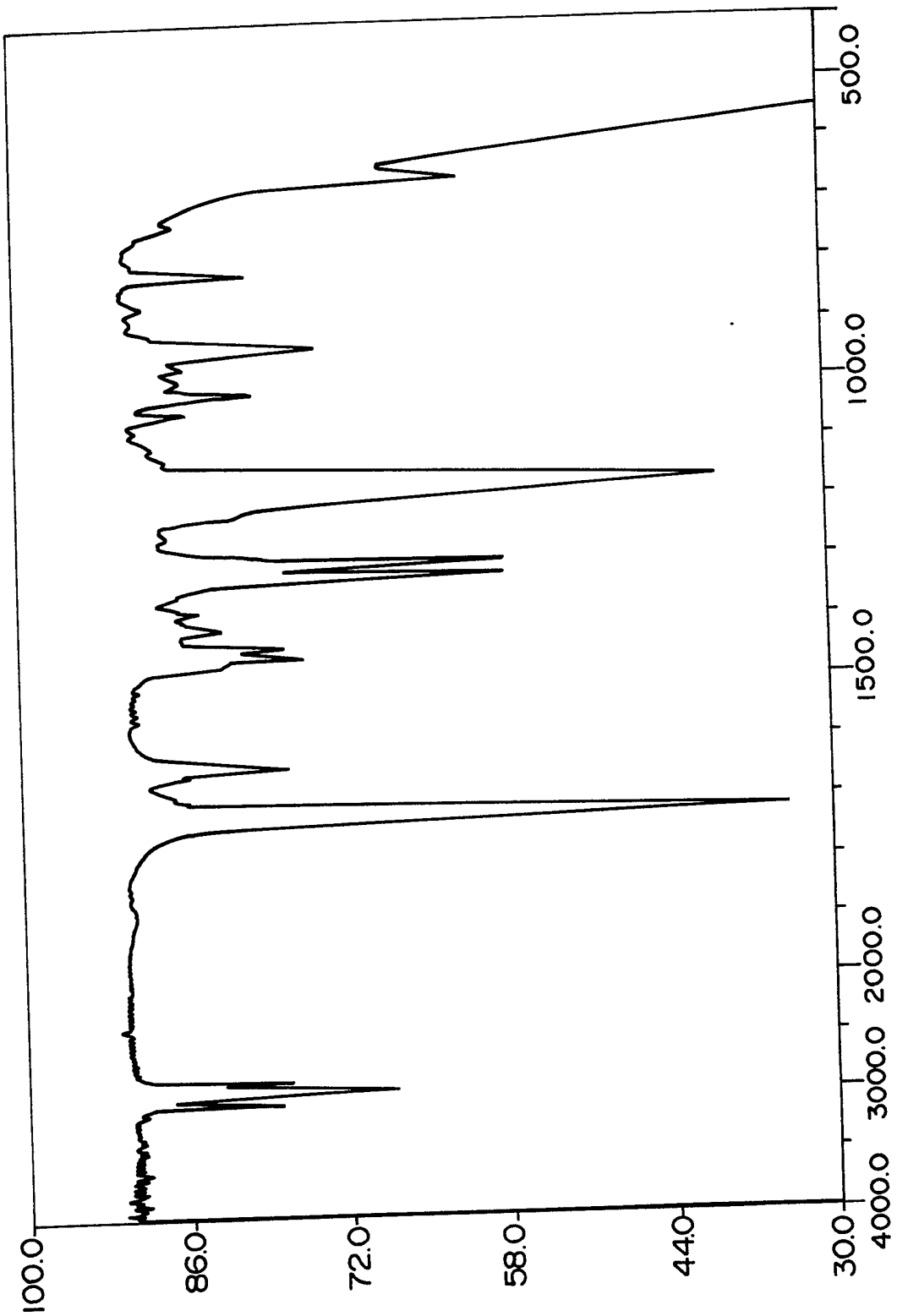


FIG. 2

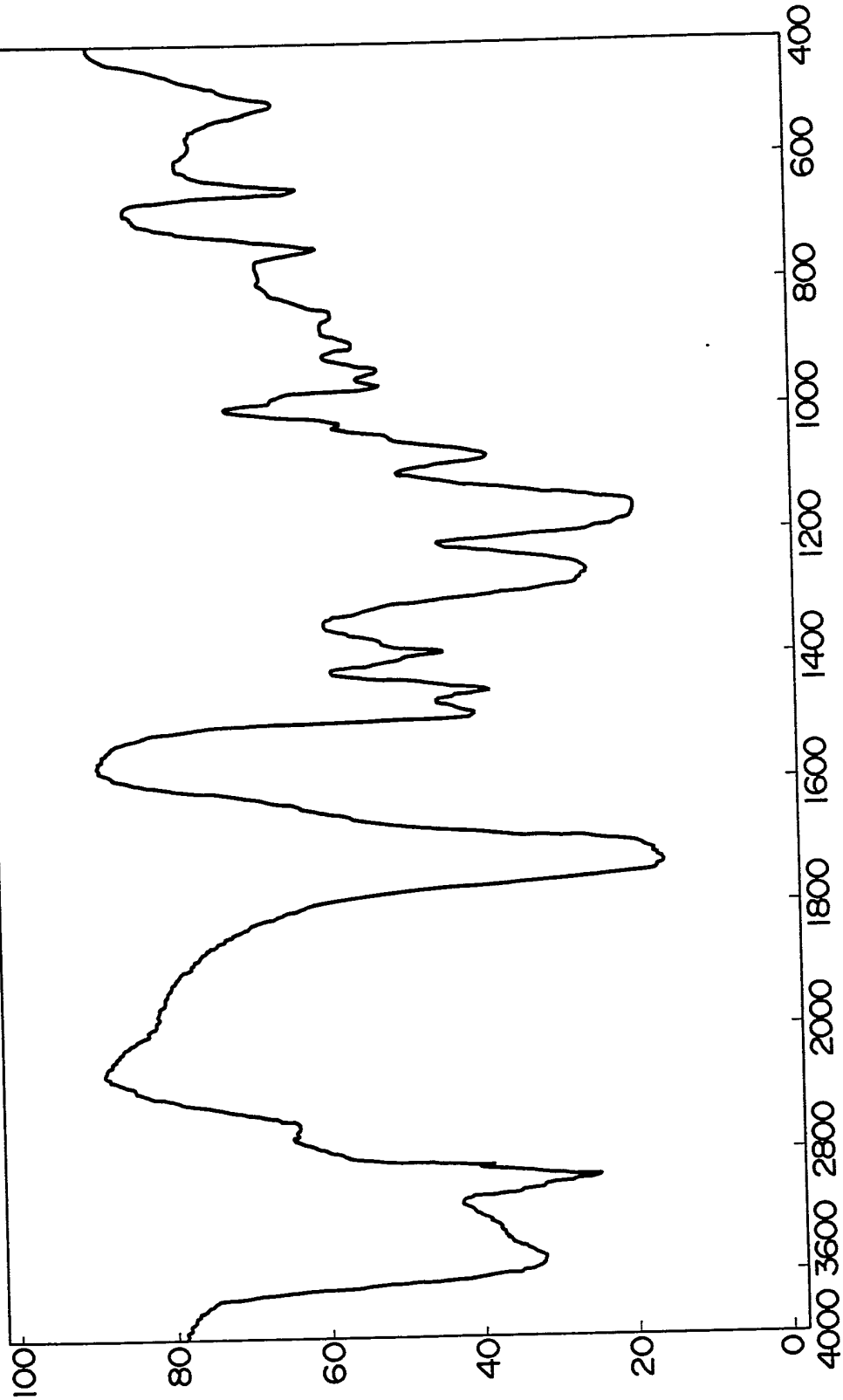

