TO: KSI/Scientific & Technical Information Division  
Attn: Miss Winnie M. Morgan

FROM: GP/Office of Assistant General Counsel for Patent Matters

SUBJECT: Announcement of NASA-Owned U.S. Patents in STAR

In accordance with the procedures agreed upon by Code GP and Code KSI, the attached NASA-owned U.S. Patent is being forwarded for abstracting and announcement in NASA STAR.

The following information is provided:

U.S. Patent No.: 3,803,090

Government or Corporate Employee: U.S. Government

Supplementary Corporate Source (if applicable): 

NASA Patent Case No.: ARC-10,592-1

NOTE - If this patent covers an invention made by a corporate employee of a NASA Contractor, the following is applicable:

YES [X]  NO [ ]

Pursuant to Section 305(a) of the National Aeronautics and Space Act, the name of the Administrator of NASA appears on the first page of the patent; however, the name of the actual inventor (author) appears at the heading of column No. 1 of the Specification, following the words "...with respect to an invention of ..."

Bonnie L. Woerner

Enclosure
THERMOGRAVIMETRIC ANALYSIS OF REPRESENTATIVE POLYMERS

- POLY-2,2'-DM-5,5'-(DPS)-BZ
- POLY-2,2'-DH-5,5'-(DPS)-BZ
- POLY-4,4'-(DPS)-BZ
- POLY-2,2'-DH-6,6'-(DPS)-NZ
- POLY-DIMETHYLSILOXANE
- PET

 percent weight retained

0 200 400 600 800
0 40 60 70 80 90 100
scale change

TEMPERATURE, °C

FIG. 1

3,803,090
COMPARISON OF POLYMER PHOTOSTABILITY BY ESR

VACUUM

150 torr O₂

INITIAL RATE OF PHOTO-INDUCED RADICAL PRODUCTION

1000

100

10

1

0.1

COMMERCIAL POLYMERS

POLYDIPHENYLZILAOXYARYLIZINES

POLY-4,4’-(DPS)-BZ

POLY-2,2-DH-6,6’-(DPS)-NZ

POLY-2,2-DH-5,5’-(DPS)-NZ

POLY-2,2-DH-7,7’-(DPS)-NZ

POLY-2,2-DH-4,4’-(DPS)-DBDP

POLY-4,4’-(DPS)-DBDP

POLY-4,4’-(DPS)-BZ

POLY-4,4’-(DPS)-BZ

MYLAR, PURIFIED

MYLAR, PURIFIED

MYLAR, PURIFIED

MYLAR, C GRADE

POLYVINYLIDENE FLUORIDE

POLYSTYRENE, PURIFIED

POLYPROPYLENE

PVC

FIG. 2.
ULTRAVIOLET AND THERMALLY STABLE POLYMER COMPOSITIONS

Filed Jan. 5, 1973

FIG. 3.
ULTRAVIOLET AND THERMALLY STABLE POLYMER COMPOSITIONS

Ronald F. Reinsch, Palo Alto, and Hermilo R. Gloria, San Jose, Calif., assignors to the United States of America as represented by the Administrator of the National Aeronautics and Space Administration

Filed Jan. 5, 1973, Ser. No. 321,179

U.S. Cl. 260-465

ABSTRACT OF THE DISCLOSURE

A new class of polymers is provided, namely, poly(diaryl siloxyl) aryazines. These novel polymers have a basic chemical composition which has the property of stabilizing the optical and physical properties of the polymer against the degradative effect of ultraviolet light and high temperatures. This stabilization occurs at wavelengths including those shorter than found on the surface of the earth and in the absence or presence of oxygen, making the polymers of the present invention useful for high performance coating applications in extraterrestrial space as well as similar applications in terrestrial service. The invention also provides novel aromatic azines which are useful in the preparation of polymers such as those of the present invention.

The invention described herein was made by employees of the United States Government and may be manufactured and used by or for the Government for governmental purposes without the payment of any royalties thereon or therefor.

BACKGROUND OF THE INVENTION

Field of the invention

Plastic materials having stable properties under conditions of ultraviolet light and high temperatures in the presence or absence of oxidizing atmospheres.

Description of the prior art

Heretofore, in the preparation of ultraviolet stable polymer films and coatings, a conventional practice has been to mix within the matrix or to coat the surface of the polymer with an ultraviolet absorbing compound, or opaque filler such as carbon black, talc, clay or a pigment.

