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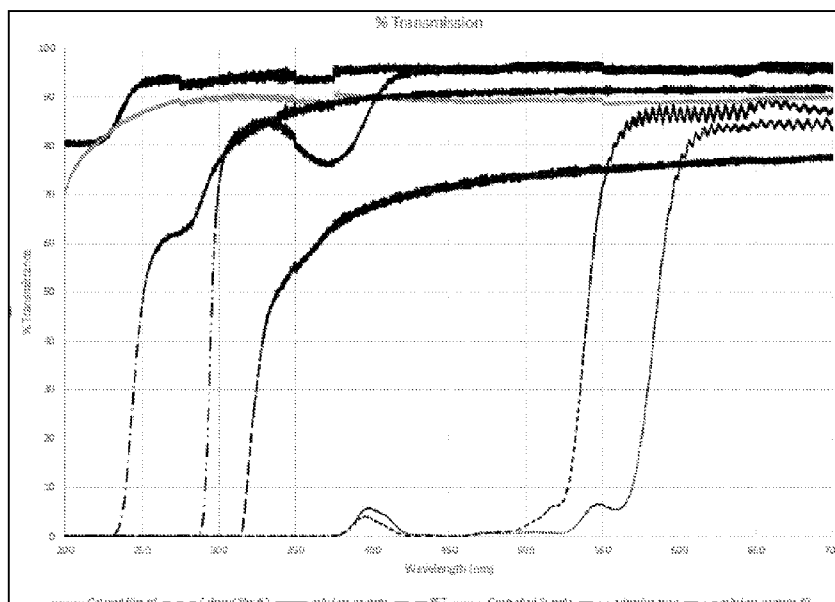


FIG. 1

(57) Abstract: The invention provides multilayer ultraviolet radiation dosage indicators, dosimeters comprising these indicators, processes for preparing these indicators, and methods of use of these indicators in several industrial and domestic applications that require a reduction or elimination of harmful microbial load that is sensitive to ultraviolet radiation. The multilayer dosage indicators comprise radiation sensitive compositions in one of the layers that are selective to ultraviolet radiation and show a differential opacity in response to this radiation.



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MULTILAYER RADIOCHROMIC STRUCTURES AND USES THEREOF

BACKGROUND

Field of the Invention

[0001] The disclosed and/or claimed inventive concept(s) provide multilayer ultraviolet (UV) radiation dosage indicators and dosimeters comprising these indicators.

Description of Related Art

[0002] In facilities where radiation sources are used, for example, in hospitals where cancer patients receive radiation treatments or in blood banks where blood products are irradiated, various methods are used to quantitatively determine the radiation exposure. The methods practiced include the use of thermoluminescent dosimeters (TLD's), ionization-type radiation detectors, photographic film, and radiochromic materials. TLD's are inconvenient because they require a complicated and time-consuming read-out process. Ionization-type radiation detectors are awkward and unwieldy and require a complicated setup. Photographic film requires a time-consuming chemical processing procedure before read-out. In case of radiochromic materials, the calculation of the dose requires a complex sequence of steps.

[0003] Photochromic polyacetylenes responsive to radiation exposure have been disclosed in several U.S. patents, namely U.S. 4,066,676; 4,581,315; 3,501,302; 3,501,297; 3,501,303; 3,501,308; 3,772,028; 3,844,791, and 3,954,816. The recording of image or dosage information using these polyacetylene compounds has presented several problems and shortcomings including an inadequate degree of resolution, clarity, color instability of an imaged pattern. Other deficiencies include a relatively slow image development, and, in some cases, the impractical need to image at extremely low temperatures or at excessively high dosage levels.

[0004] A preferred radiation sensitive material in radiation dosimeters includes dispersions of crystalline 10,12-pentacosadiynoic acid (PCDA). Subjecting monomeric PCDA crystals to ionizing radiation results in progressive polymerization, the degree of polymerization increasing with radiation dose. The amount of polymerization (and hence, the radiation dose) can be determined by measuring either the optical density or the spectral absorption of the exposed dosimeter. However, it has been found that these parameters also vary with both the temperature

of the device when measured as well as the thickness of PCDA dispersion. Maximum accuracy of dose measurement must account for the temperature and thickness effects.

[0005] Radiation dosimetry film provides a means for measuring radiation exposure at a point, but its principal utility is in obtaining a two-dimensional map of radiation exposure, i.e. radiation exposure at multiple points in a two-dimensional array. A typical user may measure an 8"×10" size film at a spatial resolution of 75 dpi, generating a map of radiation doses at 450,000 points. Of course, other resolutions can be used to generate the radiation exposure map.

[0006] U.S. Pat. No. 5,637,876 discloses a radiation dosimeter, exemplarily for use in determining a level of radiation to which a patient is subjected during radiation treatment, which comprises a substrate provided with a layer of radiation sensitive material. The radiation sensitive material has an optical density which varies systematically in accordance with the degree of radiation exposure. The dosimeter may take the form of a card or a flexible substrate which is positionable on the patient or other irradiation subject and which is also positionable in, or slidable through a slot in, a dose reader which includes a reflection or transmission densitometer.

[0007] U.S. Pat. No. 9,797,771 discloses a film manufactured for use in indicating an exposure and/or measuring dose of the exposure of long wavelength UV that is commonly used for UV curing of coating, pathogen inactivation and other industrial and medical applications.

[0008] U.S. Pat. No. 7,589,331 discloses a UV sensitive composition that undergoes a color change upon exposure to a predetermined dosage of UV-C radiation. The UV-C sensitive composition comprises a halogenated polymer, such as polyvinylidene chloride, that produces an acid upon exposure to UV radiation, and a pH sensitive dye. Upon exposure to UV-C radiation, the halogenated polymer undergoes degradation and produces HCl.

[0009] U.S. Pat. No. 9,097,588 discloses a UV radiation (UVR) response indicator which comprises a UVR sensitive material which has been modified so as to display an altered characteristic in a delayed manner in response to UVR exposure. The radiation sensitive material comprises a UVB and/or UVA radiation-driven acid release agent and a pH indicator which displays altered color between deprotonated and acid forms.

[0010] U.S. Pat. No. 8,829,457 discloses a UV radiation dosimeter apparatus for measuring an individual's UV radiation exposure from incoming UV rays, including an UV radiation dosimeter body; an UV filter in the UV radiation dosimeter body; a detector semiconductor substrate in the UV radiation dosimeter body connected to the UV filter for detecting the incoming UV rays and producing a signal, the semiconductor substrate made of ZnSe(Te), and a chip in the UV radiation dosimeter body for receiving the signal and measuring the individual's UV radiation exposure from the incoming UV rays.

[0011] U.S. Pat. No. 9,658,101 discloses dosimeters for measuring or detecting UV radiation wherein the dosimeters can be used with sunscreen, if desired, without the sunscreen having corrosive effects on photochromic materials in the dosimeters. The dosimeters are flexible enough to allow them to be shaped into a desired configuration to be worn on the wrist or other body part of a user, or on the clothes or equipment of a user.

