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(54) Title: PHOTOCURABLE COMPOSITION FOR PREPARING LENSES

(57) Abstract

A photocurable composition for preparation of a lens including: a polythiol compound; an aromatic monomer containing at least two vinyl groups; an acrylate monomer containing at least two unsaturated groups and selected from the group consisting of acrylates, methacrylates, acrylic acid anhydrides and methacylic anhydrides; and a photoinitiator.

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PHOTOCURABLE COMPOSITION FOR PREPARING LENSES

The present invention relates to photocurable compositions and their use in forming high index lenses.

The manufacture of lenses such as opthalmic lenses by photocuring of a resin composition has been previously described, for example in our prior Applications PCT/AU96/00247 and PCT/AU97/00310. One photocurable system known as the thiol-ene system involves polythiol olefin copolymerization. The thiol-ene system has the potential of allowing lenses to be prepared with high refractive index to allow lens thickness to be minimised and to be more attractive. However we have found that the thiol-ene system is difficult to cure efficiently particularly in resin thicknesses of at least 1mm which are generally required for opthalmic lenses. Further the thiol-ene system involves a complex range of reactions which are difficult to control and may result in an unacceptable proportion of unreacted monomer and poor rigidity.

Alternative systems such as urethane acrylate systems disclosed in PCT/AU97/00247 may be cured efficiently. Such systems however do not generally allow the high refractive indexes theoretically possible with the thiol-ene system to be readily attained. Furthermore the preparation of lenses, which are typically of thickness of 1 to 15 mm, creates significant difficulties for compositions containing vinyl substituted aromatics. It is difficult to achieve even curing of a cast of this thickness since the depth of penetration of active rays in a photosensitive composition is generally small. This is especially a problem for compositions containing divinyl benzene or the like.

As a result it has not generally been possible to achieve the balance of good mechanical properties and high refractive index required for the commercial use of photosensitive compositions in preparing lenses.

There is therefore a need for a photocurable lens composition and method of preparing lenses which allows lenses to be cured rapidly to provide a high refractive index and the required mechanical properties.

The invention provides a photocurable resin composition for preparing

lenses including:

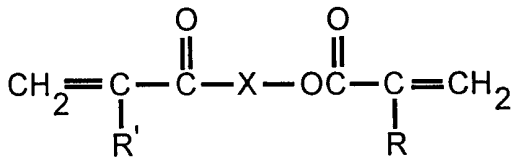
a polythiol compound;

an aromatic monomer containing at least two vinyl functional groups;

an acrylate monomer containing at least two unsaturated groups and selected from acrylates, methacrylates, acrylic acid anhydrides and methacrylic anhydrides; and

a photoinitiator preferably including a phosphine oxide.

The acrylate monomer is preferably of Formula I:



I

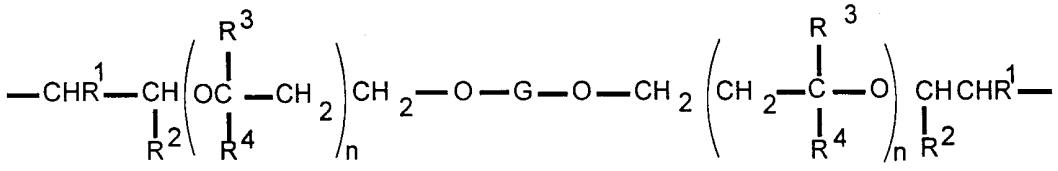
wherein R and R' are independently selected from hydrogen and C_1 to C_4 alkyl and most preferably are hydrogen or methyl;

X is a bond or a group of Formula -O(L)- wherein L is a linking group optionally substituted with one or more acrylic or methacrylic groups.

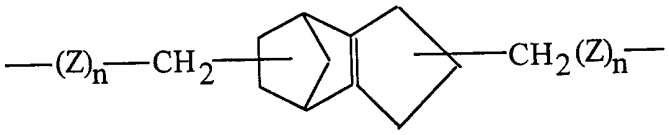
Examples of linking groups include alkylene, preferably of 1 to 6 carbon atoms, carbocyclic, heterocyclic and polyalkylene oxides. Such linking groups may be further substituted with from 1 to 3 acrylate and/or methacrylate groups.

Suitable examples of linking groups include alkylene glyol di(meth)-acrylates and polyalkylene glycol di(meth)acrylates.

Further examples of L include groups of Formula IIa or Iib



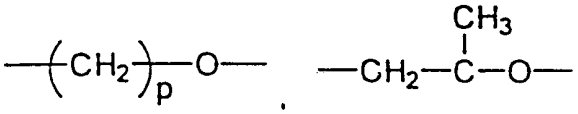
IIa



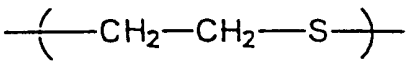
IIb

wherein

Z is a spacer group selected from one or more of the following



or

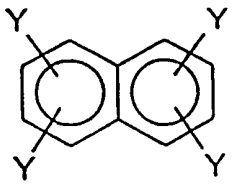


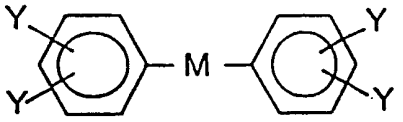
, or derivatives thereof;

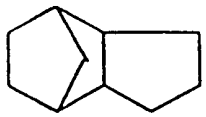
wherein p = 1-4

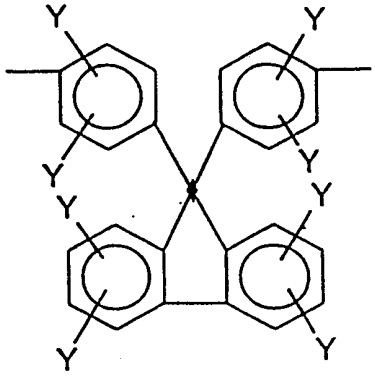
R^2 is hydrogen or hydroxy;