Although this invention is not predicated on its theory of operation, it is believed that the poly(diaryl siloxyl) aryazines of the present invention derive their photostability from the contribution of the repeating units to the rapid deactivation of electronically excited states, a process which is believed to allow the polymer to relax to the ground state at a rate faster than any bond scission process.

Many of the protective compounds of the prior art are low molecular weight substances which are incorporated in the polymer melt, coated on the surface of the polymeric structure or dissolved in the casting solvent. While many of the substances involved are effective when used in relatively thick layers or high concentrations, the results obtained therewith are far from optimum because the incorporation of 0.01 to 10% by weight of stabilizer often adversely affects the physical, chemical and optical properties for which the polymeric materials were initially chosen. Moreover, conventional techniques of protection from ultraviolet have been tailored to terrestrial conditions and are often deficient in the space environment. For example, the commercially employed benzophenone derivatives actually become sensitizers for the photodegradation of polymers in vacuum at wavelengths below 3000Å.

A further deficiency of commercial ultraviolet absorbers is that as additives they show a tendency to be leached out of plastics by solvents, to migrate off the surface or to be lost by evaporation during the hot molding operations and other thermal processing to which plastics may be subjected. This loss of absorber is accompanied by a loss of protection against photodegradation.

The use of copolymerizable groups as the ultraviolet stabilizer has also been suggested as a means of covalently binding the stabilizer in the polymer chain. However, in practice only a small fraction of the copolymerizable stabilizer is actually incorporated in the polymer chain, leaving the unreacted stabilizer to be leached out in the same manner as the additives mentioned above. Since the randomly incorporated stabilizer constitutes only a small fraction of the polymer chain, no protection is afforded to long stretches of the chain that are devoid of stabilizer groups.

Many of the commercial polymers that have a degree of photostability are not soluble in common solvents and therefore cannot be used economically in paints, coatings or films.

Heretofore, 2,2'-dihydroxynaphthalazine (2,2'-DHNZ) has been described [N. Kaisianpaller & D. Du Hon, Appl. Optics 3,287 (1964)] as a yellow organic phosphor with constant fluorescent efficiency between 1000 and 3500 Å, but there has been no any recommendation for the use of such compounds as a repeating unit in an alternating copolymer with diphenylsiloxane units to enhance the ultraviolet stability of a polymer.

Alternating copolymers composed of arylsiloxy and diphenylsiloxy repeating units have been prepared by the melt condensation of arylols and bis-(anilino)diphenylsilane, [J. E. Curry & J. D. Byrd, J. Appl. Polymer Sci., 9, 295 (1965)] although the resulting polymers displayed good thermal stability up to 300°C. and coatings cast from tetrahydrofuran had considerable ultraviolet stability in vacuum; they did not contain the diazine linkage, \((-N=N-N=N-\) \) as the basic repeating unit which is essential to achieve the objectives of the present invention.

Sulfonated derivatives of 2,2'-dihydroxynaphthalazine have been described [U.S. Pat. No. 2,818,432] as useful fluorescent dyes for animal fibers or polymides. However, the resulting dyed polymers did not contain a diazine linkage as the basic repeating unit of the polymer backbone because the 2,2'-dihydroxynaphthalazine group was attached to the polymer backbone as a pendant side-chain through the sulfonic acid group.

Polymers derived from hydrazine have been described [U.S. Pat. No. 2,957,852] as elastomers that exhibit improved stability to ultraviolet light. However, the repeating units of these polymers were linked through carbonyl groups where X and X' represent hydrogen or a monovalent organic radical. These elastomers, although derived from hydrazine, do not contain the diazine-linkage essential to the present invention.

Heretofore, polymers based on the reaction of the hydrazine class of compounds and halosiloxanes have been described [U.S. Pat. No. 2,877,201] as being water repellent compounds useful for application to aircraft windshields. However, since this reaction is limited to linear aliphatic hydrazine compounds having at least one functional hydrogen atom attached to each nitrogen atom, it was not possible to prepare by this method polymers which have the diazine linkage as the repeating unit in the polymer backbone as described in the present invention.
3 SUMMARY OF THE INVENTION

This invention relates to a new class of polymers whose basic chemical composition has the property of stabilizing the optical and physical properties of the polymer against the degradative effect of ultraviolet light and high temperatures. More particularly, this stabilization also occurs at wavelengths shorter than those found on the surface of the earth and in the absence or presence of oxygen; making this invention useful for high performance coating applications in extraterrestrial space as well as in terrestrial use.