[0012] U.S. Pat. No. 5,436,115 discloses a film that is substantially oxygen impermeable and changes color in response to exposure to UV radiation. The film comprises a mixture of a hydrophilic polymer, a nitro-substituted aromatic aldehyde for producing hydrogen ions in response to UV radiation and at least one dye that is substantially insensitive to changes in temperature and sensitive to changes in hydrogen ion concentration.

[0013] U.S. Pat. No. 6,426,503 discloses a dosimeter for measuring UV radiation. A UV filter has transmission characteristics that correspond to an erythema action curve, having two different decays in the high UV range. A photodiode receives UV radiation from the filter and outputs a current representing the amount of received radiation.

[0014] Welch et al. in *Radiation Protection Dosimetry*, 2017, volume 176 (4), pp. 341-346, examined unlaminate Gafchromic EBT3 film for UV radiation monitoring by exposing the film to select wavelengths in the UV spectrum ranging from 207 to 328 nanometers. The authors found the response of the film to be wavelength dependent and the highest sensitivity was observed for 254 nanometers.

[0015] It has been found that UV radiation dosage indicators and dosimeters comprising these indicators according to the disclosed and/or claimed inventive concept(s) (a) have superior properties such as the ease of usage and the nonrequirement of additional processing or

instrumentation to obtain the indicator response compared to the currently available commercial dosimeters and (b) are amenable for usage in several domestic, industrial, and healthcare applications that require reduction or elimination of microbial load that is particularly sensitive to UV radiation.

SUMMARY

[0016] In a first aspect, the disclosed and/or claimed inventive concept(s) provides a multilayer radiation dosage indicator comprising: (a) a base substrate comprising a visible mark; (b) at least one first layer on top of the base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; (c) at least one second layer on top of the first layer of ultraviolet radiation filtering composition wherein the second layer comprises a viewing zone through which the visible mark on the base substrate is viewable depending on opacity of the first layer prior to exposure to the ultraviolet radiation; and (d) an optional protective layer that's optionally transparent to the ultraviolet radiation.

[0017] In a second aspect, the disclosed and/or claimed inventive concept(s) provides a multilayer radiation dosage indicator comprising: (a) a base substrate comprising a visible mark; (b) at least one first layer on top of the base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; and (c) at least one second layer on top of the first layer of an ultraviolet radiation bandpass filter wherein the second layer comprises a viewing zone through which the visible mark on the base substrate is viewable depending on opacity of the first layer prior to exposure to the ultraviolet radiation.

[0018] In a third aspect, the disclosed and/or claimed inventive concept(s) provides a multilayer radiation dosage indicator comprising: (a) a base substrate; (b) at least one first layer on top of the base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; (c) at least one second layer on top of the first layer of ultraviolet radiation filtering composition; and (d) an optional protective layer that's optionally transparent to the ultraviolet radiation.

[0019] In a fourth aspect, the disclosed and/or claimed inventive concept(s) provides a multilayer radiation dosage indicator comprising: (a) a base substrate; (b) at least one first layer on top of the base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; and (c) at least one second layer on top of the first layer of an ultraviolet radiation bandpass filter.

[0020] In a fifth aspect, the disclosed and/or claimed inventive concept(s) provides a process for preparing a multilayer radiation dosage indicator comprising: (a) selecting a base substrate comprising a visible mark; (b) coating on the base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain a coated base substrate; and (c) coating on the coated base substrate at least one second layer of an ultraviolet radiation filtering composition wherein the second layer comprises a viewing zone through which the visible mark on the base substrate is viewable depending on opacity of the first layer prior to exposure to the ultraviolet radiation; and (d) optionally providing a protective layer on top of the second layer that's optionally transparent to the ultraviolet radiation.

[0021] In a sixth aspect, the disclosed and/or claimed inventive concept(s) provides a process for preparing a multilayer radiation dosage indicator comprising: (a) selecting a base substrate; (b) coating on the base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain a coated base substrate; and (c) coating on the coated base substrate at least one second layer of an ultraviolet radiation filtering composition; and (d) optionally providing a protective layer on top of the second layer that's optionally transparent to the ultraviolet radiation.

[0022] In a seventh aspect, the disclosed and/or claimed inventive concept(s) provides a process for preparing a multilayer radiation dosage indicator comprising: (a) selecting a base substrate; (b) printing on the base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain a printed base substrate; and (c) printing on the printed base substrate at least one second layer of an ultraviolet radiation filtering composition; and (d)

optionally providing a protective layer on top of the second layer that's optionally transparent to the ultraviolet radiation.

BRIEF DESCRIPTION OF THE DRAWINGS

[0023] The foregoing will be apparent from the following more particular description of exemplary, non-limiting embodiments of the disclosed and/or claimed inventive concept(s), as illustrated in the accompanying drawings. The drawings are not necessarily to scale, but emphasis is placed upon illustrating embodiments of the disclosed and/or claimed inventive concept(s).

[0024] FIG. 1 is a chart of percentage transmittance of different materials subjected to radiation (Y axis) as a function of the wavelength of radiation (in nanometers, X axis).

[0025] FIG. 2 is a schematic diagram (side view) of one exemplary, non-limiting embodiment of the multilayer radiation dosage indicator assembled in accordance with the disclosed and/or claimed inventive concept(s).

[0026] FIG. 3 is a schematic diagram (side view) of one exemplary, non-limiting embodiment of the multilayer radiation dosage indicator assembled in accordance with the disclosed and/or claimed inventive concept(s).

[0027] FIG. 4 is a schematic diagram (side view) of one exemplary, non-limiting embodiment of the multilayer radiation dosage indicator assembled in accordance with the disclosed and/or claimed inventive concept(s).

DETAILED DESCRIPTION

[0028] Before explaining at least one aspect of the disclosed and/or claimed inventive concept(s) in detail, it is to be understood that the disclosed and/or claimed inventive concept(s) is not limited in its application to the details of construction and the arrangement of the components or steps or methodologies set forth in the following description or illustrated in the drawings. The disclosed and/or claimed inventive concept(s) is capable of other aspects or of being practiced or carried out in various ways. Also, it is to be understood that the phraseology and terminology employed herein is for the purpose of description and should not be regarded as limiting.

[0029] Unless otherwise defined herein, technical terms used in connection with the disclosed and/or claimed inventive concept(s) shall have the meanings that are commonly understood by those of ordinary skill in the art. Further, unless otherwise required by context, singular terms shall include pluralities and plural terms shall include the singular.

[0030] All patents, published patent applications, and non-patent publications referenced in any portion of this application are herein expressly incorporated by reference in their entirety to the same extent as if each individual patent or publication was specifically and individually indicated to be incorporated by reference.

[0031] All articles and/or methods disclosed herein can be made and executed without undue experimentation based on the present disclosure. While the articles and methods of the disclosed and/or claimed inventive concept(s) have been described in terms of aspects, it will be apparent to those of ordinary skill in the art that variations may be applied to the articles and/or methods and in the steps or in the sequence of steps of the method described herein without departing from the concept, spirit and scope of the disclosed and/or claimed inventive concept(s). All such similar substitutes and modifications apparent to those skilled in the art are deemed to be within the spirit, scope, and concept of the disclosed and/or claimed inventive concept(s).