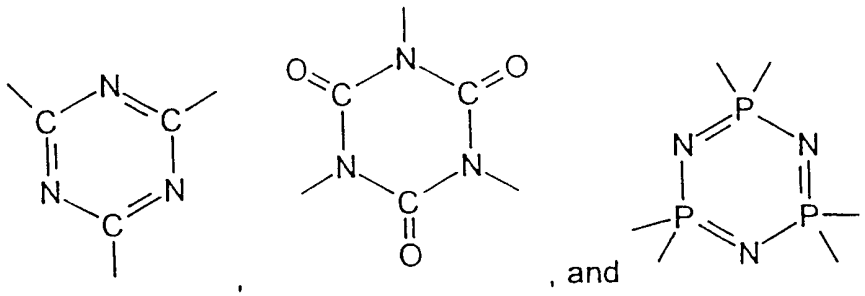
G is selected from the group consisting of



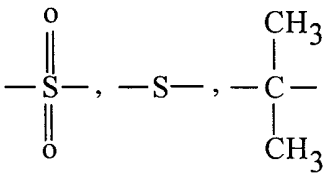








wherein M is selected from the group consisting of a bond,



methylene and $-CH_2-S-CH_2-$

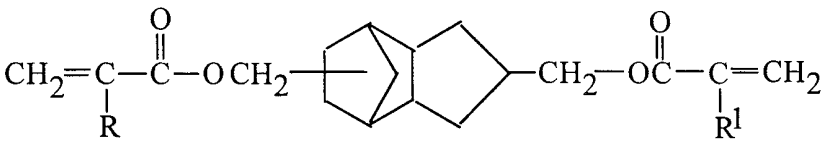
R_1, R_3 and R_4 are independently selected from the group consisting of hydrogen and alkyl of 1 to 6 carbon atoms; and

n is an integer having a value of 0 to 10,

Y is hydrogen or a halogen, preferably fluorine, chlorine or bromine.

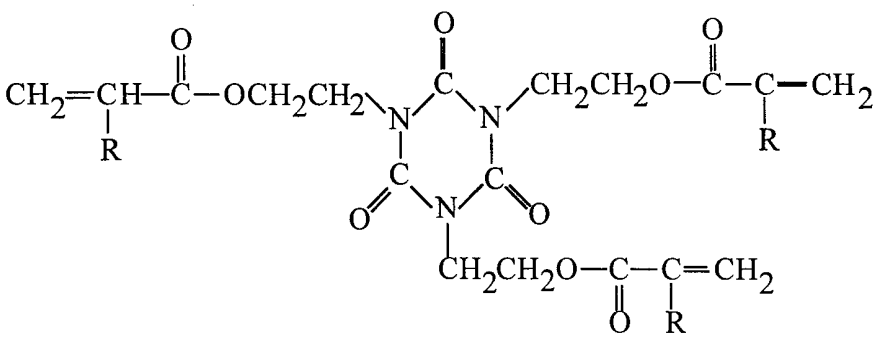
Preferred compounds of Formula I include compounds wherein X is a bond and the most preferred acrylate monomer of this type is methacrylic acid anhydride. Compounds of Formula I in which X is a bond have been found to significantly improve curing efficiency.

Further particularly preferred acrylate monomers are selected from the group of compounds of formula

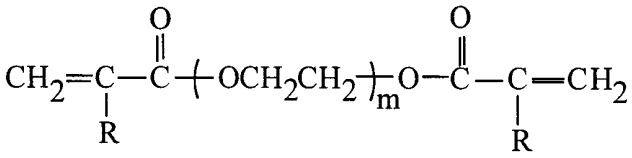


wherein R and R^1 are independently selected from hydrogen and methyl;

compounds of formula:



where R is methyl or hydrogen; and compounds of formula:



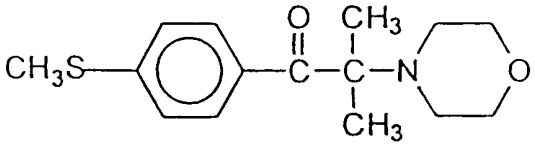
wherein R is methyl or hydrogen, and m is an integer of 2 to 15 and most preferably 9.

The acrylate component is typically present in an amount of from 10 to 40% by weight and preferably from 15 to 25% by weight of the total composition.

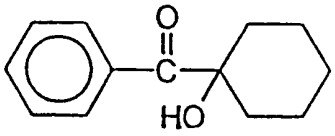
The compositions of the invention include a photoinitiator which has sufficient absorption in the far UV and near visible regions of the spectrum. Photoinitiators exhibiting strong absorption in the range of from 350 to 450nm are particularly preferred. The preferred chemical class of initiators is the phosphine oxides particularly the acylphosphine oxides including bisacyl phosphine oxides or their derivatives such as ester derivatives. The most preferred photoinitiators are 2,4,6-trimethylbenzoyl-diphenylphosphine oxide (available under the brand name LUCIRIN TPO from BASF), bis-(2,4,6-trimethylbenzoyl)phenyl phosphine oxide (available under the brand IRGACURE 819 from Ciba) and bis-(2,6-dimethoxybenzoyl)-2,4,4-trimethylpentyl phosphine oxide (known by the brand name BAPO-1 and available in admixture with an hydroxy-α,α-dialkylacetophenone from Ciba. Another suitable product is LUCIRIN LR 8893X brand initiator from BASF) ethyl 2,4,6-trimethyl benzoyl phenyl phosphinate.