100
80
60
40
20
0
4000
3600
2800
2000
1800
1600
1400
1200
1000
800
600
400

FIG. 3

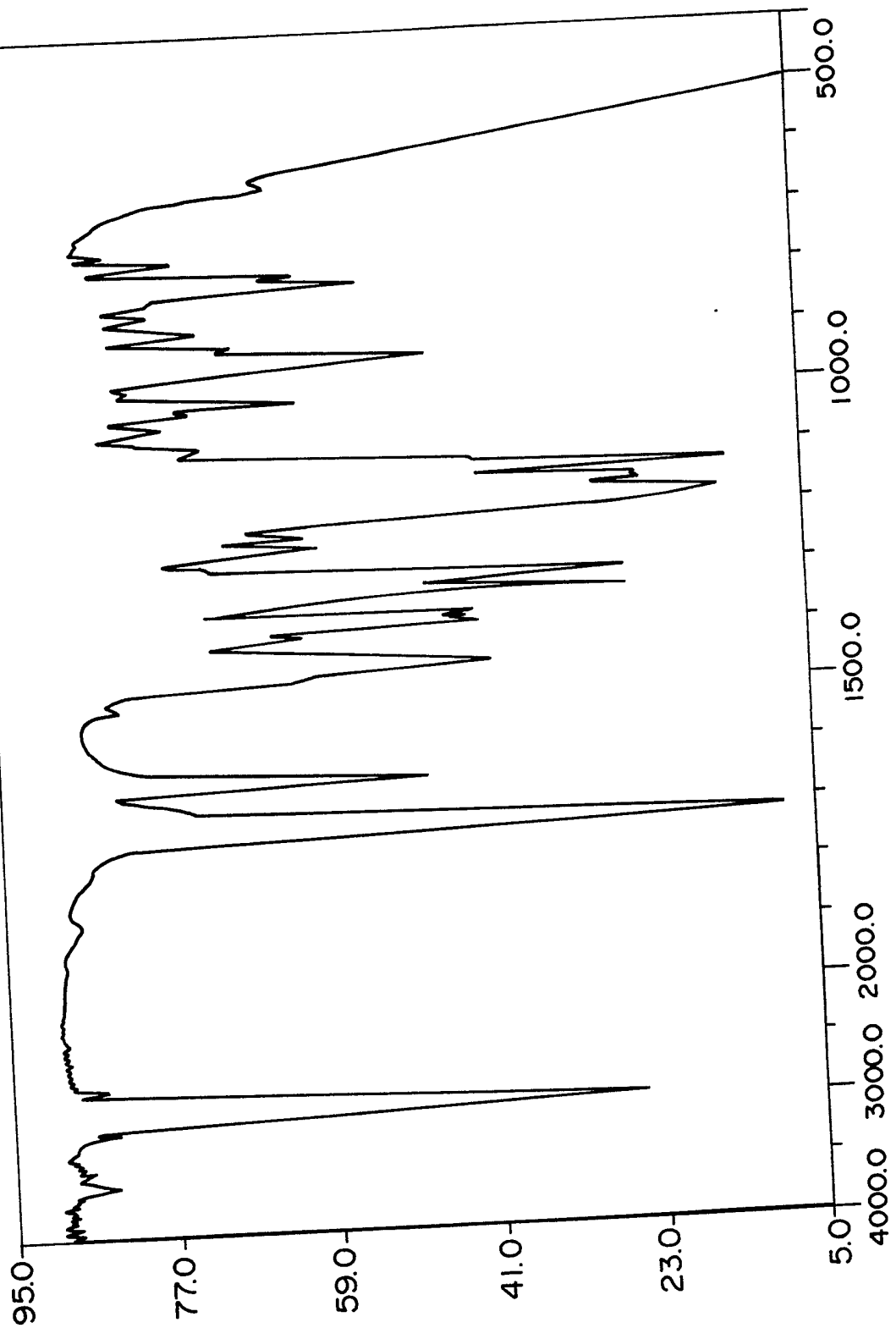
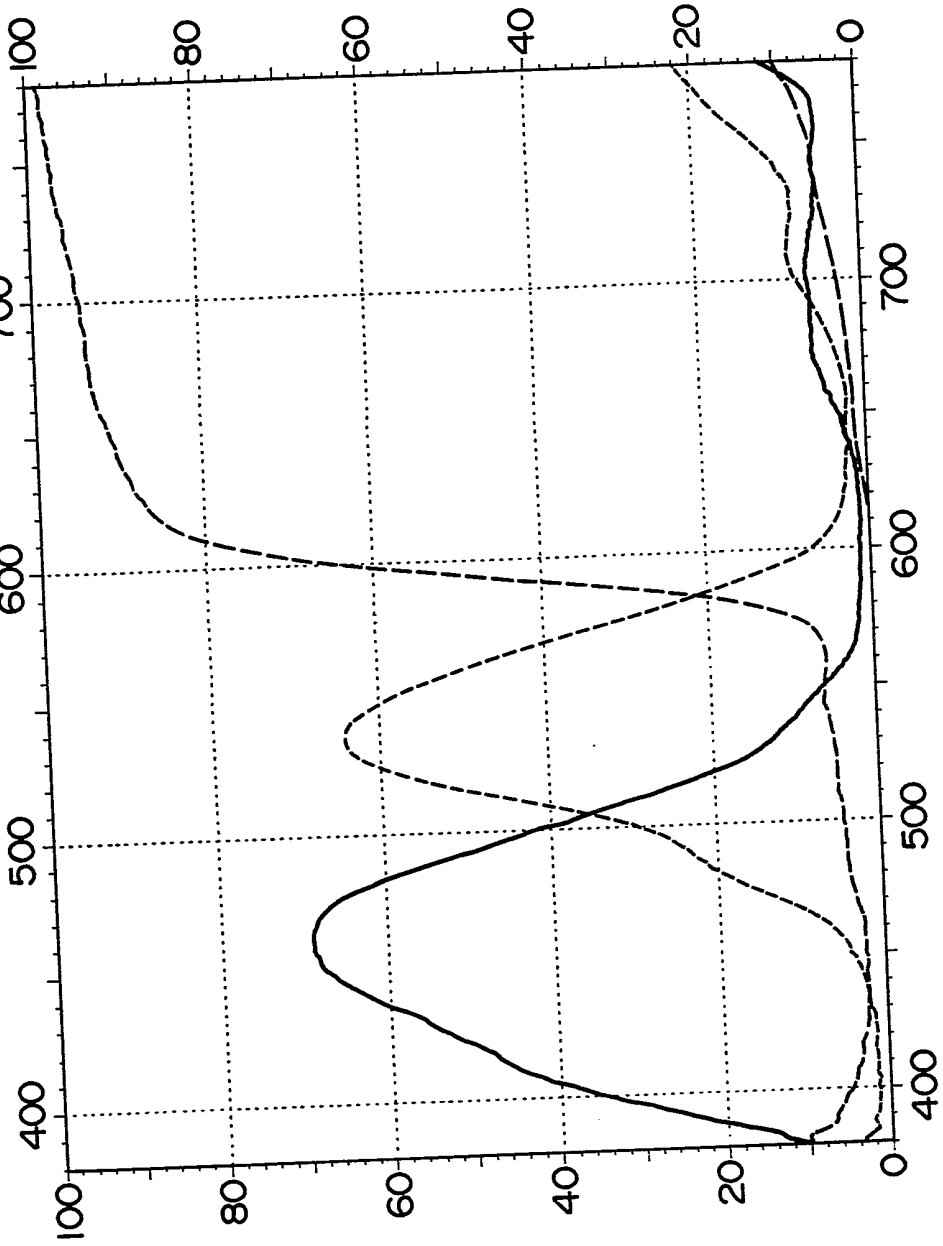


