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3,488,290

## STABILIZED PHOTOCHROMIC COMPOSITIONS CONTAINING ANTIOXIDANT

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No Drawing. Filed Apr. 15, 1965, Ser. No. 448,274

Int. Cl. G02b 5/20; G03c 1/68

U.S. Cl. 252-300

16 Claims

### ABSTRACT OF THE DISCLOSURE

This invention relates to the provision of light-stabilized photochromic compositions. More particularly, it relates to polymeric photochromic compositions containing normally unstable organic mercuric complexes of diarylthiocarbazonates and certain additive materials capable of stabilizing these mercuric complexes against degradation by light. It also relates to polymeric photochromic compositions containing organic mercuric complexes of diarylthiocarbazonates and said additive materials.

The photochromic composition provided by this invention comprises the combination of certain organo(arylazo)thioformic acid 2-arylhydrazidato] - mercury compounds and certain antioxidant compounds of the hindered phenol type, this type, or phosphite type.

This invention relates to the provision of light-stabilized photochromic composition. More particularly, it relates to polymeric photochromic compositions containing normally unstable organic mercuric complexes of diarylthiocarbazonates and certain additive materials capable of stabilizing these mercuric complexes against degradation by light. It also relates to polymeric photochromic compositions containing organic mercuric complexes of diarylthiocarbazonates and said additive materials.

The term "photochromic composition" is intended to means a composition which is capable of a reversible color change in response to a change in environmental light.

Conventional dyes have substantially constant spectral characteristics regardless of the source or intensity of the incident visible radiation. They are colored because they absorb a portion of the incident visible radiation. If the dyed compositions are transparent, they transmit colored light. If they are non-transparent, they reflect colored light. Colored transparent materials are well-known for such commonplace uses as sunglasses, lenses, goggles, windows and skylights. Colored non-transparent materials are encountered in textile, paper and coating fields. In normal light filter systems, the degree and quality of filtration are fixed when the filter is produced, and, therefore, the color is unvarying thereafter. It is not always desirable, however, to have an inflexible filter since, for many applications, the intensity of incident light goes through variations. In such cases, it may be desirable to have a filter which could vary in automatic response to a variation in light intensity. Non-transparent or reflective materials which would respond to light variations with automatic color and hue change would likewise be useful in obtaining unique camouflage, design and novelty effects. Photochromic materials (i.e., materials capable of changing color as a result of visible light radiation) are known which are capable of satisfying the general demands in this area. However, such materials have quite limited light stability. After several hours of exposure, it is found that much of the photochromic material has been decomposed by the light and the remaining material is not capable of giving as great a color change as was originally obtained. The lack of durability of present

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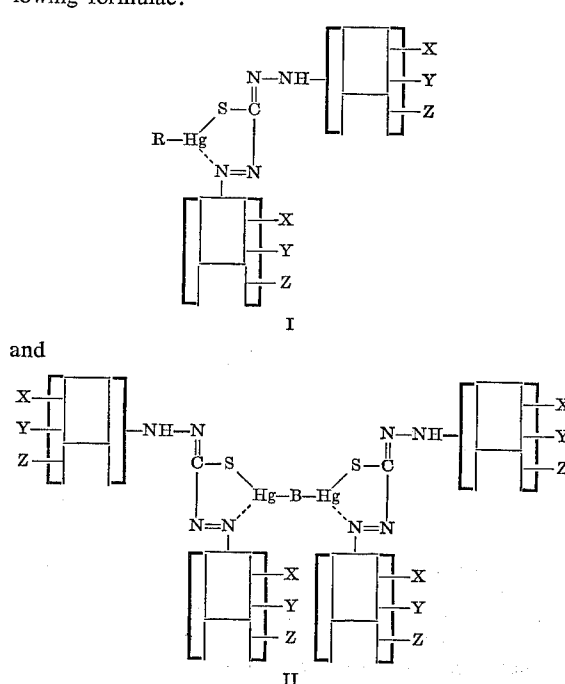
photochromic systems is the major factor for their limited acceptance in applications.

Prior to the present invention, very little had been accomplished toward providing photochromic materials having improved light stability, and it is in this area that the present invention resides.