Photoinitiator combinations may be used. For example, acylphosphine oxides may be used in combination with benzophenones, benzoin eithers or hydroxy-α,α-dialkylacetophenones. Specific examples of other photoinitiators include:

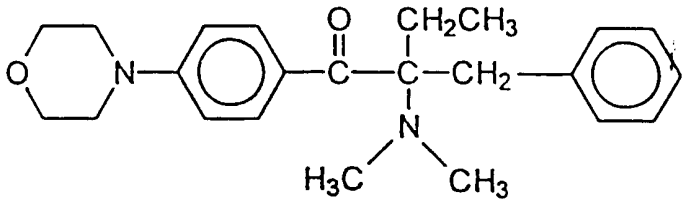
2-methyl-1-[4-(methylthio)phenyl]-2-morpholinopropane-1-one (Irgacure 907)



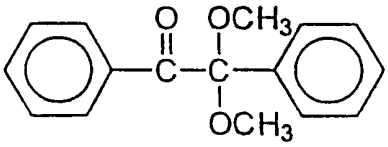
(hydroxycyclohexyl) phenyl ketone (Irgacure 184)



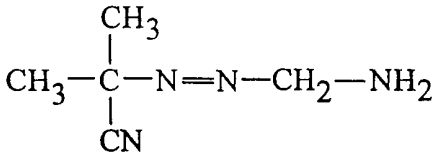
(2-benzyl-2-N-dimethylamino-1-(4-morpholinophenyl)-1-butanone) (Irgacure 369)



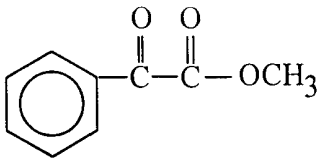
(Benzyl Dimethyl Ketal) (Lucirin BDK; Irgacure 651)



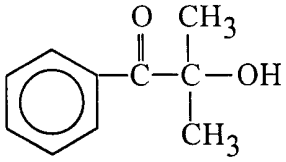
2-(carbamoylazo)-substituted (Vicure 30)



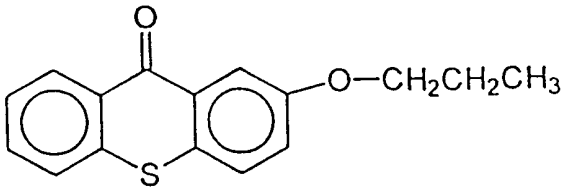
1-methoxy-2-phenyl-1,2-ethandione (VICURE 55)



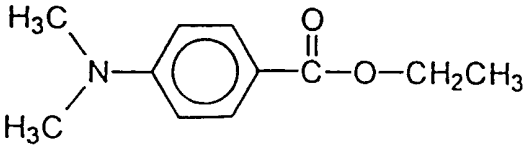
α-hydroxy-α,α-dimethylacetophene (DAROCURE 1173)



2-n-propoxy-9H-thioxanthen-9-one (WB 4744)



Ethyl 4-(dimethylamino)benzoate (Quantacure EPD)



Examples of suitable mixture of initiators containing phosphine oxides include:

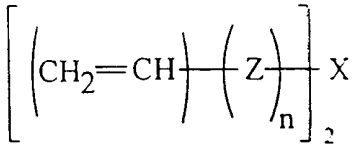
IRGACURE 1700 - a mixture of 25% BAPO-1 and 75% DAROCURE 1173,

IRGACURE 1800 - a mixture of 25% BAPO-1 and 75% Irgacure 184, and

IRGACURE 1850 - a mixture of 50% BAPO-1 and 50% IRGACURE 184.

The amount of photoinitiator present in the compositions of the invention is typical in the range from 0.1 to 2% by weight. We have obtained particularly good results by using from 0.2 to 1.5% by weight of an acylphosphine oxide photoinitiator.

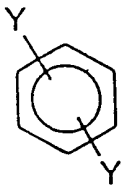
The aromatic monomer containing at least two vinyl functional groups is preferably of Formula III



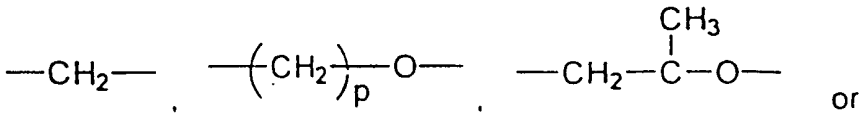
III

wherein

X is



Z is a spacer group selected from one or more of the following



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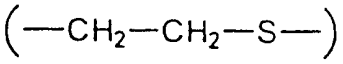


, or derivatives thereof;

Y is hydrogen or halogen;

n = 0, 1 or 2;

p = 1 - 4.

In the preferred compounds of Formula III n is zero, that is the linking group is not present, and X is an aromatic group particularly benzene. The most preferred said monomer containing at least two vinyl groups is divinyl benzene.

The amount of said monomer containing at least two vinyl functional groups is typically in the range of from 20 to 40% by weight and more preferably from 26 to 34% by weight based on the total composition.

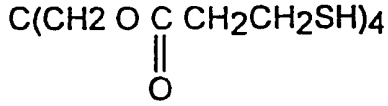
In the composition of the present invention it is particularly preferred that an aromatic divinyl compound be present in a stoichiometric (molar) excess with respect to the polythiol compound.

We have obtained particularly good results using divinyl benzene. This was contrary to what was expected because photopolymer compositions containing divinyl benzene in combination with acrylates are generally difficult to cure particularly in thick casts. The presence of an excess of divinyl compound would merely be expected to exacerbate these difficulties. We have found that compositions containing divinyl benzene, a polythiol compound and an acrylate type monomer, preferably where the divinyl benzene is present in excess of the polythiol result in lenses high refractive index lenses and good mechanical properties when a photoinitiator having significant absorption in the range 350 to 450nm is used (particularly an acylphosphine oxide) in an amount of from 0.1 to 2% by weight of the total composition.