FIG. 4



Photoreactive compounds and photocurable resin compositions prepared therefrom

This invention relates to photoreactive compounds and to photocurable resin compositions prepared therefrom. More particularly the invention relates to a photocurable resin composition for forming a relief printed image. The photocurable resin composition of the invention may be used for part-processing in the electronics field and in the application field of general photoresists such as plate-making materials in the printing industry. Further, since the photocurable resin composition of the present invention is highly sensitive to ultraviolet radiation, it is very suitable for producing colour filters of the pigment dispersion type.

In recent years, for photocurable resin composition for forming printed images, a high curing speed has been required for the reasons of productivity, energy-saving, etc., and further, a high solubility of unexposed part, in an alkaline aqueous solution has been required for the reasons of prevention of polution of operational environments, economy, etc.For such purposes, a number of

photocurable compositions have so far been proposed.

These compositions include a composition obtained by dimerization and crosslinking at photosensitive group by light (Japanese patent publication Nos. Sho 49-46396, Sho 50-15026, etc.), and a photopolymerizable composition consisting of a compound containing an addition-polymerizable ethylenic double bond and a photopolymerization initiator and if desired, an organic high molecular weight binder and a sensitizer for broadening the photosensitive wave range.

As examples of photopolymerizable compositions, a number of the compositions have been known, such as a composition having an addition-polymerizable compound containing an ethylenic unsaturated double bond to be subjected to photopolymerization with a system consisting of a substituted triazine and a (p-dialkylamino-benzylidene)ketone (Japanese patent application laid-open No. Sho 63-139339), a composition to be photopolymerized with a system consisting of keto-coumarin and an activator (Japanese patent application laid-open Nos. Sho 52-112681, 58-15503), a composition to be photopolymerized with an organic peroxide and a pigment (Japanese patent application laid-open No. Sho 59-140203), etc.

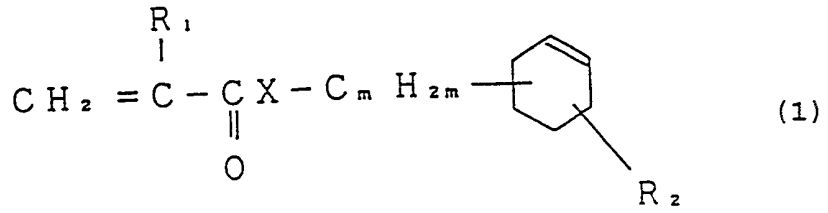
These photocurable resin compositions are coated onto a base to prepare a photosensitive layer, followed by exposing this layer to an image, polymerizing and

curing the resulting exposed part and dissolving and removing the resulting uncured part with a solvent capable of dissolving only the part to form a cured printed image.

However, these photocurable resin composition have drawbacks that the unexposed part cannot be dissolved with an aqueous solvent; the curing speed in the presence of oxygen is low due to their photopolymerization system; when the pretreatment temperature prior to their exposure varies, the coating thickness after development varies to a large extent; etc. Thus, they could not have been regarded as fully satisfying properties necessary for forming printed image.

In order to overcome these drawbacks, the present inventors have made extensive research, and as a result, have found that a combination of a high molecular weight compound containing an alicyclic compound having an unsaturated bond in its ring, with an azide has an extremely high sensitivity, and as a result, have achieved the present invention. Further, we have made extensive research in a photocurable resin composition capable of dissolving and removing the unexposed part with an alkali aqueous solution and having a high curing speed, and as a result, have achieved the present invention.

Basically, the present invention provides a photoreactive compound of the formula:



(1)

(wherein R_1 is a hydrogen atom or a methyl or ethyl group; X is -O- or -NH-; R_2 is a hydrogen or halogen atom or a C_1-C_4 alkyl or C_1-C_4 alkoxy group and m is an integer of from 1 to 5).

The invention also provides a photocurable resin composition obtained by adding an azide compound, as a photosensitivity-imparting agent, to a high molecular weight compound obtained by polymerizing a photoreactive compound of formula (1). In such photocurable resin compositions the polymer is preferably one obtained by copolymerizing the photoreactive compound with a compound containing an ethylenic unsaturated double bond and a carboxylic group in the same molecule. Such a composition forms the basis for a colour filter material having a colouring pigment dispersed in the photocurable resin composition.

BRIEF DESCRIPTION OF THE ACCOMPANYING DRAWINGS

Fig. 1 shows an IR spectra of a compound obtained in Example 1.

Fig. 2 shows an IR spectra of a high molecular weight compound obtained in Example 1.

Fig. 3 shows an IR spectra of a compound obtained in Example 3.