It is known from commonly assigned application, Ser. No. 369,587, filed May 22, 1964, and now abandoned by John Kazan, Jr., and a continuation-in-part thereof, Ser. No. 421,097, filed Dec. 24, 1964, that light stability of photochromic materials can be materially improved by admixing them with certain substantially colorless materials which have the capacity to absorb light in the ultraviolet range. Mixtures of photochromic materials and ultraviolet absorbers have much greater durability than the photochromic materials per se. The limits of stability to light of these compositions are still not satisfactory for many purposes and, at any rate, the Kazan photochromic compositions still do not possess anywhere near the stability of conventional dye compositions. There is a need for compositions which are photochromic and yet are not encumbered by poor durability in normal fields of application due to lack of light stability.

It is an object of the present invention to provide photochromic compositions which have improved stability to light. Other objects will become apparent from the ensuing description.

It is known that organo-mercuric complexes of the following formulae:



are photochromic materials. In the formulae, X, Y and Z are intended to represent either hydrogen, a lower alkyl radical, hydroxy, a lower alkoxy radical, a halo radical, a nitro radical, an amino radical, a di-lower alkyl-amino radical or a phenylazo radical. The symbol R is intended to represent an alkyl radical of up to eighteen carbon atoms (including cycloalkyl of five or six carbon atoms), an alkenyl radical of 2-10 carbon atoms, an aralkyl radical, especially monocyclic ar(lower alkyl) (such as benzyl or phenethyl), an aryl radical such as phenyl, biphenyl, naphthyl or a heterocyclic aromatic radical having five or six ring members such as furyl and pyridyl. The symbol B is intended to represent a dival-

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ent organic linkage such as an alkylene radical of 2-10 carbon atoms or an arylene radical such as phenylene.

The symbol  $\square$  is intended to represent an aryl radical such as phenyl or naphthyl. The compounds of Formulae I and II may bear inert substituents and still be within the scope of the present invention. For example, R can be substituted by hydroxy, lower alkoxy, di-lower alkyl-amino, carbamoyl, halo, trifluoromethyl, nitro, carboxy, carb-lower alkoxy or lower alkanoyl. The radical B can likewise bear substituents such as those optionally found on radical R.

The present invention is based on the discovery that the combination of a compound of Formula I or Formula II with one or more antioxidants of the type disclosed below leads to a composition having desirable photochromic properties and very greatly enhanced stability. Such compositions, in being capable of performing their photochromic functions for a far greater period than previously known compositions, are suitable in certain end uses where long life is of prime consideration.

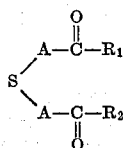
The success of the present invention is surprising for several reasons. In the first place, photochromic dyes are quite unstable and they lack analogy to known dye materials. The problem they present is not solved by mere reference to experience with other dyestuffs. Secondly, conventional dyestuffs are not normally used with antioxidants, since such a need has not been recognized. Thirdly, although some antioxidants are capable of accomplishing the objectives of this invention, other common antioxidants are not useful in this respect. Three such antioxidants found to be incompatible with the mercuric complexes of Formulae I and II are phenothiazine, dibutyl tin dilaurate and the mercuric salt of N,N-di-benzylidithiocarbamic acid.

The antioxidants which may be used in conjunction with the above-described photochromic material are selected from the following classes of compounds:

(A) Antioxidants of the hindered phenol type including those having one to three phenolic groups. By "hindered phenol" is meant a phenol to which in either one or both of the ortho positions to the hydroxyl group is attached at least one bulky group, for example t-butyl. Specific examples of the antioxidants are the following:

2,6-di-tert-butyl-4-methylphenol  
2,4,6-tri-tert-butylphenol  
4,4'-thiobis(6-tert-butyl-m-cresol)  
4,4'-methylenebis(2,6-di-tert-butylphenol)  
4,4'-methylenebis(6-tert-butyl-m-cresol)  
2,6-bis(2-hydroxy-3-t-butyl-5methylbenzyl)-p-cresol  
2,2'-methylenebis(4-ethyl-6-tert-butylphenol)  
2,2'-methylenebis(4,6-dinonylphenol)  
2,2'-methylenebis(4-methyl-6-tert-butylphenol);  
4,4'-butylidenebis(6-tert-butyl-m-cresol)