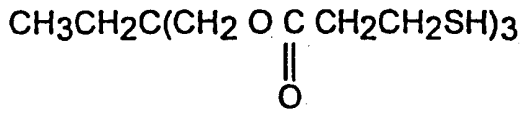
Accordingly a particularly preferred embodiment of the invention proves a photocurable composition for the preparation of lenses including divinyl benzene; a polythiol compound wherein the divinyl benzene is present in a stoichiometric excess of the polythiol compound; an acrylate type monomer (preferably methacrylic acid anhydride); and a photo initiator, preferably an acylphosphine oxide, having significant absorption at a wavelength in the range of from 350 to 450nm in an amount of from 0.1 to 2% by weight of the total composition.

The polythiol compound used in the composition of the invention will include at least two thiol groups and preferably at least three thiol groups. The polythiol compound is preferably selected from one or more of the following:

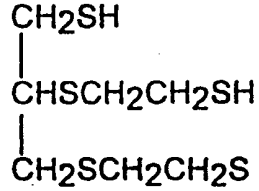
(a) Pentaerythritol Tetrakis (3-mercapto-propionate) [PTMP]



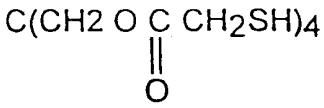
(b) Trimethylolpropane Tris (3-mercaptopropionate) [TTMP]



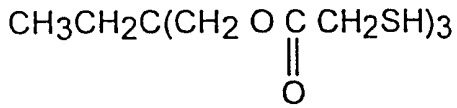
(c) 4-mercaptomethyl-3,6-dithia-1,8-octanedithiol [MDO]



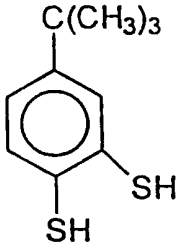
(d) Pentaerythritol Tetrakis (3-mercaptoacetate) [PTMA]



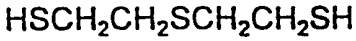
(e) Trimethylolpropane Tris (3-mercaptoacetate) [TTMA]



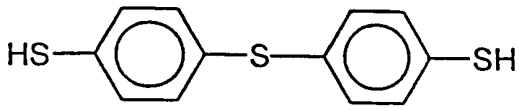
(f) 4-t-butyl-1,2-benzenedithiol



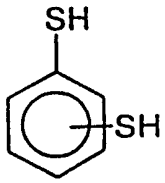
(g) 2-mercaptoethylsulfide



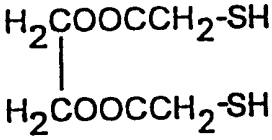
(h) 4,4'-thiodibenzenethiol



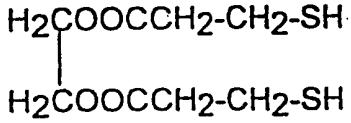
(i) benzenedithiol



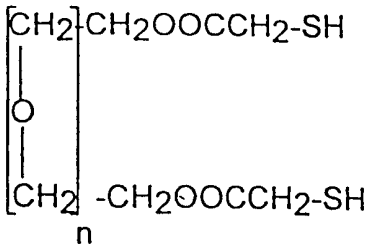
(j) Glycol Dimercaptoacetate



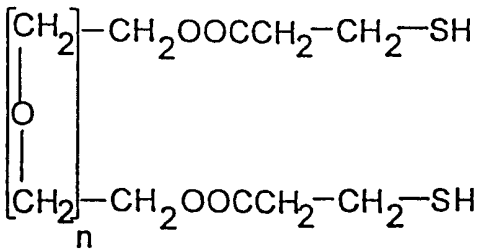
(k) Glycol Dimercaptopropionate Ethylene bis(3-Mercaptopropionate)



(l) Polyethylene Glycol Dimercaptoacetates



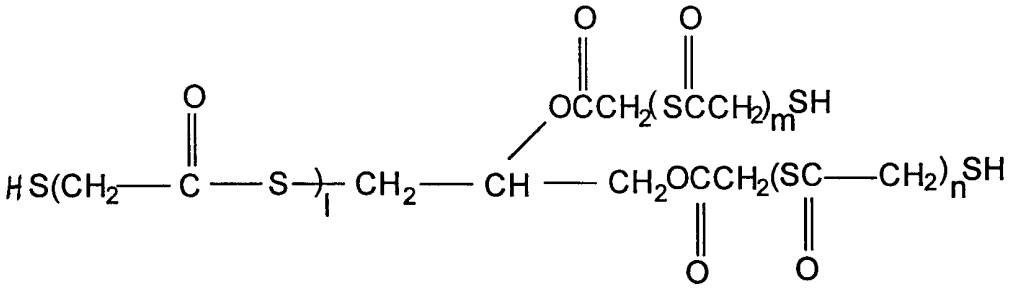
(m) Polyethylene Glycol Di(3-Mercaptopropionates)



(n) 2,5-bis(mercaptomethyl)-1,4-dithiane

(o) 3,6-dithia-1,8-octanedithiol

(p) TGBMA (a mixture of oligomers)

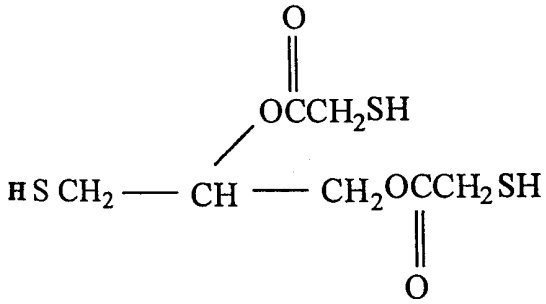


l = 0, 1, 2, 3 ...