[0032] As utilized in accordance with the disclosure, the following terms, unless otherwise indicated, shall be understood to have the following meanings.

[0033] The use of the word “a” or “an” when used in conjunction with the term “comprising” may mean “one,” but it is also consistent with the meaning of “one or more,” “at least one,” and “one or more than one.” The use of the term “or” is used to mean “and/or” unless explicitly indicated to refer to alternatives only if the alternatives are mutually exclusive, although the disclosure supports a definition that refers to only alternatives and “and/or.”

[0034] Throughout this application, the term “about” is used to indicate that a value includes the inherent variation of error for the quantifying device, the method being employed to determine the value, or the variation that exists among the study subjects. For example, but not by way of limitation, when the term “about” is utilized, the designated value may vary by plus or minus twelve percent, or eleven percent, or ten percent, or nine percent, or eight percent, or seven percent, or six percent, or five percent, or four percent, or three percent, or two percent, or one percent.

[0035] The use of the term “at least one” will be understood to include one as well as any quantity more than one, including but not limited to, 1, 2, 3, 4, 5, 10, 15, 20, 30, 40, 50, 100, etc. The term “at least one” may extend up to 100 or 1000 or more depending on the term to which it is attached. In addition, the quantities of 100/1000 are not to be considered limiting as lower or higher limits may also produce satisfactory results. In addition, the use of the term “at least one of X, Y, and Z” will be understood to include X alone, Y alone, and Z alone, as well as any combination of X, Y, and Z. The use of ordinal number terminology (i.e., “first”, “second”, “third”, “fourth”, etc.) is solely for the purpose of differentiating between two or more items and, unless otherwise stated, is not meant to imply any sequence or order or importance to one item over another or any order of addition.

[0036] As used herein, the words “comprising” (and any form of comprising, such as “comprise” and “comprises”), “having” (and any form of having, such as “have” and “has”), “including” (and any form of including, such as “includes” and “include”) or “containing” (and any form of containing, such as “contains” and “contain”) are inclusive or open-ended and do not exclude additional, unrecited elements or method steps. The term “or combinations thereof” as used herein refers to all permutations and combinations of the listed items preceding the term. For example, “A, B_{Xn}, B_{Xn+1}, or combinations thereof” is intended to include at least one of: A, B_{Xn}, B_{Xn+1}, AB_{Xn}, A B_{Xn+1}, B_{Xn}B_{Xn+1}, or AB_{Xn}B_{Xn+1} and, if order is important in a particular context, also B_{Xn}A, B_{Xn+1}A, B_{Xn+1}B_{Xn}, B_{Xn+1}B_{Xn}A, B_{Xn}B_{Xn+1}A, AB_{Xn+1}B_{Xn}, B_{Xn}AB_{Xn+1}, or B_{Xn+1}AB_{Xn}. Continuing with this example, expressly included are combinations that contain repeats of one or more item or term, such as B_{Xn}B_{Xn}, AAA, MB_{Xn}, B_{Xn}B_{Xn}B_{Xn+1}, AAAB_{Xn}B_{Xn+1}B_{Xn+1}B_{Xn+1}, B_{Xn+1}B_{Xn}B_{Xn}AAA, B_{Xn+1}A B_{Xn}AB_{Xn}B_{Xn}, and so forth. The skilled artisan will understand that typically there is no limit on the number of items or terms in any combination, unless otherwise apparent from the context.

[0037] The term “each independently selected from the group consisting of” means when a group appears more than once in a structure, that group may be selected independently each time it appears.

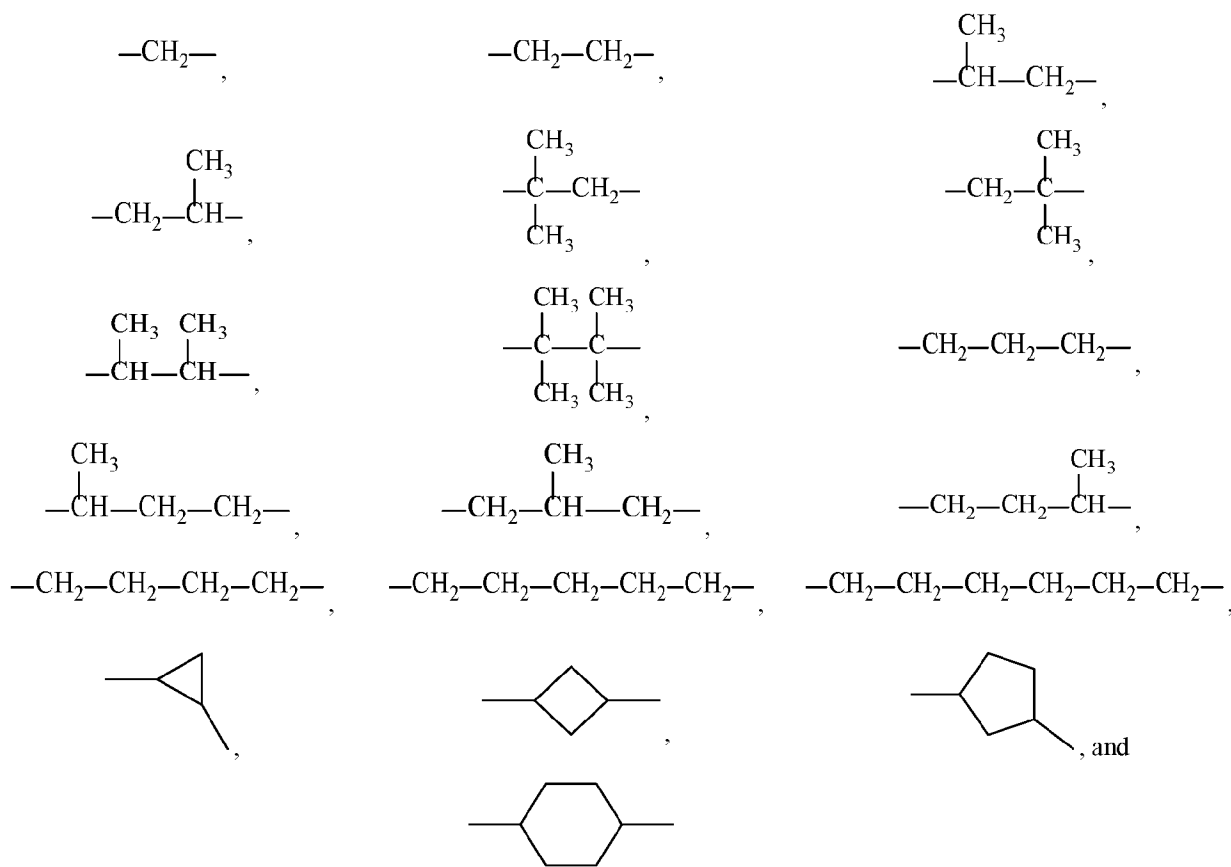
[0038] The term “hydrocarbyl” includes straight-chain and branched-chain alkyl, alkenyl, alkynyl, cycloalkyl, cycloalkenyl, aryl groups, and combinations thereof with optional heteroatom(s). A hydrocarbyl group may be mono-, di- or polyvalent.