(B) Antioxidants of the thio type as exemplified by dilauryl thiodipropionate. These may be defined more broadly by the formula:



wherein R<sub>1</sub> and R<sub>2</sub> are substituents selected from the group consisting of —OH,



and —OR<sub>5</sub> wherein R<sub>3</sub> and R<sub>4</sub> are substituents selected from the group consisting of hydrogen and lower alkyl radicals having 1-6 carbon atoms and R<sub>5</sub> is an alkyl

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radical having from 4-18 carbon atoms, and A is an alkylene radical containing from 1-3 carbon atoms.

Illustrative thio compounds within the general Formula III are alkyl esters of 3,3'-thiodipropionic acid, wherein the alkyl group has 4-18 carbon atoms, such as dibutyl, diamyl, dihexyl, dioctyl, diisooctyl, di-2-ethyl-hexyl, dilauryl, distearyl, laurylstearyl, and mixtures of these.

(C) Antioxidants of the phosphite type exemplified by trialkyl phosphites where the alkyl radical has from one to twenty carbon atoms, such as trisodecyl phosphite, trioctyl phosphite; mixed aryl alkyl phosphites such as phenyl didecyl phosphite, diphenyl decyl phosphite; and triaryl phosphites such as tris(p-nonylphenyl) phosphite.

The amount of antioxidant used is between 0.1% and 5.0%, preferably between 0.5% and 2.0%, based on the weight of the substrate.

It has been found that an even greater degree of stability can be imparted to the photochromic dyes if the foregoing antioxidants are employed in conjunction with an ultraviolet absorber. When both additives are used, photochromicity is often retained for periods which are greater than when either additive is used exclusively.

As ultraviolet absorbers which may be added to obtain maximum periods of stability, the following may be mentioned as illustrative:

(A) 2 - hydroxybenzophenones: 2 - hydroxy - 4 - methoxybenzophenone, 2,4 - dihydroxybenzophenone, 2,2'-dihydroxy - 4 - methoxybenzophenone, 2,2',4,4' - tetrahydroxybenzophenone, 2,2' - dihydroxy - 4,4' - dimethoxybenzophenone, 2 - hydroxy - 4 - butoxybenzophenone, 2 - hydroxy - 4 - octyloxybenzophenone, 2 - hydroxy - 4 - dodecyloxybenzophenone, 2,2' - dihydroxy - 4 - octyloxybenzophenone, 4' - chloro - 2 - hydroxy-4-octyloxybenzophenone, etc.

(B) Benzotriazoles: 2 - (2 - hydroxy - 5 - methylphenyl)benzotriazole, 2 - (2 - hydroxy - 5 - octylphenyl)benzotriazole, 2 - (2 - hydroxy - 4 - methoxyphenyl)benzotriazole, etc.

(C) Esters: Phenyl salicylate, tertiary - butylphenyl salicylate, phenyl resorcyate, p-octylphenyl benzoate, bis(p - nonylphenyl)isophthalate, bis(p - nonylphenyl)terephthalate, etc.

(D) Triazines: 2,4,6 - tris(2 - hydroxy - 4 - octyloxyphenyl) - s - triazine, 2 - (2 - hydroxy - 4 - octyloxyphenyl) - 4,6 - dixyl - s - triazine, etc. These compounds are the subject of U.S. Patent 3,118,887, issued Jan. 21, 1964, to Hardy et al.

(E) Benzothiazoles: 2 - (4 - methoxyphenylimino)-3 - ethylbenzothiazole, 2 - phenylimino - 3 - ethylbenzothiazole, etc.

(F) Benzylidenemalonate esters: Diethyl p - methoxybenzylidenemalonate, diethyl o - methoxybenzylidenemalonate, diethyl p - hydroxybenzylidenemalonate, diethyl diphenylmethylenemalonate, etc. This class of ultraviolet absorbers is disclosed in copending application of Susi, Ser. No. 362,182, filed Apr. 23, 1964, now abandoned.

(G) Arylaminoethylenes: N - methyl - p - methoxyanilinomethylenemalonitriles disclosed in U.S. Patent 3,079,366.