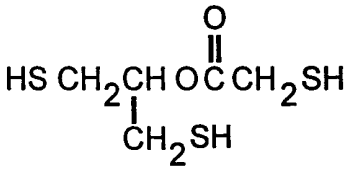
m = 0, 1, 2, 3 ...

n = 0, 1, 2, 3 ...

12



(q) DMPMA



The thiol compound may preferably be present in amounts of from approximately 30 to 50% by weight, more preferably approximately 35 to 45% by weight, based on the total weight of the casting composition.

We have found that the compositions of the invention may include one or more UV absorbers without having a significantly deleterious effect on curing efficiency. As UV absorbers reduce the penetration of light in thick compositions they have in many cases been found to lead to inefficient curing particularly at the centre of the cast.

Examples of preferred UV absorbers include:

2(2'-hydroxy-5'methyl phenyl) benzotriazole, (Ciba Geigy "Tinuvin P"),

2-hydroxy-4-n-octoxybenzo-phenone, (Cyanamid "Cyasorb UV" 531),

2(2-hydroxy-5-t-octylphenyl)-benzotriazole, (Cyanamid "Cyasorb UV5411"),

2 hydroxy-4-(2-acryloyloxyethoxy)-benzophenone (Cyanamid "UV 2098"),

2 hydroxy-4-(2 hydroxy-3-methacryloxy)propoxy benzophenone (National Starch and Chemicals "Permasorb MA"),

2,2'-dihyroxy-4-methoxybenzophenone (Cyanamid "UV24"),

2,4 dihydroxy-benzophenone (BASF "UVINUL 400"),

2,2'-dihydroxy-4,4'dimethoxy-benzophenone (BASF "UVINUL D-49"),

2,2', 4,4' tetrahydroxy benzophenone (BASF "UVINUL D-50"),
ethyl-2-cyano-3,3-diphenyl acrylate (BASF "UVINUL D-35")
2-ethexyl-2-cyano-3,3-diphenyl acrylate (BASF "UVINUL N-539") and
Ciba Geigy Tinuvin 213.

The compositions of the invention are preferably used in casting of lense of thickness in the range of from 1 to 15mm. Accordingly in a further embodiment the invention provides a method of preparing lenses including forming a cast of the hereinabove described composition with a thickness in the range of from 1 to 15mm and irradiating the cast composition with light in the UV-visible range. Preferably the light includes a significant wavelength component in the range of from 350 to 450nm. The "FUSION V" brand lamp is a good source of the appropriate range of UV light.

The period of irradiation of UV-visible light may be reduced significantly when compared with previously known high index lens casting compositions and is typically from several minutes to an hour. Indeed it is preferred that irradiation occurs for a period of from 0.1 to 5 minutes.

In a preferred embodiment the invention utilises the post irradiation heat treatment. In this embodiment, the composition will typically include a heat curing agent adapted to induce curing a relatively high temperature of for example from 80°C to 130°C.

Examples of heat curing agents include:

- AIBN (Azo radical heat initiator)
  Azodiisobutyronitrile,
- Trigonox TX-29 (Dialkyl Peroxide radical head intiator)
  1,1-di-(butyl peroxy-3,3,5-trimethyl cyclohexane),
- TBPEH (Alkyl Perester radical heat initiator)
  t-butyl per-2-ethylhexanoate,
- (Diacyl Peroxide radical heat initiator)
  Benzoyl Peroxide,
- (Peroxy Dicarbonate radical heat initiator)
  Ethyl Hexyl Percarbonate,

- (Ketone Peroxide radical heat initiator)
  Methyl ethyl ketone peroxide,
- 1,1-bis(t-butylperoxy)cyclohexane,
- 2,2-bis(t-butylperoxy)butane,
- Dicumyl peroxide,
- t-Butyl cumyl peroxide,
- 2,5-Dimethyl hexane 2,5-di-t-butyl peroxide,
- Di-t-butyl peroxide,
- Bis(t-butyl peroxide) diisopropylbenzene,
- t-butyl perbenzoate, and
- t-butyl peroxy neodecanoate

Examples of other additives include:

- Amicure DBU,
- Amicure BDMA and
- DABCO

Amicure DBU and/or Amicure BDMA are preferred.

Dialkyl peroxide radical heat initiators are particularly preferred. Fatty alkyl peroxides such as 2,5-dimethyl hexane 2,5-di-t-butylperoxide are the most preferred.

The amount of heat initiator is typically in the range of from 0.05 to 1% by weight and generally the thermal treatment will be carried out by maintaining a temperature of at least 80°C for a period of at least 10 minutes. When present the heat initiator is generally used in amounts of at least 0.05%.

The invention will now be described with reference to the following examples. It should be understood, however, that the description following is illustrative only and should not be taken as a restriction on the generability of the invention described above.

Example 1

The composition listed in the table below was cast into lens moulds to provide a thickness of 2.0mm

	AMOUNT
DVB	32.5g
Methacrylic anhydride	20.0g
TG BMA	39.0g
PTMA	8.5g
UV 5411	0.2g
Irganox 1010	0.2g
TPO (Lucirin)	1.2g
TX-29	0.5g

The composition was irradiated using a EPIQ 6000 V-bulb spectrum lamp (Fusion UV curing Systems Corporation) having a power of 240w/cm with one pass under the lamp at a rate of 9mm/second. Post irradiation heat treatment was carried out at 100°C for 1 hour.

The resulting lens was found to have a refractive index of 1.599 and Abbe number of 36.5.

Example 2

The procedure of Example 1 was repeated with the exception that the heat initiator was omitted. The composition was cured under the conditions used in Example 1. The heat treatment condition was 120°C for 1.5 hours.