Fig. 4 shows a spectra of a colored printed image formed in Example 7.

DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

The high molecular weight compound in the composition of the present invention is characterized by having a compound expressed by the above-mentioned formula in the molecular structure thereof, and includes a homopolymer consisting only of the above structural unit and a copolymer having repetition units having the above structural unit combined with at least one structural unit expressed by a structure obtained by subjecting the unsaturated double bond of other vinyl monomer to scission.

The photoreactive compound of the present invention is a novel compound and may be easily obtained by reacting a cyclohexenylalkanol or a cyclohexenylalkylamine with a (meth)acrylic chloride.

The photoreactive compound of the present invention is expressed by the above formula and is characterized by containing two unsaturated double bonds having different reactivities in the molecular structure

thereof. The unsaturated double bond in the cyclohexene ring is abundant in the photoreactivity with azides, but it is low in the reactivity in the case of radical polymerization reaction. Whereas, the other unsaturated double bond is abundant in the radical polymerizability, but it is low in the photoreactivity with azides. When the compound is mixed with a photo-inactive, high molecular weight compound, followed by adding a radical initiator or an azide compound, it is, of course, possible to prepare a photocurable resin, but it is more preferred that making use of the above-mentioned difference between the reactivities of the two unsaturated double bonds, the compound is firstly subjected to radical polymerization to form a high molecular weight compound, followed by utilizing the photoreactivity of the double bond of the cyclohexene ring with an azide compound, in the resulting high molecular weight compound.

The high molecular weight compound prepared using the compound may be either of the homopolymer consisting only of the compound or a copolymer of the compound with other vinyl monomer.

Examples of the vinyl monomers to be copolymerized with the compound of the present invention are styrenes such as styrene, methylstyrene, etc., acrylic acid, methacrylic acid, esters of α-methylene aliphatic monocarboxylic acids such as methyl (meth)acrylate,

ethyl (meth)acrylate, n-butyl (meth)acrylate, isobutyl (meth)acrylate, etc., vinyl esters such as vinyl acetate, etc. However, in order to give a suitable solubility of the high molecular weight compound in an alkali aqueous solution, it is preferred to introduce a carboxyl group into the high molecular weight polymer by using a monoester obtained by reacting an acid anhydride such as maleic anhydride, succinic anhydride, etc. with methacrylic acid, acrylic acid or 2-hydroxyethyl (meth)-acrylate.

The molecular weight of the high molecular weight compound of the photocurable composition of the present invention has no particular limitation, but since it is necessary to coat the photocurable resin composition onto a base in the form of a uniform coating, usually it is preferred that the molecular weight is about 5,000 to 500,000 in terms of an average molecular weight as calculated based upon polystyrene. However, it is also possible to select an average molecular weight as calculated based upon polystyrene, other than those within the above range, depending upon the thickness of the coating formed, the coating process and the use object.

As the azide compound as a photosensitivity-imparting agent used in the present invention, known compounds such as 4-azidocalcon, 4,4'-diazidocalcon, 2,6-bis(4-azido-benzylidene)-4-methylcyclohexanone, etc. may be used.

The quantity of the photosensitivity-imparting agent blended has no particular limitation, but usually it is preferably in the range of 0.5 to 15 parts by weight based upon 100 parts by weight of the high molecular weight compound. If it is less than 0.5 part by weight, the curing speed of the coating is insufficient, while, if it exceeds 15 parts by weight, deposition of the photosensitivity-imparting agent occurs.

In the photocurable resin composition of the present invention, it is possible, if necessary, to add a sensitizer to the above monoazides or diazides.

As concrete examples of the sensitizer, naphthothiazolines and benzothiazolines represented by 1-methyl-2-benzoylmethylene-β-naphthothiazoline (hereinafter abbreviated to NT-1), and ketocoumalins represented by 3,3'-carbonyl-bis(7-diethylaminocoumalin), are preferred.

The quantity of the sensitizer blended has no particular limitation, but it is usually in the range of 0.5 to 15 parts by weight, preferably in the range of 1 to 10 parts by weight, based upon 100 parts by weight of the high molecular weight compound. If it is less than 0.5 part by weight, the effect of the sensitizer is small, and the curing speed does not increase as compared with that at the time of no addition, while if it exceeds 15 parts by weight, the solubility of the unexposed part in an alkali aqueous solution

lowers. By blending the above-mentioned respective constituting materials in the respective definite quantities, a photosensitive material, that is, the photocurable resin composition of the present invention is obtained.

When the above photosensitive material is used, conventional processes may be applied. For example, by tightly contacting a transparent original print having a linear printed image with a photosensitive surface, followed by exposing, thereafter developing with an alkali aqueous solution, a positive type or negative type relief image is obtained.

As to the photocurable resin composition of the present invention, when the composition is coated onto a base according to a conventional coating process, followed by irradiating by ultraviolet ray, it is possible to form an objective relief image due to the crosslinkability of azides. The thus obtained relief image has a high chemical resistance. The composition has a high mechanical strength. Further, its adhesion onto the base is high. Further, since it is highly sensitive, it is also suitable as a material for forming pigment-dispersion type color filters. Thus, the photocurable resin composition of the present invention is very useful.