The amount of ultraviolet absorber should be between 0.05 and 5.0%, preferably between 0.2 and 2.0%, based on the weight of the polymeric substrate.

The amount of organic dithizonate which is useful for the practice of this invention depends mainly upon the desired effect and the thickness of the substrate. In general, between about 0.001% and 10.0%, preferably between about 0.005% and 5.0%, of the dithizonate based on the weight of the substrate will be employed, but in view of the many fields of application and possible materials which can be used, special circumstances may require that more or less of the photochromic compounds be used.

When compounds of Formula I or II are applied to or incorporated in various materials along with an antioxidant and preferably an ultraviolet absorber, the materials are rendered durably photochromic. For example, polymers containing a measurable quantity of a compound of Formula I or II and a specified amount of antioxidant, change color upon exposure to normal illuminants. The color change is believed to follow from the transformation of the above compounds from a stable form (the less colored form) to a meta-stable form. When the source of radiation is removed, the compound returns from the meta-stable to the stable form.

Either transparent or reflective substrates can be used in the present invention, depending on whether a photochromic light filter or a photochromic reflective surface is desired. The photochromic compound can be combined with the substrate with which it is to be used, by conventional blending, coating, dyeing, milling or molding methods for the given substrate. The choice of application technique is not an aspect of the present invention and will depend on the nature of the substrate, available equipment, and the desired end-use for the product. Among the many materials to which the photochromic compounds can be applied are such natural polymeric materials as cotton, paper and wood. In the case of cotton and paper, a dyeing method may be used. In the case of wood, the photochromic compound may be suspended in a conventional paint or other coating vehicle, along with pigments if desired and applied by spraying or brushing techniques.

The photochromic compounds used in the present invention may be applied to synthetic polymeric materials which may be either solid or liquid, and transparent, translucent or opaque. Among the many usable polymeric compositions are cellulose esters such as cellulose acetate, cellulose butyrate, cellulose nitrate, cellulose acetate butyrate, cellulose propionate, cellulose triacetate, polymeric esters of acrylic acid and methacrylic acid (e.g., methyl methacrylate), polyolefins, polystyrenes, polyvinyl halides, polyvinyl chloride-acetate copolymers, polycarbonates, polyacetals, polyvinyl butyral, polyvinyl acetate, cellulose ethers such as cyanoethylated cellulose and carbamoyl ethylated cellulose, melamine resins, polyacrylonitrile, polyester resins, etc.

Along with the photochromic compounds of Formula I and Formula II, there may be added to the substrate other materials such as dyestuffs which are not themselves photochromic, to give unique color effects. For example, a blue dyestuff and a mercuric dithizonate give a green color mixture before exposure to light. As the photochromic mixture becomes colored (upon exposure), the color changes to gray or blue. Without the non-photochromic dye, a change in color from yellow to gray to blue would be observed. The addition of such auxiliary dyestuffs is entirely optional and does not form a necessary part of the present invention. Suitable dyes are those which are soluble in or compatible with the polymeric substrate. Examples include the class of dyes commonly known as Solvent Dyes and so named in the Color Index. These dyes include C.I. Solvent Yellow 1, 14, 15, 17, 19, 23, etc.; C.I. Solvent Orange 6, 9, 10, 34, etc.; C.I. Solvent Red 7, 9, 10, 12, 14, 16, etc.; C.I. Solvent Violet 4, 5, 6, 11, 12, 13, etc.; C.I. Solvent Blue 1, 10, 16, 18, 19, 21, 38, 59, etc.; C.I. Solvent Green 3, 11, etc.; C.I. Solvent Brown 5, 8, 15, etc.; C.I. Solvent Black 1, 2, 6, 15, etc.; Disperse Blue 3, 7, 77, 78, etc. and Disperse Red 9, etc.

The following examples, in which parts and percentages are by weight, are presented to further illustrate the present invention.

#### EXAMPLE 1

Five plastic compositions were prepared by hot milling molding grades of cellulose acetate-butyrate for five minutes with methyl-[(phenylazo)thioformic acid 2-

phenylhydrazidato]mercury, 2,2'-dihydroxy-4-methoxybenzophenone and 2,2'-methylenebis(4-methyl-6-tertiary-butylphenol) (Antioxidant A) or 2,6-di-tertiary-butyl-p-cresol (Antioxidant B). The formulations are shown in Table I. The milled compositions were then compression-molded into sheets 0.015 to 0.020" thick.