[0039] The term “alkyl” refers to a functionalized or unfunctionalized, monovalent, straight-chain, branched-chain, or cyclic C₁-C₆₀ hydrocarbyl group optionally having one or more heteroatoms. In one non-limiting embodiment, an alkyl is a C₁-C₄₅ hydrocarbyl group. In another non-limiting embodiment, an alkyl is a C₁-C₃₀ hydrocarbyl group. Non-limiting examples of alkyl include methyl, ethyl, *n*-propyl, isopropyl, *n*-butyl, isobutyl, *tert*-butyl, *n*-pentyl, isopentyl, *n*-hexyl, *n*-heptyl, *n*-octyl, 2-ethylhexyl, *tert*-octyl, *iso*-norbornyl, *n*-dodecyl, *tert*-dodecyl, *n*-tetradecyl, *n*-hexadecyl, *n*-octadecyl, *n*-eicosyl, cyclobutyl, cyclopentyl, cyclohexyl, and the like. The definition of “alkyl” also includes groups obtained by combinations of straight-chain, branched-chain and/or cyclic structures.

[0040] The term “aryl” refers to a functionalized or unfunctionalized, monovalent, aromatic hydrocarbyl group optionally having one or more heteroatoms. The definition of aryl includes carbocyclic and heterocyclic aromatic groups. Non-limiting examples of aryl groups include phenyl, naphthyl, indenyl, indanyl, azulenyl, fluorenyl, anthracenyl, furyl, thienyl, pyridyl, pyrrolyl, oxazolyl, thiazolyl, imidazolyl, pyrazolyl, 2-pyrazolynyl, pyrazolidinyl, isoxazolyl, isothiazolyl, 1,2,3-oxadiazolyl, 1,2,3-triazolyl, 1,3,4-thiadiazolyl, pyridazinyl, pyrimidinyl, pyrazinyl, 1,3,5-triazinyl, 1,3,5-trithianyl, indoliziny, indolyl, isoindolyl, 3H-indolyl, indolinyl, benzo[b]furanyl, 2,3-dihydrobenzofuranyl, benzo[b]thiophenyl, 1H-indazolyl, benzimidazolyl, benzthiazolyl, purinyl, 4H-quinoliziny, isoquinoliny, cinnoliny, phthalazinyl, quinazolinyl, quinoxaliny, 1,8-naphthridiny, pteridinyl, carbazolyl, acridiny, phenazinyl, phenothiaziny, phenoxyazinyl, pyrazolo[1,5-c]triazinyl, and the like.

[0041] The term “aralkyl” refers to an alkyl group comprising one or more aryl substituent(s) wherein “aryl” and “alkyl” are as defined above. Non-limiting examples of aralkyl groups include benzyl, 2-phenyl-ethyl, 3-phenyl-propyl, 4-phenyl-butyl, 5-phenyl-pentyl, 4-phenylcyclohexyl, 4-benzylcyclohexyl, 4-phenylcyclohexylmethyl, 4-benzylcyclohexylmethyl, and the like.

[0042] The term “alkylene” refers to a functionalized or unfunctionalized, divalent, straight-chain, branched-chain, or cyclic C₁-C₄₀ hydrocarbyl group optionally having one or more heteroatoms. In one non-limiting embodiment, an alkylene is a C₁-C₃₀ group. In another non-limiting embodiment, an alkylene is a C₁-C₂₀ group. Non-limiting examples of alkylene groups include:



[0043] The term “arylene” refers to a functionalized or unfunctionalized, divalent, aromatic hydrocarbyl group optionally having one or more heteroatoms. The definition of arylene includes carbocyclic and heterocyclic groups. Non-limiting examples of arylene groups include phenylene, naphthylene, pyridinylene, and the like.

[0044] The term “heteroatom” refers to oxygen, nitrogen, sulfur, silicon, phosphorous, or halogen. The heteroatom(s) may be present as a part of one or more heteroatom-containing functional groups. Non-limiting examples of heteroatom-containing functional groups include ether, hydroxy, epoxy, carbonyl, carboxamide, carboxylic ester, carboxylic acid, imine, imide, amine, sulfonic, sulfonamide, phosphonic, and silane groups. The heteroatom(s) may also be present as a part of a ring such as in heteroaryl and heteroarylene groups.

[0045] The term “halogen” or “halo” refers to Cl, Br, I, or F.

[0046] The term “ammonium” includes protonated NH_3 as well as protonated primary, secondary, and tertiary organic amines.

[0047] The term “functionalized” with reference to any moiety refers to the presence of one or more “functional groups” in the moiety. Various functional groups may be introduced in a moiety by way of one or more functionalization reactions known to a person having ordinary skill in the art. Non-limiting examples of functionalization reactions include: alkylation, epoxidation, sulfonation, hydrolysis, amidation, esterification, hydroxylation, dihydroxylation, amination, ammonolysis, acylation, nitration, oxidation, dehydration, elimination, hydration, dehydrogenation, hydrogenation, acetalization, halogenation, dehydrohalogenation, Michael addition, aldol condensation, Canizzaro reaction, Mannich reaction, Claisen condensation, Suzuki coupling, carboxylation, sulfonation, carboxylic acid salt formation, sulfonic acid salt formation, and the like. The term “unfunctionalized” with reference to any moiety refers to the absence of functional groups in the moiety.

[0048] The term “monomer” refers to a small molecule that chemically bonds during polymerization to one or more monomers of the same or different kind to form a polymer.

[0049] The term “polymer” refers to a large molecule comprising one or more types of monomer residues (repeating units) connected by covalent chemical bonds. By this definition, polymer encompasses compounds wherein the number of monomer units may range from very few, which more commonly may be called as oligomers, to very many. Non-limiting examples of polymers include homopolymers, and non-homopolymers such as copolymers, terpolymers, tetra-polymers and the higher analogues. The polymer may have a random, block, and/or alternating architecture. The polymers may be nonionic, or may be cationic, anionic, or amphoteric in nature.

[0050] The term “homopolymer” refers to a polymer that consists essentially of a single monomer type.

[0051] The term “non-homopolymer” refers to a polymer that comprises more than one monomer types.

[0052] The term “copolymer” refers to a non-homopolymer that comprises two different monomer types.

[0053] The term “terpolymer” refers to a non-homopolymer that comprises three different monomer types.

[0054] The term “branched” refers to any non-linear molecular structure. The term includes both branched and hyper-branched structures.

[0055] The term “radiation sensitive” refers to the condition of exhibiting an alteration in one or more intrinsic or extrinsic properties in response to an incident radiation.

[0056] The term “metal” refers to a material that, when freshly prepared, polished, or fractured, typically shows a lustrous appearance, and is a good conductor of electricity and heat. This definition of a metal includes the several scientifically accepted categories of metals such as alkali metals, alkaline earth metals, lanthanoids, actinoids, transition metals, and post-transition metals.