The polymers are particularly useful for coating metal or glass surfaces employed on the exterior panels of spacecraft, improved protective coatings for solar cells, photostable paints for stabilizing inflatable structures, and numerous other structural applications where the combination of photostability below 3000 A, and thermal stability in the region of 480° F. (250° C.) is required.

An object of the present invention is to provide a novel and useful, high temperature stable film having desirable thermal and optical properties that can be conveniently prepared from economical starting materials, e.g., hydroxylated arylazines and diphenyldihalosilanes.

Another object is to provide an efficient and economical process for preparing these ultraviolet and thermally stable polymers, which is adaptable to conventional, batch-type, melt polymerization processes and which can be carried out with existing plants and equipment.

A further object is to provide an ultraviolet stable polymer which avoids the use of sacrificial additives to achieve protection of the host polymer from photodegradation.

A still further object of this invention is to provide a plastic film which can be formed by casting from volatile, economical organic solvents and which can be cast with a doctor knife and dried at room temperature in a forced draft enclosure.

Another object of the invention is to provide a transparent, plastic layer that has excellent adhesion to glass. This property of bonding to glass as well as ultraviolet and thermal stability make the poly(diphenylsiloxy) arylazines useful compounds for laminating aircraft windshields or spacecraft windows. The bonding agent does not support combustion. Purified poly(diphenylsiloxy) arylazines melt at about 170° C., and resolidify when cooled to 150° C. The bonding agent by not suffering significant photodegradation when subjected to ultraviolet radiation in the wavelength region below 3000 A.

Another object of the invention is to provide a polymer that does not support combustion. Purified poly(diphenylsiloxy) arylazines melt at about 170° C., and resolidify when cooled to 150° C. The bonding agent by not suffering significant photodegradation when subjected to ultraviolet radiation in the wavelength region below 3000 A.

Another object of the invention is to provide a polymer which avoids the use of sacrificial additives to achieve protection of the host polymer from photodegradation.

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was open at both ends to allow the passage of a small droxy-U'-naphthalazine (2,2',6,6'-THNZ); 2,2',7,7'-tetradydroxy-1',1'-naphthalazine (2,2',7,7'-THNZ); and 2,2'-dimethoxy-6,6'-dihydroxy-1',1'-naphthalazine (2,2'-DM,6,6'-DHNZ).

The aryl azines are prepared by methods well known to those skilled in the art and consists of causing an ethanolic solution of hydrazine sulfate in a nitrogen atmosphere. Although any hydroxy substituted aromatic aldehyde may be suitable for the preparation of the required aryl azine monomer, a series of new aldehydes is 2,2'-dihydroxy-5,5'-dimethoxybenzaldehyde and 2,2'-dimethoxy-5,5'-dihydroxybenzaldehyde. Aromatic aldehydes that have a methoxy group in the 2(ortho) position of the benzene ring and a hydroxy group in one of the remaining unsubstituted positions may be prepared by the method of F. M. Dean, [Tetrahedron Letters 4153 (1966)], by allowing an aromatic aldehyde containing two methoxy groups to react with boron trichloride. The methoxy group in the 2 position is not cleaved but the remaining ether group is selectively cleaved to yield a hydroxy group.

Examples of diaryl- or dialkyl-arylalkyl-silylamines that are suitable for use in the present invention include: bis(aniline) diphenylsilane, bis(aniline) dimethyldisilane and bis(aniline) phenylmethylsilane. These compounds may be prepared by allowing aniline to react with the appropriate diaryl or arylalkyldichlorosilane in the presence of 2,3,6-trimethylpyridine according to the method of Curry et al. [J. Appl. Polymer Sci. 9, 295 (1965)]. The diaryl or dialkyl dichlorosilanes are commercially available.

The following non-limiting examples illustrate preferred embodiments of the invention.

**EXAMPLE 1**

Preparation of poly(4,4'-diphenylsiloxy) benzalazine (poly-4,4'-(DPS)-BZ)

The apparatus used in these melt condensation reactions was an all-glass system having polymerization tubes 2.5 cm. in diameter and 10.5 cm. long. The reaction vessel was equipped with two concentric tubes which extend to the bottom of the polymerization tube. The outer tube was open at both ends to allow the passage of a small droxy-U'-naphthalazine. The ratio of monomers that had been previously purified by methanol precipitation, was placed between two sheets of clean glass. Upon heating the glass sandwich to 190° C. and maintaining the laminate at 190° C. under light pressure for 30 minutes, the two glass sheets were bonded together.