The molded sheets were exposed to artificial light in a Fade-Ometer and to sunlight. The percent of the original amount of mercury complex remaining after the exposure was determined spectrophotometrically. The results are shown in Table II and Table III.

TABLE I

Components	Parts				
	I	II	III	IV	V
Cellulose acetate-butyrate	100	100	100	100	100
Mercuric complex	0.04	0.04	0.04	0.04	0.04
U.V. absorber			0.3	0.3	0.3
Antioxidant A		1.0		1.0	
Antioxidant B			1.0		

TABLE II

Hours exposure:	Percent complex remaining, Fade-Ometer				
	I	II	III	IV	V
10	15	68	83	88	58
20		40	51	68	13
30		28	29	53	
40		17	14	39	

TABLE III

Hours exposure:	Percent complex remaining, sunlight				
	I	II	III	IV	V
5	67	83	87	91	85
10	37	63	72	84	69
15	13	42	54	73	53
20		33	43	67	41
25		23	30	56	25

The data in the above tables show that 1.0% of 2,2'-methylenebis(4-methyl-6-tertiary-butylphenol) stabilizes the mercuric complex against U.V. radiation from sunlight and from artificial light sources.

The data also show that the combination of Antioxidant A and the U.V. absorber gives better short- and long-term protection than when either is taken alone. It also shows that the combination of Antioxidant B with a U.V. absorber is more effective than the U.V. absorber by itself under artificial lighting, though not under natural light.

#### EXAMPLE 2

Plastic compositions were prepared by hot milling cellulose acetate-butyrate for five minutes with 0.04% of methyl[(phenylazo)thioformic acid 2-phenylhydrazidato]mercury, 0.3% of 2,2'-dihydroxy-4-methoxybenzophenone and 1.0% of an antioxidant. Control compositions were also prepared with the antioxidant omitted.

#### Antioxidants

- (A) 2,2'-methylenebis(4-methyl-6-tertiary-butylphenol)
- (B) 2,6-di-tertiary-butyl-p-cresol
- (C) Phenyl didecyl phosphite
- (D) Mixture of 7.5-10% of 2,4-dinonylphenol, 40-42.5% 2,2'-methylenebis(4,6-dinonylphenol) and 50% of phenyl didecyl phosphite
- (E) Didodecyl ester of 3,3'-thiodipropionic acid
- (F) 4,4'-(n-propylmethylene)bis(2-tertiary-butyl-6-methylphenol)
- (G) 4,4'-thiobis(2-tertiary-butyl-5-methylphenol)

The milled compositions were compression-molded into sheets 0.015" to 0.020" thick. The molded sheets were exposed in a Fade-Ometer and to sunlight. The percent of the original amount of mercury complex remain-

ing after the exposure was determined spectrophotometrically. The results are shown in Tables IV and V.

TABLE IV

Antioxidant:	Percent complex remaining, sunlight exposure			
	5 hours	10 hours	15 hours	20 hours
A.....	90	86	79	72
D.....	86	77	68	61
G.....	89	81	78	67
None.....	82	71	59	52

TABLE V

Antioxidant:	Percent complex remaining, Fade-Ometer exposure			
	5 hours	10 hours	15 hours	20 hours
A.....	89	79	71	62
B.....	87	70	56	46
C.....	94	84	75	58
D.....	87	72	61	42
F.....	84	71	63	53
G.....	85	73	66	54
None.....	76	56	42	33

Of those described, Antioxidant A is preferred since it provides the highest level of stability. The other antioxidants are effective to a lesser degree, but they provide improved stability as against the control composition. Other antioxidants such as phenothiazine, dibutyl tin dilaurate and mercury N,N-dibenzyl dithiocarbamic acid, do not provide significant improvement in stability of the photochromic compound.

## EXAMPLE 3

Two plastic compositions were prepared by hot milling cellulose acetate-butyrate for five minutes with 0.04% of methyl[(phenylazothioformic acid 2-phenylhydrazidato)mercury and 1.0% of an antioxidant. A plastic composition was also prepared with the antioxidant omitted.