[0057] The term “alkali metal” refers to metal elements lithium, sodium, potassium, rubidium, cesium, and francium.

[0058] The term “alkaline earth metal” refers to metal elements beryllium, magnesium, calcium, strontium, barium, and radium.

[0059] The term “lanthanoid” refers to metal elements with atomic numbers 57 through 71 (inclusive) in the periodic table, from lanthanum through lutetium.

[0060] The term “actinoid” refers to metal elements with atomic numbers 89 through 103 (inclusive) in the periodic table, from actinium through lawrencium.

[0061] The term “transition metal” refers to a metal element whose atom has a partially filled d sub-shell, or which can give rise to cations with an incomplete d sub-shell. Particular, yet non-limiting examples of post-transition metals include scandium, titanium, vanadium, chromium, manganese, iron, cobalt, nickel, copper, zinc, rhodium, and palladium.

[0062] The term “post-transition metal” refers to a metal element that is typically soft, has poor mechanical strength, and/or has melting point lower than those of transition metal elements. Particular, yet non-limiting examples of post-transition metals include aluminum, gallium, indium, thallium, tin, lead, bismuth, and polonium.

[0063] The term “metalloid” refers to an element which demonstrates properties which are intermediate between the properties of typical metals and typical non-metals. For example, a metalloid may be an element which has the physical appearance and properties of a metal but behaves chemically as a non-metal. Particular, yet non-limiting examples of metalloids include silicon, boron, arsenic, germanium, antimony, and tellurium.

[0064] The term “coating composition” refers to a composition in the form of, for example, a solution, an emulsion, a suspension, or a dispersion, that is suitable for applying onto a surface of a substrate.

[0065] The term “substrate” refers to a material that serves as a base for a composition such as a coating composition.

[0066] The term “device” refers to a fabricated material.

[0067] The term “discontinuous coating” refers to a coating that unlike a film does not provide a complete coverage of the surface of a substrate on which the coating is applied. Particular, yet non-limiting examples of discontinuous coatings include those obtained from printing processes such as inkjet printing, dot matrix printing, layer printing, pad printing and the like.

[0068] The term “bandpass filter” refers to an optical filter that allows transmission of a specific range of wavelengths and can reject wavelengths both above and below that range.

[0069] All percentages, ratio, and proportions used herein are based on a weight basis unless other specified.

[0070] In a first aspect, the disclosed and/or claimed inventive concept(s) provides a multilayer radiation dosage indicator comprising: (a) a base substrate comprising a visible mark; (b) at least one first layer on top of the base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; (c) at least one second layer on top of the first layer of ultraviolet radiation filtering composition wherein the second layer comprises a viewing zone through which the visible mark on the base substrate is viewable depending on opacity of the first layer prior to exposure to the ultraviolet radiation; and (d) an optional protective layer that’s optionally transparent to the ultraviolet radiation.

[0071] In a second aspect, the disclosed and/or claimed inventive concept(s) provides a multilayer radiation dosage indicator comprising: (a) a base substrate comprising a visible mark; (b) at least one first layer on top of the base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; and (c) at least one second layer on top of the first layer of an ultraviolet radiation bandpass filter wherein the second layer comprises a viewing zone through which the visible mark on the base substrate is viewable depending on opacity of the first layer prior to exposure to the ultraviolet radiation.

[0072] In a third aspect, the disclosed and/or claimed inventive concept(s) provides a multilayer radiation dosage indicator comprising: (a) a base substrate; (b) at least one first layer on top of the base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; (c) at least one second layer on top of the first layer of ultraviolet radiation filtering composition; and (d) an optional protective layer that's optionally transparent to the ultraviolet radiation.

[0073] In a fourth aspect, the disclosed and/or claimed inventive concept(s) provides a multilayer radiation dosage indicator comprising: (a) a base substrate; (b) at least one first layer on top of the base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; and (c) at least one second layer on top of the first layer of an ultraviolet radiation bandpass filter.

[0074] In one non-limiting embodiment, the first layer is optically transparent prior to exposure to the ultraviolet radiation such that the visible mark on the base substrate is viewable. In another non-limiting embodiment, the first layer is opaque prior to exposure to the ultraviolet radiation such that the visible mark on the base substrate is nonviewable.

[0075] In one non-limiting embodiment, the first layer becomes opaque after exposure to the ultraviolet radiation such that the visible mark on the base substrate is nonviewable. In another non-limiting embodiment, the first layer becomes optically transparent after exposure to the ultraviolet radiation such that the visible mark on the base substrate is viewable.

[0076] In one non-limiting embodiment, the ultraviolet radiation has a wavelength in the range from about 100 nanometer to about 400 nanometer.

[0077] In one non-limiting embodiment, the ultraviolet radiation is ultraviolet-C radiation having a wavelength in the range from about 100 nanometer to 280 nanometer. In another non-limiting embodiment, the ultraviolet radiation is ultraviolet-B radiation having a wavelength in the range from 281 nanometer to 310 nanometer. In yet another non-limiting embodiment, the ultraviolet radiation is ultraviolet-A radiation having a wavelength in the range from 311 nanometer to about 400 nanometer.

[0078] In one non-limiting embodiment, the ultraviolet radiation has a wavelength in the range from about 100 nanometer to about 310 nanometer. In another non-limiting embodiment, the ultraviolet radiation has a wavelength in the range from about 280 nanometer to about 400 nanometer. In yet another non-limiting embodiment, the ultraviolet radiation has a wavelength in the range from about 100 nanometer to about 280 nanometer or from about 311 nanometer to about 400 nanometer.

[0079] In one non-limiting embodiment, the multilayer radiation dosage indicator according to the disclosed and/or claimed inventive concept(s) is selectively responsive to exposure to ultraviolet-C radiation having a wavelength in the range from about 100 nanometer to 280 nanometer.

[0080] In one non-limiting embodiment, the radiation sensitive composition according to the disclosed and/or claimed inventive concept(s) comprises at least one acetylenic compound.

[0081] In one non-limiting embodiment, the acetylenic compound comprises at least one acetylene moiety and at least one non-acetylenic functional group.

[0082] In one non-limiting embodiment, the non-acetylenic functional group is selected from the group consisting of carboxyl, carboxylate, hydroxy, hydroxide, alkoxy, alkoxide, epoxy, amino, ammonium, aldehyde, keto, amide, ester, nitrile, urethane, ether, and combinations thereof. In another non-limiting embodiment, the non-acetylenic functional group is selected from the group consisting of carboxyl, carboxylate, and combinations thereof.

[0083] In one non-limiting embodiment, the acetylenic compound is selected from the group consisting of decadiynoic acids, undecadiynoic acids, dodecadiynoic acids, tridecadiynoic acids, tetradecadiynoic acids, pentadecadiynoic acids, hexadecadiynoic acids, heptadecadiynoic acids,

octadecadiynoic acids, nonadecadiynoic acids, icosadiynoic acids, heneicosadiynoic acids, docosadiynoic acids, tricosadiynoic acids, tetracosadiynoic acids, pentacosadiynoic acids, hexacosadiynoic acids, heptacosadiynoic acids, octacosadiynoic acids, nonacosadiynoic acids, triacontadiynoic acids, salts thereof, and combinations thereof.