Example 3

The composition of Example 1 used in this example with the exception that the photoinitiator used was a mixture of 1g of TPO and 1.0g of DAROCURE 1173. DAROCUR 1173, available from Ciba, has as the active agent 2-hydroxy-2-methyl-1-phenylpropan-1-one.

A 2.0mm thick lens was cured by passing it twice beneath the lamp at a speed of 24mm/sec and was heat treated at 100°C for 1 hour.

Example 4

The procedure of Example 1 was repeated except that methacrylic anhydride was replaced with the same amount of ethylene glycol dimethacrylate. Effective case was provided under the conditions used in Example 1.

Examples 5-9

Formulation Examples 5 to 9 were prepared from the compositions shown in Table 1 and were cast into semi-finished lenses of 8mm thickness.

The lenses were cured by subjecting the photochemical compositions to the following conditions.

(a) UV curing using a FUSION V lamp with the back surface uppermost for a period of about 0.5 minutes.

(b) Maintain the composition at about 120 to 130°C for about one and half hours; and

(c) Subjecting the lens with the front surface uppermost to further irradiation for a period of about 0.5 minutes.

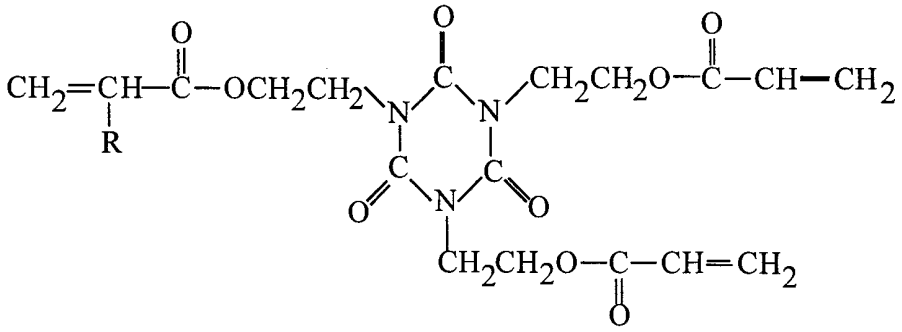
The properties of the resulting cured lenses are provided in the table.

Following formulations were casted into thick semi-finished lenses (8 mm) and stock lenses with the corresponding curing conditions, giving low strain and fully cured lenses.

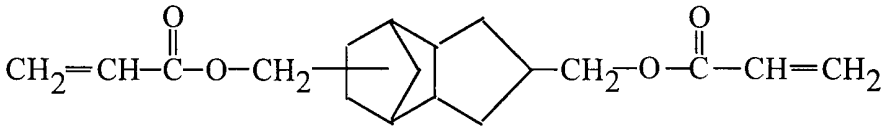
components	formulation 5 (grams)	formulation 6 (grams)	formulation 7 (grams)	formulation 8 (grams)	formulation 9 (grams)
DVB	32.5	28.5	28	32.5	29.8
MAA	20			10	8.1
A-9300		32	17	8.5	17.6
DCP-A			16.5		
SR-480				10	
NK ESTER 9G					4
TGBMA	39	39.5	38.5	39	40.5
PTMA	8.5				
TBC	0.1	0.1	0.1	0.1	0.1
UV5411	0.2	0.2	0.2	0.2	0.2
Irganox 1010	0.2	0.2	0.2	0.2	0.2
blue dye	1 ppm	1 ppm	1 ppm	1 ppm	1 ppm
Q1301	20 ppm	20 ppm	20 ppm	20 ppm	20 ppm
Irgacure 819	0.9	0.3	0.35	0.6	0.6
lucirin TPO	0.2	0.2	0.2	0.2	0.2
Interox DHBP	0	0.2	0.2	0	0
properties of lenses					
Refractive index	1.599	1.596	1.592	1.595	1.593
Abbe number	36.5	37	38	36.5	37
final conversion	95.0%	96.0%	95.8%	95.6%	95.0%
Tg°C	112	90	90	95	95

DVB - divinyl benzene

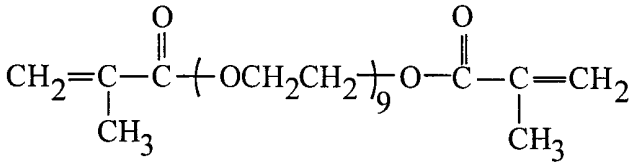
A-9300



DCP-A - Diacryloyl oxymethyl tricyclodecane



NK - Ester 9G Polyethylene Glycol Dimethacrylate.



SR480 - ethoxylated (10) Bisphenol A dimethacrylate

MAA - Methacrylic anhydride

Claims:

1. A photocurable composition for preparation of a lens including:

a polythiol compound;

an aromatic monomer containing at least two vinyl groups

an acrylate monomer containing at least two unsaturated groups and selected from the group consisting of acrylates, methacrylates, acrylic acid anhydrides and methacrylic anhydrides; and

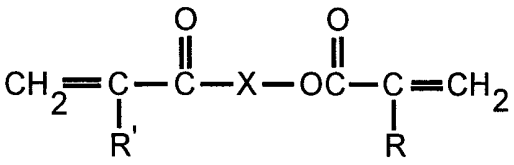
a photoinitiator.

2. A photocurable composition according to claim 1 wherein the aromatic monomer containing at least two vinyl groups is present in an amount of from 20 to 40% by weight of the composition.

3. A photocurable composition according to claim 1 wherein aromatic monomer containing at least two vinyl groups is present in a stoichiometric excess compared with the number of moles of the polythiol compound and preferably in the range of from 26 to 34% by weight.