The present invention will be described below in more detail by way of Examples, but it should not be construed to be limited thereto.

In Examples and Comparative examples, an ultra-high pressure mercury vapor lamp (UI-501C, trade name of product made by Ushio Co., Ltd.) was used for exposure. In the case of measuring the sensitivity on the side of a long wave length of 400 nm or more, HOYA color filter glass L42 (trade name of product made by Hoya Co., Ltd.) (hereinafter abbreviated to L42) was used. For measuring the quantity of exposure, an integrating actinometer (UIT-102, trade name of product made by Ushio Co., Ltd.) was used.

The development was carried out in a 0.05% aqueous solution of tetramethylammonium hydroxide, and a quantity of exposure in which the film thickness became constant after the exposure and development (hereinafter referred to as saturated coating thickness) was defined as sensitivity. The exposure was always carried out in air.

Example 1

3-Cyclohexene-1-methanol (112 g), methacrylic acid (172 g), sulfuric acid (2.8 g) and as polymerization inhibitors, phenothiazine (0.28 g) and 4-methoxyphenol (0.28 g), were placed in a flask equipped with a stirrer, followed by stirring at 80°C, for 4 hours, under 200 mmHg.

At that time, water (26 g) distilled off. The reaction solution was distilled. A fraction of 53-55°C at 2 mmHg (153 g) (GC purity: 99%) (GC: abbreviation of gas liquid chromatography) was obtained. Its structure was confirmed to be of 3-cyclohexenylmethyl methacrylate, according to IR spectra and H^1NMR spectra.

Next, 3-cyclohexenylmethyl methacrylate (30 g), 2-hydroxyethyl methacrylate (30 g) and methacrylic acid (30 g) were dissolved in methanol (225 g), followed by adding 2,2'-azobisisobutyronitrile (0.27 g), heating the mixture at 65°C for 6 hours, feeding the resulting material into ethyl acetate (1,500 g) and drying the deposited precipitates under reduced pressure to obtain a high molecular weight compound (65 g).

This high molecular weight compound (1.5 g), 2,6-di(4-azidobenzylidene)-4-methylcyclohexanone (hereinafter abbreviated to AZ-1) (0.045 g), 1-methyl-2-benzoylmethylene-ß-naphtothiazoline (0.045 g), methyl cellosolve (9 mℓ) and N-methyl-2-pyrrolidone (1 mℓ) were mixed and dissolved to obtain a photocurable resin composition.

This photocurable resin composition was spin-coated onto a glass base at 1,000 rpm, followed by heating at 110°C for 10 minutes, and then exposing. The coating thickness before the exposure was 1.50 μm and the saturated coating thickness after exposure and development was 1.35 μm. The sensitivity at that time was 30 mJ/cm^2.

Further, when the heat treatment before the exposure was carried out at 85°C for 10 minutes, the coating thickness before the exposure was 1.55 μm and the sensitivity at that time was 45 mJ/cm^2.

Example 2 (Use example)

The photocurable resin composition of Example 1 was spin-coated onto a glass base at 1,000 rpm, followed by heating at 110°C for 10 minutes and then exposing the resulting material through L42. The sensitivity at that time was 80 mJ/cm^2 and the saturated coating thickness after the exposure and development was 1.30 μm.

Example 3

Acetone (350 g), triethylamine (121 g), α-terpineol (154 g) and phenothiazine as a polymerization inhibitor (0.7 g) were placed in a flask equipped with a stirrer, followed by slowly dropwise adding methacrylic chloride (110 g) while keeping the temperature at 0° to 5°C, thereafter agitating the mixture at 50°C for 2 hours, filtering off the deposited salt, and distilling off acetone for concentration to obtain a fraction of 97 to 103°C at 2 mmHg (115 g) (GC purity: 95%). When its structure was determined by IR spectra and H^1NMR spectra, it was confirmed to be 4-methyl-3-cyclohexen-1-yl-dimethyl methacrylate.

Next, the above compound (30 g) and mono(2-methacryloxyethyl) maleate (40 g) were dissolved in

methanol (220 g), followed by adding 2,2'-azobisbutyronitrile (0.28 g), heating the mixture at 65°C for 6 hours, feeding the resulting material into ethyl acetate (1,500 g) and drying the deposited precipitates under reduced pressure to obtain a high molecular weight compound (43 g).

The above high molecular weight compound (1.5 g) AZ-1 (0.05 g), methyl cellosolve (9 mℓ) and N-methyl-2-pyrrolidone (1 mℓ) were mixed and dissolved to obtain a photocurable resin composition, followed by exposing this composition in the same manner as in Example 1. The coating thickness before exposure at the time of heating at 110°C was 1.50 μm, the saturated coating thickness after exposure and development was 1.30 μm and the sensitivity was 80 mJ/cm^2. The coating thickness before exposure at the time of heating at 85°C was 1.60 μm, the saturated coating thickness after exposure and development was 1.30 μm and the sensitivity at that time was 120 mJ/cm^2.