[0084] In one non-limiting embodiment, the acetylenic compound is selected from the group consisting of 10,12-pentacosadiynoic acid, salts thereof, and combinations thereof.

[0085] In one non-limiting embodiment, the salt as disclosed herein is obtained by the reaction of the functionalized or unfunctionalized acetylenic compound and a substance selected from the group consisting of organic acids, organic bases, inorganic acids, inorganic bases, complex formers, crystal formers, cocrystal formers, and combinations thereof.

[0086] In another non-limiting embodiment, the salt as disclosed herein is obtained by the reaction of the functionalized or unfunctionalized acetylenic compound and a substance selected from the group consisting of functionalized or unfunctionalized aliphatic amines, alicyclic amines, heterocyclic amines, aromatic amines, heteroaromatic amines, and combinations thereof.

[0087] In yet another non-limiting embodiment, the salt as disclosed herein is obtained by the reaction of the functionalized or unfunctionalized acetylenic compound and a substance selected from the group consisting of functionalized or unfunctionalized alkyl amines, dialkyl amines, trialkyl amines, quaternary amines, pyridines, azopyridines, bipyridyls, pyrimidines, pyrazines, piperidines, bipiperidines, morpholines, and combinations thereof.

[0088] In yet another non-limiting embodiment, the salt as disclosed herein is obtained by the reaction of the functionalized or unfunctionalized acetylenic compound and a substance selected from the group consisting of metals and inorganic bases.

[0089] In yet another non-limiting embodiment, the salt as disclosed herein is obtained by the reaction of the functionalized or unfunctionalized acetylenic compound and a substance selected from the group consisting of hydrides, oxides, hydroxides, cyanides, carbonates, and bicarbonates of alkali and alkaline earth metal elements, and combinations thereof.

[0090] Non-limiting, yet particular examples of organic bases include 4,4'-azopyridine, 4,4'-bipyridyl, trans-1,2-bis(4-pyridyl)ethylene, 4,4'-bipiperidine, morpholine, diethylamine, n-

butylamine, and combinations thereof. Other suitable examples of organic bases can be found in ULLMANN's Encyclopedia of Industrial Chemistry, 7th Edition, 2002, Wiley-VCH Verlag GmbH & Co. KGaA, the contents of which are herein incorporated by reference in its entirety.

[0091] Non-limiting, yet particular examples of inorganic bases can be found in ULLMANN's Encyclopedia of Industrial Chemistry, 7th Edition, 2002, Wiley-VCH Verlag GmbH & Co. KGaA, the contents of which are herein incorporated by reference in its entirety.

[0092] Additional insight into the properties, functionality and application(s) of radiation sensitive acetylene compounds is disclosed in Hall et al. in *Chemical Science*, 2020, volume 11, 8025-8035, the disclosure of which is herein incorporated by reference in its entirety.

[0093] In one non-limiting embodiment, the radiation sensitive composition according to the disclosed and/or claimed inventive concept(s) comprises at least one radiation sensitive dye.

[0094] In one non-limiting embodiment, the radiation sensitive dye is selected from the group consisting of spiropyrans, spirothiopyrans, spironaphthooxazines, spirobenzopyrans, spiroindolobenzopyrans, chromenes, 2,2,-dichlorochromenes, leuco quinines, anthroquinone dyes, thiazine leuco dyes, oxazine leuco dyes, phenazine leuco dyes, monoarylmethane phthalides, diarylmethane phthalides, triarylmethane phthalides, monoheterocyclic phthalides, bisheterocyclic phthalides, alkenylphthalides, bridged phthalides, bisphthalides, diarylmethanes, triarylmethanes, triarylmethane lactones, fluoran leuco dyes, tetrazolium salts, diazo dyes, nitro dyes, phthalein dyes, triphenylmethane dyes, benzeins, indophenols, quinolines, anthraquinones, indigo dyes, indamines, thiazines, pH-sensitive dyes, and UV-oxidizable dyes.

[0095] Non-limiting, yet particular examples of radiation sensitive dye include diphenyl iodonium (DPI) chloride, DPI- hexafluorophosphate, DPI-perfluor-1-butanesulfonate, DPI-triflate, 4-iodophenyl diphenyl sulfonium triflate, 4-methylthiophenyl diphenyl sulfonium triflate, 2-naphthyl diphenyl sulfonium triflate, 4- chlorophenyl diphenyl sulfonium triflate, and 4-bromophenyl diphenyl sulfonium triflate, thymol blue, malachite green, bromocresol green, indophenol blue, hydroxyethyl amino-azobenzene, methyl red, phenol red, ethyl orange, *m*-Cresol purple, New Fuchsin, *p*-methyl red, lissamine green, aniline blue, methyl violet, crystal violet, ethyl violet, brilliant green, orallochite green oxalate, methyl green, cresol red, quinaldine red, para methyl red, metanil yellow, orange IV, phenylazoaniline, erythrosin B, benzopurpurin, congo red,

methyl orange, bromocresol green, resazurin, alizarin red, bromocresol purple, chlorophenol red, bromophenol blue, carbazoyl methane, bisindophthalide, fluoran, 4-(pyrrolidino)azobenzene, methylene blue, calcein, nitro blue tetrazolium salt, victoria blue B carbinol, auramine carbinol, p-phenylazophenol, 4-phenylazodiphenylamine, 4-phenylazo-1-naphthylamine, 4-phenylazoresorcinol, 3-methyl-4-phenylazophenol, p-phenylazophenyl isocyanate, 4-(p-phenylazophenyl) semicarbazide, Rhodamine 6G, quinaldine red, benzophenylsafranine, Bismarck brown, Sudan orange, safranine O, and the like.

[0096] In one non-limiting embodiment, the base substrate is a material selected from the group consisting of paper, polymer, plastic, textile, metal, alloy, cloth, wood, leather, ceramic, glass, stone, and combinations thereof.

[0097] In one non-limiting embodiment, the second layer is a continuous film or a discontinuous coating on the first layer of the multilayer radiation dosage indicator according to the disclosed and/or claimed inventive concept(s).

[0098] Details of printing processes that provide discontinuous coatings on the surface of a substrate can be obtained from the Handbook of Print Media: Technologies and Production Methods (2001), Ed. Helmut Kipphan, Springer Science & Business Media, that is herein incorporated in its entirety by reference. Details of coating processes that provide continuous coatings on the surface of a substrate can be obtained from Modern Coating and Drying Technology (1992), Eds. E.D. Cohen and E.B. Gutoff, Wiley, that is herein incorporated in its entirety by reference.

[0099] In one non-limiting embodiment, the second layer comprises a film obtained from a polymer selected from the group consisting of carbohydrates, polysaccharides, polyethers, polyesters, polyamides, polyethylene terephthalates, polyolefins, polyurethanes, polycarbonates, polycarbamates, polylactides, polyglycolides, copolymers of lactides and glycolides, polymers derived from vinylic monomers, polymers derived from (meth)acrylic monomers, polyvinyl alcohols, polyvinyl acetates, polyvinyl butyrals, and combinations thereof.