## Antioxidant

- (A) 2,2'-methylenebis(4-methyl-6-tertiary-butylphenol)  
(B) Phenyl didecyl phosphite

The milled compositions were then compression-molded into sheets 0.015" to 0.020" thick. The molded sheets were exposed in a xenon-type of "Weather-Ometer." The percent of the original amount of mercury complex remaining after the exposure was determined spectrophotometrically. The results are shown in Table VI.

TABLE VI

Antioxidant:	Percent complex remaining, Weather-Ometer			
	5 hours	10 hours	15 hours	20 hours
A.....	91	80	64	51
B.....	81	63	41	23
None.....	78	58	34	18

Polymeric compositions of like properties are prepared from polystyrene, poly(vinyl chloride), poly(methyl methacrylate), poly(vinyl butyral) and poly(ethylene adipate).

## EXAMPLE 4

Two plastic compositions were prepared by hot milling cellulose acetate-butyrate for five minutes with 0.04% methyl[(phenylazothioformic acid 2-phenylhydrazidato)mercury, 0.3% ultraviolet absorber and 1.0% 2,2'-methylene-bis(4-methyl-6-tertiary-butylphenol). Similar plastic compositions were prepared with the antioxidant omitted.

## Ultraviolet absorber

- (A) 2,2'-dihydroxy-4-methoxybenzophenone  
(B) Carbonyl cyanide m-methoxyphenylhydrazone

The milled compositions were then compression-molded into sheets 0.015" to 0.020" thick. The molded sheets

were exposed in a xenon-type of "Weather-Ometer" for 20 hours. The percent of the original amount of mercury complex remaining after the exposure was determined spectrophotometrically. The results are shown in Table VII.

TABLE VII

U.V. Absorber:	Antioxidant	Percent complex remaining, 20 hours exposure in Weather-Ometer
A.....	No.....	54
A.....	Yes.....	64
B.....	No.....	64
B.....	Yes.....	79

This example shows the improvement in stability which can be obtained by use of combination of a U.V. absorber and an antioxidant. Similar results are obtained by use of other U.V. absorbers such as those listed in the specification.

It is feasible to use combinations of two or more antioxidants, either alone or with a U.V. absorber.

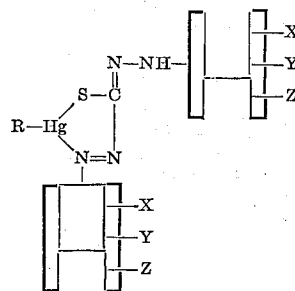
## EXAMPLE 5

The following stabilizer combinations are prepared and used in the procedure of Example 1.

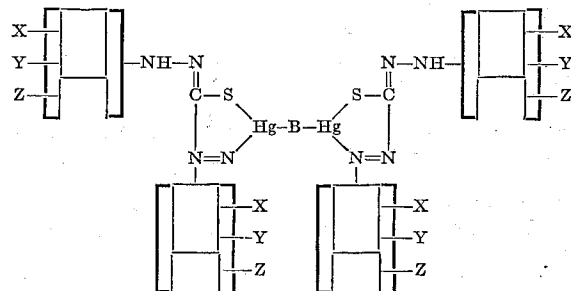
- (1) 2,4,6-tri-tert-butylphenol (0.5 part) and 2-(2-hydroxy-5-methylphenyl)benzotriazole (0.2 part)
- (2) Dilauryl 3,3'-thiodipropionate (0.5 part) and phenyl salicylate (0.2 part)
- (3) Phenyl didecyl phosphite (0.5 part) and 2,4,6-tris-(2-hydroxy-4-octyloxyphenyl)-s-triazine (0.5 part)
- (4) 2,6-di-tert-butyl-4-methylphenol (0.5 part) and diethyl p-methoxybenzylidenemalonate (0.2 part)
- (5) 2,4,6-tri-tert-butylphenol (0.5 part) and N-methyl-p-methoxyanilinomethylenemalononitrile (0.2 part).