Example 4 (Use example)

The photocurable resin composition of Example 3 was exposed in the same manner as in Example 2. The sensitivity at that time was 200 mJ/cm^2.

Example 5

Acetone (500 g), 2-(1-cyclohexenyl)ethylamine (77 g), triethylamine (155 g) and phenothiazine as a polymerization inhibitor (0.5 g) were placed in a flask equipped with a stirrer, followed by slowly dropwise adding methacrylic chloride (71 g) while keeping the temperature at 0° to 5°C,

thereafter agitating at 25°C for 2 hours, filtering off the deposited salt, distilling off acetone for concentration and washing with water to obtain a fraction of 145 to 147°C at 1 mmHg (80 g) (GC purity: 94%). When its structure was determined by IR spectra and H^1NMR spectra, it was confirmed to be N-[2-(1-cyclohexenyl)ethyl] methacrylamide.

Next, N-[2-(1-cyclohexenyl)ethyl]methacrylamide (30 g), 2-hydroxyethyl methacrylate (40 g) and methacrylic acid (30 g) were dissolved in methanol (300 g), followed by adding 2,2'-azobisisobutyronitrile (0.5 g), heating the mixture at 65°C for 4 hours, feeding the resulting material into ethyl acetate (2,000 g), and drying the deposited precipitates to obtain a high molecular weight compound (54 g).

The above compound (1.5 g), 4,4'-diazidocalcon (0.06 g), methyl cellosolve (9 mℓ) and N-methyl-2-pyrrolidone (1 mℓ) were mixed and dissolved to obtain a photocurable resin composition, followed by exposing this composition in the same manner as in Example 1. The coating thickness before the exposure at the time of heating at 110°C was 1.20 μm, the saturated coating thickness after exposure and development was 1.10 μm and the sensitivity at that time was 80 mJ/cm^2. The coating thickness before exposure at the time of heating at 85°C was 1.30 μm, the saturated coating thickness after exposure and development was 1.05 μm and the sensitivity at that time was 120 mJ/cm^2.

Example 6 (Use example)

3,3'-carbonylbis(7-diethylaminocoumarin) (0.075 g) was added to the photocurable resin composition of Example 5, followed by exposure in the same manner as in Example 2. The sensitivity at that time was 150 mJ/cm^2.

Comparative example 1

Methyl methacrylate-methacrylic acid copolymer (wt. ratio 7:3) (1.0 g), trimethylolpropane triacrylate (0.3 g), 2,4,6-tris(trichloromethyl)triazine (hereinafter abbreviated to TCT) (0.012 g), NT-1 (0.04g), methyl cellosolve(9 mℓ) and N-methyl-2-pyrrolidone (1 mℓ) were mixed and dissolved to obtain a photocurable resin composition.

This composition was spin-coated onto a glass base at 900 rpm, followed by heating at 85°C for 10 minutes, and then exposing the resulting material. The coating thickness before the exposure was 1.10 μm, the saturated coating thickness after exposure and development was 0.70 μm and the sensitivity at that time was 150 mJ/cm^2.

Further, when the heat treatment before the exposure was carried out at 75°C for 10 minutes, the coating thickness before exposure was 1.20 μm, the saturated coating thickness after exposure and development was 0.50 μm and the sensitivity at that time was 240 mJ/cm^2.

Comparative example 2 (Comparative use example)

The photocurable resin composition of Comparative exmaple 1 was spin-coated onto a glass base at 900 rpm, followed by heating at 85°C for 10 minutes and then exposing the resulting material through L42. The sensitivity at that time was 180 mJ/cm^2 and the saturated coating thickness after exposure and development was 0.40 μm.

Example 7 (Application example)

Using the high molecular weight compound obtained in Example 1, together with other components, the following pigment inks for color filters were prepared.

Red pigment ink:

Polymer	17 g
2,6-Di(4-azidobenzylidene)-4-methylcyclohexanone	0.5 g
Cromophtal Red A3B (trade name of product made by Chiba Geigy Co., Ltd.)	5 g
Seikafast Yellow 2700 (trade name of product made by Dainichi Seika Co., Ltd.)	1.0 g
Diethylene glycol monoethyl ether	90 mℓ
N-methyl-2-pyrrolidone	10 mℓ

Green pigment ink:

Polymer	17 g
2,6-Di(4-azidobenzylidene)-4-methylcyclohexanone	0.5 g
Cyanine Green S573-2y (trade name of product made by Dainichi Seika Co., Ltd.)	6 g
Seikafast Yellow 2700 (trade name of product made by Dainichi Seika Co., Ltd.)	1.2 g
Diethylene glycol monoethyl ether	90 mℓ
N-methyl-2-pyrrolidone	10 mℓ

Blue pigment ink:

Polymer	17 g
4,4'-Diazidodiphenylmethane	0.7 g
Chromofine Blue 4920 (trade name of product made by Dainichi Seika Co., Ltd.)	5 g
Chromofine Violet RE (trade name of product made by Dainichi Seika Co., Ltd.)	1.2 g
Diethylene glycol monoethyl ether	90 mℓ
N-methyl-2-pyrrolidone	10 mℓ