4. A photocurable composition according to claim 1 wherein the photoinitiator includes a phosphine oxide initiator.

5. A photocurable composition according to claim 1 wherein the acrylate monomer is of Formula I

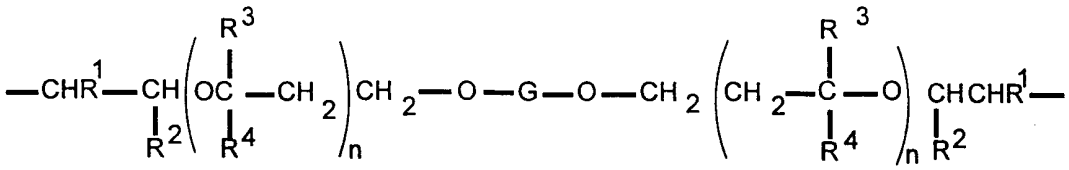


I

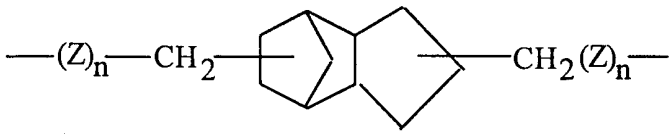
wherein R and R' are independently selected from hydrogen and C_1 to C_4 alkyl and most preferably are hydrogen or methyl;

X is a bond or a group of Formula -O(L)- wherein L is a linking group optionally substituted with one or more acrylic or methacrylic groups.

6. A photocurable composition according to claim 5 wherein X is of formula -O(L)- and L is of formula



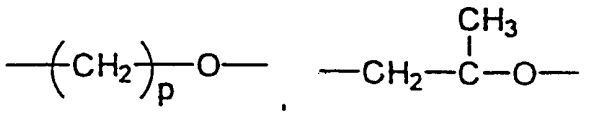
IIa



or IIb

wherein

Z is a spacer group selected from one or more of the following

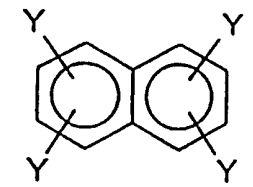


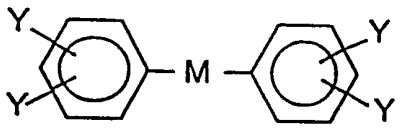
, or derivatives thereof;

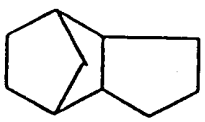
wherein p = 1-4

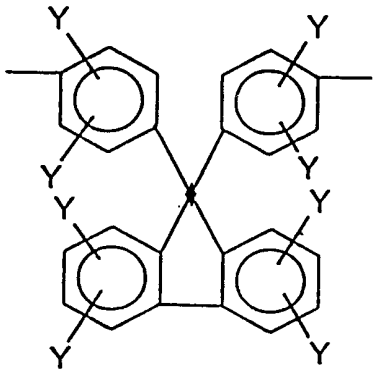
R^2 is hydrogen or hydroxy;

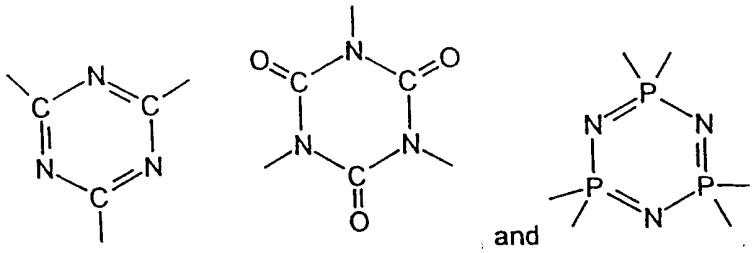
G is selected from the group consisting of



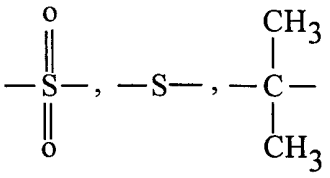








wherein M is selected from the group consisting of a bond,



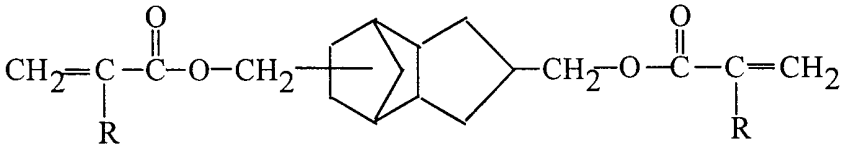
methylene and $-CH_2-S-CH_2-$

R_1, R_3 and R_4 are independently selected from the group consisting of hydrogen and alkyl of 1 to 6 carbon atoms;

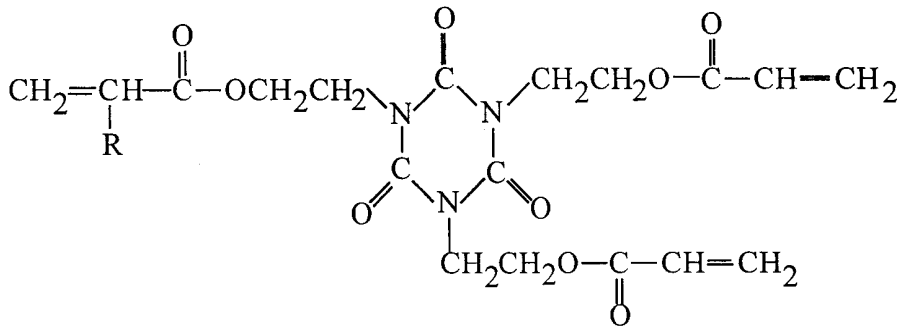
n is an integer having a value of 0 to 10, and

Y is hydrogen or a halogen, preferably fluorine, chlorine or bromine.