We claim:

1. A photochromic composition consisting essentially of:
  - (a) a photochromic compound of the formulae:



and

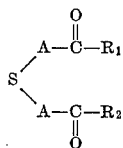


wherein X, Y and Z are individually selected from the group consisting of hydrogen, lower alkyl, hydroxy, lower alkoxy, halo, nitro, amino, di-lower alkylamino and phenylazo; R is an alkyl of up to eighteen carbons, alkenyl of 2-10 carbons, monocyclic ar(lower alkyl) and aromatic of up to two

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rings of 5-6 carbons; B is a member selected from the group consisting of alkylene of 2-10 carbons and monocyclic arylene; and  $\square$  is an aryl radical of up to six membered rings; and

- (b) 0.5-50 parts per part of photochromic compound of an antioxidant selected from the group consisting of hindered phenols, tri(hydrocarbon)phosphites and thio compounds of the formula:



wherein  $\text{R}_1$  and  $\text{R}_2$  are individually selected from the group consisting of  $-\text{OH}$ ,



and  $-\text{OR}_5$  wherein  $\text{R}_3$  and  $\text{R}_4$  are substituents selected from the group consisting of hydrogen and lower alkyl radicals having 1-6 carbon atoms and  $\text{R}_5$  is an alkyl radical having from 4-18 carbon atoms, and A is an alkylene radical containing from 1-3 carbon atoms.

2. The photochromic composition of claim 1 wherein the antioxidant is a hindered phenol.

3. The photochromic composition of claim 1 wherein the antioxidant is 2,6-di-tert-butyl-4-methylphenol.

4. The photochromic composition of claim 1 wherein the antioxidant is 2,2'-methylenebis(4-methyl-6-tertiary-butylphenol).

5. The photochromic composition of claim 1 containing 0.005 to 5000 parts per part of photochromic compound of an ultraviolet absorber selected from the group consisting of 2-hydroxy benzophenones, 2-(2-hydroxyphenyl)benzotriazoles, aryl esters of phenyl carboxylic acids, triaryl triazines wherein one aryl radical is substituted by a 2-hydroxy group, 2-(phenylimino)-3-lower alkyl benzothiazoles, benzylidenemalononic acid esters and arylamino ethylenes.

6. The photochromic composition of claim 5 wherein the ultraviolet absorber is 2,2'-dihydroxy-4-methoxybenzophenone.

7. The photochromic composition of claim 5 wherein the ultraviolet absorber is 2-(2-hydroxy-5-methylphenyl)benzotriazole.

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8. A stabilized photochromic polymeric composition consisting essentially of a polymeric material selected from the group consisting of cellulose ethers, polyolefins, polystyrenes, poly(vinyl halides), polycarbonates, polyacetals, poly(vinyl butyral), melamines, polyacrylonitrile and polyesters; and the stabilized photochromic composition of claim 1 in sufficient concentration to provide 0.001 to 10.0% by weight of the photochromic compound.

9. The photochromic composition of claim 8 wherein the polymer is cellulose acetate butyrate.

10. The composition of claim 8 wherein the antioxidant is 2,2'-methylenebis(4-methyl-6-tertiary-butylphenol).

11. The composition of claim 8 wherein the antioxidant is 2,6-di-tert-butyl-4-methylphenol.

12. A stabilized photochromic polymeric composition consisting essentially of:

- (a) a polymer selected from the group consisting of cellulose ethers, polyolefins, polystyrenes, poly(vinyl halides), polycarbonates, polyacetals, poly(vinylbutyral), poly(vinylacetate), melamines, polyacrylonitrile and polyesters; and

- (b) the stabilized photochromic mixture of claim 5 in sufficient quantity to provide the photochromic compound in a concentration of 0.001 to 10 percent by weight.

13. The composition of claim 12 wherein the ultraviolet absorber is 2,2'-dihydroxy-4-methoxybenzophenone.

14. The composition of claim 12 wherein the antioxidant is 2,6-di-tert-butyl-4-methylphenol.

15. The composition of claim 12 wherein the antioxidant is 2,2'-methylenebis(4-methyl-6-tertiarybutylphenol).

16. The composition of claim 12 wherein the polymer is cellulose acetate-butyrate.

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RICHARD D. LOVERING, Primary Examiner

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96-90; 260-45.75; 350-160