Poly(4,4'-diphenylsiloxy) benzalazine poly-4,4'-(DPS)-BZ polymers were also prepared under a variety of conditions, but following the general outline given above. The reaction time was varied from one hour to 24 hours and the temperature varied from 170° C. to 230° C. The monomer ratio of the arylazine to the silylamine was varied from 1:0.8 to 2:3 and in all cases polymers satisfactory for the purposes of the present invention were obtained.

The following polymers were made by the process of Example 1.

**Example 2:** Poly2,2'-dimethoxy-5,5'-(diphenylsiloxy) benzalazine, poly-2,2'-DM,5,5'-(DPS)-BZ.

**Example 3:** Poly-4,4'-diphenylsiloxyl) - dibenzidine - p-phenylene diamine, poly-4,4'-(DPS)-DBPD.

**Example 4:** Poly-2,2'-diethylene-6,6'-(diphenylsiloxyl) naphthalazine, poly-2,2'-DH,6,6'-(DPS)-NZ.

**Example 5:** Poly-2,2'- diethylene-7,7'- (diphenylsiloxyl) naphthalazine, poly-2,2'-DH,7,7'-(DPS)-NZ.

**Example 6:** Poly-2,2'-diethylene-5,5'-(diphenylsiloxyl) benzalazine, poly-2,2'-DH,5,5'-(DPS)-BZ.

The infrared spectra of all polymers were obtained. They exhibit the 1610 cm⁻¹ absorption band attributed to the C=N linkage while the bands between 815 and 1050 cm⁻¹ are assigned to the C—H out-of-plane deformation in the 1,4-disubstituted aromatic rings. In addition, the infrared spectrum contains all of the bands between 690 and 720 cm⁻¹ attributed to the monosubstituted benzene rings attached to silicon while the bands observed at 1429 and 1125 cm⁻¹ may be assigned to the silicon phenyl linkage. The presence of the bands at 1250 and 1050 cm⁻¹ attributed to the silicomethyl and Si—O—C linkages, respectively, confirm that the polymers have the general formula indicated.

The molecular weight of the polymers was found to depend on temperature, reaction time and the ratio of monomers. In general, the formation of low molecular weight products was avoided by maintaining the reaction temperature below the decomposition point of the arylazine and by keeping the ratio of silylamine to arylazine at about 3:2. When the reaction temperatures were greater than 200° C., low molecular weight, insoluble copolymers were obtained. Upon lowering the reaction temperature from 230° C. and maintaining the same ratio of silylamine to arylazine in the two experiments, a material was obtained which was soluble in tetrahydrofuran and had a molecular weight which was an order-of-magnitude greater than that obtained by reaction at the higher temperatures. Thus, the preferred ceiling temperature was established at 195° C.

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of weight loss using a thermogravimetric balance and the results are plotted in FIG. 1. The photostability of the polymers was determined by ultraviolet irradiation of the polymers and monomers in the optical cavity of an electron spin resonance (ESR) spectrometer. The initial rates of photo-induced radical production, in arbitrary units of radicals per minute, were obtained by measuring the ESR signal strength and are plotted in FIG. 2 for the polymers and in FIG. 3 for the monomers. For purposes of comparison, the rates of radical production are used as a criterion of ultraviolet stability. A comparison of the thermogravimetric, FIG. 1, and electron spin resonance analysis, FIG. 2, shows that polymers that do not possess ortho hydroxyl groups exhibit the greatest thermal and photochemical stability. For example, poly-4,4'(diphenylsiloxyl)benzalazine poly-4,4'(DPS)-BZ shows little weight loss at 300° C. and has the lowest rate of photo-induced radical production of the arylazine polymers. The nature of the aromatic ring appears to have little influence on the photostabilities of these molecules. In general, as shown by comparing FIGS. 2 and 3, the structural features of the monomeric azines were transferred to the corresponding polymers because the polymers appear to be about a factor of two more ultraviolet stable than the related monomer, and the polymers are all more ultraviolet stable than a typical silane, diphenyldimethylsilane.