7. A photocurable composition according to claim 1 wherein the acrylate monomer is selected from methacrylic acid anhydride; polyalkylene glycol dimethacrylates; polyalkylene glycol diacrylates; compounds of formula:



and compounds of formula

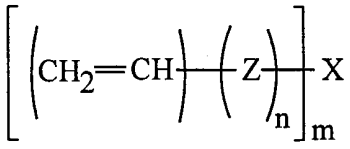


wherein the groups R are independently selected from hydrogen and

methyl.

8. A photocurable composition according to claim 1 wherein the acrylate monomer is present in an amount of from 10 to 40% by weight of the total composition.

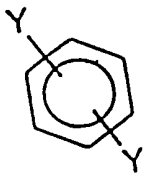
9. A photocurable composition according to claim 1 wherein the monomer containing at least two vinyl groups includes one or more compounds of Formula III



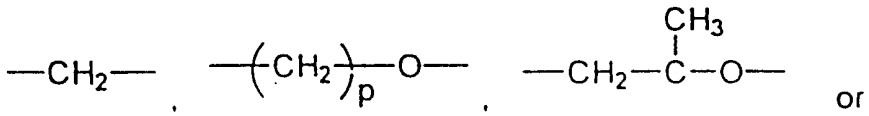
III

wherein

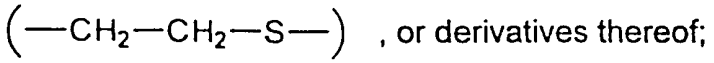
X is



Z is a spacer group selected from one or more of the following



or



, or derivatives thereof;

Y is hydrogen or halogen;

n = 0, 1 or 2;

m = 2 - 6 and

p = 1 - 4.

10. A composition according to claim 1 wherein the compound containing at least two vinyl groups is divinyl benzene.

11. A composition according to claim 1 wherein the photoinitiator includes an acyl phosphine oxide.

12. A composition according to claim 11 wherein the acyl phosphine oxide is a bis-acyl phosphine oxide.

13. A composition according to claim 11 wherein the acylphosphine oxide is selected from the group consisting of bix-(2,4,6-trimethylbenzoyl)phenyl phosphine oxide, bis-(2,6-dimethylbenzoyl)-2,4,4-trimethylpentyl phosphine oxide and ethyl 2,4,6-trimethyl benzoyl phenyl phosphinate.

14. A composition according to claim 11 wherein the acyl phosphoric oxide is present in an amount of from 0.1 to 2% by weight of the total photocurable composition.

15. A composition according to claim 1 wherein the polythiol compound includes at least one compound selected from the group consisting of: Pentaerythritol tetrakis (3-mercapto-propionate) [PTMP], trimethylolpropane tris (3-mercaptopropionate) [TTMP], 4-mercaptomethyl-3,6-dithia-1,8-octanedithiol [MDO], pentaerythritol tetrakis (3-mercaptoacetate) [PTMA], trimethylolpropane tris (3-mercaptoacetate) [TTMA], 4-t-butyl-1,2-benzenedithiol, 2-mercaptoethyl-sulfide, 4,4'-thiodibenzenethiol, benzenedithiol, glycol dimercaptoacetate, glycol dimercaptopropionate ethylene bis(3-mercaptopropionate), polyethylene glycol dimercaptoacetates, polyethylene glycol di(3-mercaptopropionates), 2,5-bis(mercaptomethyl)-1,4-dithiane, 3,6-dithia-1,8-octanedithiol, TGBMA and DMPMA.

16. A composition according to claim 15 wherein the polythiol compound is TGBMA with or without PTMA.

17. A composition according to claim 1 wherein the polythiol compound is present in an amount of from 30 to 50% by weight of the total composition.

18. A composition according to claim 1 further comprising a heat curing agent adapted to induce curing at a temperature in the range of from 80°C to 140°C.

19. A method of preparing a lens comprising:

forming a cast of a photocurable composition according to any one of the preceeding claims and having a thickness in the range of from 1 to 15 mm;

subjecting the composition to UV radiation of wavelength having significant proportion in the wavelength range of from 350 to 450 nm; and

optionally heating the composition to a temperature in the range of from 80° to 140°C in the presence of a heat curing initiator.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/AU00/00349

A. CLASSIFICATION OF SUBJECT MATTER

Int. Cl. [7]: C08F 212/34, 212/36, 220/08, 220/20 G02B 001/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C08F 212/34, 212/36, 220/08, 220/20
G02B 001/04

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
AU: IPC AS ABOVE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
WPAT
JAPIO

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,X	AU 24558/99 A (PPG INDUSTRIES OHIO, INC.) 2 August 1999 Pages 14,15. Examples 1-7	1-19
X	JP 04045401 A (MITSUBISHI GAS CHEM KK) 14 February 1992 Abstract	1-3,8-10,15-19
A	WO 9638486 A (SOLA INTERNATIONAL HOLDINGS LTD) 5 December 1996 Entire Document	1-19

[X] Further documents are listed in the continuation of Box C [X] See patent family annex

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

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Date of the actual completion of the international search	Date of mailing of the international search report
19 May 2000	27 JUN 2000
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA E-mail address: pct@ipaustralia.gov.au Facsimile No. (02) 6285 3929	Authorized officer **N.L. KING** Telephone No : (02) 6283 2150

INTERNATIONAL SEARCH REPORT

International application No.
PCT/AU00/00349

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 02283731 A (TORAY IND INC) 21 November 1990 Abstract	1-19

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/AU00/00349

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report		Patent Family Member					
AU	24558/99	US	5917006	WO	9936450		
WO	9638486	AU	56807/96	BR	9608514	CA	2221939
		CN	1185789	EP	828766	IL	118440
		US	5977276				

END OF ANNEX