[00100] Non-limiting, yet particular examples of polyolefins include polyethylene (PE), low-density polyethylene (LDPE), linear low-density polyethylene (LLDPE), very low-density polyethylene (VLDPE), ultra-low-density polyethylene (ULDPE), medium-density polyethylene

(MDPE), high-density polyethylene (HDPE), ultra-high-density polyethylene (UHDPE), ethylene/butene-1 copolymers, ethylene/hexene-1 copolymers, ethylene/octene-1 copolymers, cyclic olefin copolymers (COC), ethylene/propylene copolymers (PEP), polypropylene (PP), propylene/ethylene copolymer (PPE), polyisoprene, polybutylene (PB), polybutene-1, poly-3-methylbutene-1, poly-4-methylpentene-1, ionomers (IO), and propylene/ α -olefins (P/AO).

[00101] Non-limiting, yet particular examples of polyesters include polyethylene terephthalate (PET), amorphous polyethylene terephthalate (APET), crystalline polyethylene terephthalate (CPET), glycol-modified polyethylene terephthalate (PETG), polybutylene terephthalate, polyethylene terephthalate/isophthalate copolymer, polylactic acid (PLA), polyglycolic acid (PGA), polylactic acid-co-glycolic acid (PLGA), polyhydroxypropionate, poly(3-hydroxybutyrate) (PH3B), poly(3-hydroxyvalerate) (PH3V), poly(4-hydroxybutyrate) (PH4B), poly(4-hydroxyvalerate) (PH4V), poly(5-hydroxyvalerate) (PH5V), and poly(6-hydroxydodecanoate) (PH6D).

[00102] Non-limiting, yet particular examples of polyamides include nylon 6 (polycaprolactam), nylon 11 (polyundecanolactam), nylon 12 (polylauryllactam), nylon 4,2 (polytetramethylene ethylenediamide), nylon 4,6 (polytetramethylene adipamide), nylon 6,6 (polyhexamethylene adipamide), nylon 6,9 (polyhexamethylene azelamide), nylon 6,10 (polyhexamethylene sebacamide), nylon 6,12 (polyhexamethylene dodecanediamide), nylon 7,7 (polyheptamethylene pimelamide), nylon 8,8 (polyoctamethylene suberamide), nylon 9,9 (polynonamethylene azelamide), nylon 10,9 (polydecamethylene azelamide), nylon 12,12 (polydodecamethylene dodecanediamide), nylon 6,6/6 copolymer (polyhexamethylene adipamide/caprolactam copolymer), nylon 6/6,6 copolymer (polycaprolactam/hexamethylene adipamide copolymer), nylon 6,2/6,2 copolymer (polyhexamethylene ethylenediamide/hexamethylene ethylenediamide copolymer), and nylon 6,6/6,9/6 copolymer (polyhexamethylene adipamide/hexamethylene azelaamide/caprolactam copolymer).

[00103] Non-limiting, yet particular examples of polysaccharides include cellulose, cellulose acetate, carboxymethyl cellulose, carboxymethyl hydroxyethyl cellulose, cellulose acetate propionate carboxylate, hydroxyethyl ethyl cellulose, hydroxypropyl cellulose, hydroxypropyl methylcellulose, methyl hydroxyethyl cellulose, microcrystalline cellulose, sodium cellulose sulfate, methyl cellulose, ethyl cellulose, alkyl celluloses, hydroxyalkyl celluloses, cationic

celluloses, starches, modified starches, carboxymethyl starch, hydroxyethyl starch, hydroxypropyl starch, epichlorohydrin crosslinked hydroxypropyl starch, amylopectin, modified amylopectin, amylose, modified amylose, galactomannans, modified galactomannans, guar gum, xanthan gum, gellan gum, welan gum, hydroxypropyl guar, carboxymethyl hydroxypropyl guar, locust bean gum, ghatti gum, karaya gum, tamarind gum, carrageenan, alginates, glycosaminoglycans, hyaluronic acid, and derivatives of hyaluronic acid. Further non-limiting, yet particular examples of cellulose polymers can be found in the book chapter Cellulose-Based Polymers for Packaging Applications by Tajeddin (2014), In *Lignocellulosic Polymer Composites*, V.K. Thakur (Ed.), Scrivener Publishing, the contents of which are herein incorporated by reference in entirety.

[00104] Non-limiting, yet particular examples of polymers derived from (meth)acrylic monomers include homopolymers, copolymers, terpolymers, and higher order polymers derived from acrylic acid, methacrylic acid, itaconic acid, β -carboxyethyl acrylate, hydroxyethyl acrylate, hydroxypropyl acrylate, hydroxyethyl methacrylate, hydroxypropyl methacrylate, methyl acrylate, butyl acrylate, amyl acrylate, hexyl acrylate, 2-ethylhexyl acrylate, octyl acrylate, isooctyl acrylate, decyl acrylate, and dodecyl acrylates.

[00105] In one non-limiting embodiment, the ultraviolet radiation filtering composition comprises at least one compound selected from the group consisting of metal compounds, metalloid compounds, organic compounds, organometallic compounds, and combinations thereof.

[00106] In one non-limiting embodiment, the metal compound is selected from the group consisting of metal oxides, metal sulfides, metal sulfates, metal hydroxides, metal carbonates, metal silicates, metal chlorides, metal fluorides, metal bromides, metal iodides, metal salts, metal complexes, metal alloys, metal nitrates, and combinations thereof.

[00107] In one non-limiting embodiment, the metal compound is selected from the group consisting of titanium monoxide, titanium dioxide, titanium trioxide, aluminum oxide, aluminum sulfate, aluminum potassium sulfate, lead sulfate, lead chloride, lead bromide, lead oxide, lead iodide, barium chloride, barium sulfate, barium oxide, tungsten carbide, tungsten oxide, cesium chloride, cesium bitartrate, cesium halide, cesium oxide, cesium sulfide, cesium carbonate, zinc oxide, and combinations thereof.

[00108] In one non-limiting embodiment, the metal compound is a cesium compound selected from the group consisting of cesium chloride, cesium bitartrate, cesium halide, cesium polyhalide, cesium polyiodide, cesium oxide, cesium sulfide, cesium polysulfide, cesium carbonate, and combinations thereof. Band et al. in *J. Phys. Chem. B* (2004), 108, 33, 12360–12367 synthesize cesium oxides and characterize them using a combination of chemical and structural analysis techniques. Ferguson and Gorrie in Cesium and Cesium Compounds, *Kirk-Othmer Encyclopedia of Chemical Technology* (2011) describe the methods of extraction, synthesis, properties, and applications of cesium compounds. The contents of these references are herein incorporated by reference in their entirety.