Black pigment ink:

Polymer	17 g
2,6-Di(4-azidobenzylidene)-4-methylcyclohexanone	0.5 g
Denka Black (trade name of product made by Denki Kagaku Co., Ltd.)	6 g
Diethylene glycol monoethyl ether	90 mℓ
N-methyl-2-pyrrolidone	10 mℓ

These coloring inks were respectively coated onto separate glass bases by means of a spin-coater at 1,200 rpm, followed by heat-treating at 110°C for 10 minutes and exposing the resulting materials in air by means of a superhigh pressure mercury vapor lamp (UI-501C, trade name of product made by Ushio Co., Ltd.), by the medium of a mask of stripe pattern.

After the exposure, the resulting materials were immersed in a 0.05% aqueous solution of sodium carbonate for 2 minutes, followed by washing with flowing water for 15 seconds to remove unexposed part. Varying the exposing quantity, the above operation was carried out, and with the respective colors, exposing quantities in which the coating thickness after exposure became constant were sought (but only the black ink was heat-treated at 110°C for 5 minutes after the exposure).

When the saturated coating thickness after the exposure was made 1.7 μm, the sensitivities were 100 mJ/cm^2 in the case of red, 150 mJ/cm^2 in the case of green, 300 mJ/cm^2 in the case of blue, and 300 mJ/cm^2 in the case of black, and even in the case where a pigment is contained and exposure is carried out in air, it is possible to form a pattern sufficiently in a quantity of exposure similar to that in the conventional process for forming color filter; hence the above sensitivities are very high.

Fig. 4 shows the spectra of the colored printed image formed under the above conditions. Next, colored patterns of red, green, blue and black were successively formed on the same glass base under the above conditions. The spectra of the respective colors accorded with those of Fig. 2. This shows that neither color mixing nor decoloring occurred during the step.

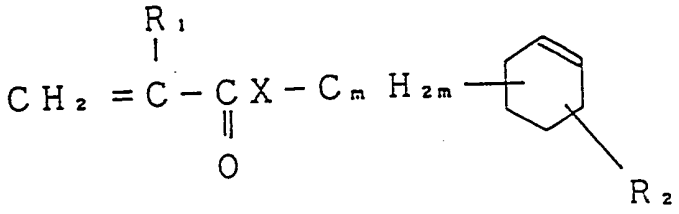
(Effectiveness of the Invention)

The photoreactive compound and the photocurable resin composition using the same, of the present invention are cured in a small quantity of exposure even in air and also the pretreatment temperature has a small influence upon the saturated coating thickness after exposure and development; hence they are more advantageous than conventional photocurable resin compositions in the productivity, energy-saving and equipments, and further, since it is possible to easily remove the unexposed part with an alkali aqueous solution, the environment of workshops is improved.

Further, since the composition is very sensitive to light, it is suitable as a material for forming color filters of pigment dispersion type.

CLAIMS:

1. Photoreactive compounds of the formula:



(I)

wherein R_1 is a hydrogen atom or a methyl or ethyl group; X is -O- or -NH-; R_2 is a hydrogen or halogen atom or a C_1-C_4 alkyl or C_1-C_4 alkoxy group; m is an integer of from 1 to 5.

2. A photocurable resin composition obtained by adding an azide compound, as a photosensitivity-imparting agent, to a high polymeric material obtained by polymerizing a photoreactive compound as claimed in claim 1.

3. A photocurable resin composition according to claim 2, in which the polymeric material is obtained by copolymerizing the photoreactive compound an ethylenic double bond and a carboxylic group in the same molecule.

4. A colour filter material comprising a colouring pigment dispersed in a photocurable resin composition as claimed in claim 3.

Patents Act 1977 21
Examiner's report to the Comptroller under Section 17 (The Search Report)

Application number

9203190.5

Relevant Technical fields

(i) UK Cl (Edition K) C2C CKG CBZ

(ii) Int CL (Edition 5) C07C

Search Examiner

D S LUCAS

Databases (see over)

(i) UK Patent Office

(ii) ONLINE DATABASE: CAS ONLINE

Date of Search

26 MARCH 1992

Documents considered relevant following a search in respect of claims 1-4

Category (see over)	Identity of document and relevant passages	Relevant to claim(s)
X	US 3536687 A (ASHLAND OIL)	1-4
X	FR 001527054 A (UGINE KUHLMANN)	1-4
X	FR 001511011 A (UGINE KUHLMANN)	1-4
X	DE 001900346 A (UGINE KUHLMANN)	1-4
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X	Tetrahedron Letters 31 (51) 7457-60 (1990) (TOKYO INST. TECH)	1-4
X	Chemical Abstracts 101 (15): 130271d (GAIBOVA)	1-4

Category	Identity of document and relevant passages	Relevant to claim(s)

Categories of documents

X: Document indicating lack of novelty or of inventive step.

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A: Document indicating technological background and/or state of the art.

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E: Patent document published on or after, but with priority date earlier than, the filing date of the present application.

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