All of the polymers exhibit varying degrees of photostability when irradiated at 2537 and 1848 A. as displayed in FIG. 2. Since the polymers degrade by different mechanisms, a straightforward comparison is often difficult to make, however, using the initial rate of photo-induced radical production as a criterion of photostability, it is seen that the poly(diphenylsiloxyl) arylazines are as ultraviolet stable as polytetrafluoroethylene, Teflon, or polyethylene terephthalate Pet (Mylar), and are more stable than many conventional polymers currently in widespread use. The presence of oxygen at 1 torr enhanced the initial rate of radical production by approximately a factor of ten in the examples of poly(4,4'-diphenylsiloxyl) benzalazine poly-4,4'-(DPS)-BZ and polyethylene terephthalate Pet (Mylar).

We claim:

1. As a new composition of matter a polymer of the formula:

\[
\begin{align*}
\text{O} & - \text{R} - \text{C} = \text{N} - (\text{R'}) - \text{N} = \text{C} - \text{R} - \text{O} - \text{Si} - \\
& \quad \text{X} & \text{Y} & \text{L} & \text{X} & \text{Y}
\end{align*}
\]

wherein \( n \) represents an integer ranging from 3 to 500; \( R \) represents a substituted aromatic group; \( X \) stands for a hydrogen atom, an alkyl radical or an aryl radical; \( Y \) represents a phenyl or methyl radical; and \( R' \) is either a phenyl group or a single covalent linkage.

2. The composition of claim 1 wherein the value of \( n \) is preferably from 15 to 90.

3. The composition according to claim 1 wherein \( n \) ranges from 15–90; \( R \) is

\[
\begin{align*}
& \quad \text{O} & \quad \text{X} & \quad \text{Y} & \quad \text{L} & \quad \text{X} & \quad \text{Y} \\
& \quad \text{HO} & \quad \text{X} & \quad \text{Y} & \quad \text{OH} & \quad \text{X} & \quad \text{Y}
\end{align*}
\]

and \( R' \) is a covalent linkage.

4. The composition according to claim 1 wherein \( n \) ranges from 15–90; \( R \) is

\[
\begin{align*}
& \quad \text{O} & \quad \text{X} & \quad \text{Y} & \quad \text{L} & \quad \text{X} & \quad \text{Y} \\
& \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y}
\end{align*}
\]

and \( R' \) is a covalent linkage.

5. The composition according to claim 1 wherein \( n \) ranges from 15 to 90; \( R \) is

\[
\begin{align*}
& \quad \text{O} & \quad \text{X} & \quad \text{Y} & \quad \text{L} & \quad \text{X} & \quad \text{Y} \\
& \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y}
\end{align*}
\]

and \( R' \) is a covalent linkage.

6. The composition according to claim 1 wherein \( n \) ranges from 15 to 90; \( R \) is

\[
\begin{align*}
& \quad \text{O} & \quad \text{X} & \quad \text{Y} & \quad \text{L} & \quad \text{X} & \quad \text{Y} \\
& \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y}
\end{align*}
\]

and \( R' \) is a covalent linkage.

7. The composition according to claim 1 wherein \( n \) ranges from 15–90; \( R \) is

\[
\begin{align*}
& \quad \text{O} & \quad \text{X} & \quad \text{Y} & \quad \text{L} & \quad \text{X} & \quad \text{Y} \\
& \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y}
\end{align*}
\]

and \( R' \) is a covalent linkage.

8. The composition according to claim 1 wherein \( n \) ranges from 15–90; \( R \) is

\[
\begin{align*}
& \quad \text{O} & \quad \text{X} & \quad \text{Y} & \quad \text{L} & \quad \text{X} & \quad \text{Y} \\
& \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y}
\end{align*}
\]

and \( R' \) is a covalent linkage.

9. The process of making a polymer having the formula:

\[
\begin{align*}
& \quad \text{O} & \quad \text{X} & \quad \text{Y} & \quad \text{L} & \quad \text{X} & \quad \text{Y} \\
& \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y} & \quad \text{X} & \quad \text{Y}
\end{align*}
\]

wherein \( n \) represents an integer ranging from 3 to 500; \( R \) represents a substituted aromatic group; \( X \) stands for a hydrogen atom, an alkyl radical or an aryl radical; \( Y \) represents a phenyl or methyl radical; and \( R' \) is either a
9. The process of claim 9 wherein the molar ratio of the arylazine to silylamine is 2:3.

10. The process of claim 9 wherein the temperature is not over 195° C.