[00109] In one non-limiting embodiment, the metal compound is a barium compound selected from the group consisting of barium sulfate, barium oxide, barium dithionate, and combinations thereof. Kresse et al. in Barium and Barium Compounds, *Ullmann's Encyclopedia of Industrial Chemistry* (2007) describe the history, production, analysis, and uses of barium and barium compounds, the contents of which are herein incorporated by reference in its entirety.

[00110] In one non-limiting embodiment, the metal compound is a tungsten compound selected from the group consisting of tungsten carbide, tungsten oxide, and combinations thereof. Lassner et al. in book chapter: Tungsten Compounds and Their Application, Tungsten (1999) provide a summary of tungsten compounds with metallic as well as non-metallic elements, the contents of which are herein incorporated by reference in its entirety.

[00111] In one non-limiting embodiment, the metal compound is an aluminum compound selected from the group consisting of aluminum oxide, aluminum hydride, aluminum sulfate, aluminum potassium sulfate, and combinations thereof. In another non-limiting embodiment, the metal compound is a lead compound selected from the group consisting of lead salts, lead sulfate, lead oxide, lead chloride, lead bromide, lead iodide, and combinations thereof. In yet another non-limiting embodiment, the metal compound is titanium dioxide.

[00112] In one non-limiting embodiment, the metalloid compound is selected from the group consisting of boron oxides, boron carbides, boron nitrides, boron halides, boric acid, organoboron compounds, silicon oxides, organosilicon compounds, silicon halides, silicon carbides, silica, silicates, silicic acid, and combinations thereof.

[00113] In one non-limiting embodiment, the ultraviolet radiation filtering composition comprises an organic compound selected from the group consisting of dibenzoyl methanes, diphenyl acrylic esters, benzophenones, triazines, benzotriazoles, benzimidazoles, benzalmalonates, tetrazolium salts, spiranes, spiropyrans, naphthopyrans, spirooxazines, pH-sensitive dyes, bipyridiliums, biquinolyls, bis(pyridinium) ethylenes, diazapyriniums, tropeolins, nitrophenols, phthaleins, naphthalenes, benzeins, indophenols, quinolines, anthraquinones, indamines, thiazines, diazoniums, bisimidazoles, bispyrroles, phenothiazines, triphenylmethanes, and combinations thereof.

[00114] In one non-limiting embodiment, the first layer is a continuous film or a discontinuous coating on the base substrate.

[00115] In one non-limiting embodiment, the base substrate is a polymer selected from the group consisting of carbohydrates, polysaccharides, polyethers, polyesters, polyethylene terephthalates, polyolefins, polyurethanes, polycarbonates, polycarbamates, polylactides, polyglycolides, copolymers of lactides and glycolides, polymers derived from vinylic monomers, polymers derived from (meth)acrylic monomers, polyvinyl alcohols, polyvinyl acetates, and combinations thereof.

[00116] In one non-limiting embodiment, the first layer in the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) becomes opaque in response to exposure to the ultraviolet radiation dosage exceeding about 0.5 mJ/cm^2 .

[00117] In one non-limiting embodiment, the first layer in the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) becomes opaque in response to exposure to the ultraviolet radiation dosage in a range from about 1 mJ/cm^2 to about 100 mJ/cm^2 .

[00118] In one non-limiting embodiment, the first layer in the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) becomes opaque in response to exposure to the ultraviolet radiation dosage exceeding about 150 mJ/cm^2 .

[00119] In one non-limiting embodiment, the radiation dosage is in the range from about 200 mJ/cm^2 to 300 mJ/cm^2 and the ultraviolet radiation is ultraviolet-C radiation having a wavelength in range from about 100 nanometer to 280 nanometer. In another non-limiting embodiment, the

radiation dosage is in the range from 301 mJ/cm² to 1999 mJ/cm² and the ultraviolet radiation is ultraviolet-B radiation having a wavelength in range from 281 nanometer to 310 nanometer. In yet another non-limiting embodiment, the radiation dosage is in the range from 2000 mJ/cm² to about 10000 mJ/cm² and the ultraviolet radiation is ultraviolet-A radiation having a wavelength in range from 311 nanometer to about 400 nanometer.

[00120] Further details of the radiation dosage ranges and methods of treatment for microbial reduction using radiation dosimeters are described in the articles by Lachert, E. in *Journal of Transfusion Medicine*, Vol. 12 (3), 2019, pp. 83-87 and Levy J. H. et al. in *Critical Care*, Vol. 22 (1), 2018, each of which are herein incorporated in its entirety by reference.

[00121] In one non-limiting embodiment, the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) comprises means to attach the indicator to an object. In one non-limiting embodiment, the object is located in a hospital, laboratory, clinic, shop, warehouse, factory, vehicle, hotel, restaurant, or residential property. In one non-limiting embodiment, the object is a blood bag, plasma bag, biological fluid bag, catheter, surgical instrument, laboratory equipment, hospital equipment, hospital surface, personnel protective equipment, apron, gloves, gown, or surgical attire.

[00122] In one non-limiting embodiment, the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) comprises an adhesive layer as a means to attach the indicator to an object.

[00123] In one non-limiting embodiment, the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) comprises means to attach the indicator to an object that requires exposure to ultraviolet radiation for a reduction in the concentration of microbes. In one non-limiting embodiment, the microbe is selected from the group consisting of bacterium, fungus, virus, pathogen, and combinations thereof. In one non-limiting embodiment, the reduction in concentration is the reduction from an unsafe concentration to safe concentration for human or animal use. In one non-limiting embodiment, the reduction in concentration is selected from the group consisting of reduction in bulk concentration, reduction in surface concentration, and combinations thereof.

[00124] In one non-limiting embodiment, the protective layer in the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) comprises a continuous film or discontinuous coating on the second layer of the multilayer radiation dosage indicator.

[00125] In one non-limiting embodiment, the protective layer in the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) comprises a continuous film or discontinuous coating comprising a polymer. In one non-limiting embodiment, the polymer is selected from the group consisting of carbohydrates, polysaccharides, polyethers, polyesters, polyethylene terephthalates, polyolefins, polyurethanes, polycarbonates, polycarbamates, polylactides, polyglycolides, copolymers of lactides and glycolides, polymers derived from vinylic monomers, polymers derived from (meth)acrylic monomers, polyvinyl alcohols, polyvinyl acetates, polyvinyl butyrals, and combinations thereof.

[00126] In one non-limiting embodiment, the ultraviolet radiation bandpass filter in the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) enables ultraviolet-C radiation having a wavelength in range from about 100 nanometer to 280 nanometer to pass. In another non-limiting embodiment, the ultraviolet radiation bandpass filter in the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) enables ultraviolet-B radiation having a wavelength in range from 281 nanometer to 310 nanometer to pass. In yet another non-limiting embodiment, the ultraviolet radiation bandpass filter in the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) enables ultraviolet-A radiation having a wavelength in range from 311 nanometer to about 400 nanometer to pass.

[00127] In one non-limiting embodiment, the radiation sensitive composition in the multilayer radiation dosage indicator according to disclosed and/or claimed inventive concept(s) comprises at least one amelioration agent.

[00128] In one non-limiting embodiment, the amelioration agent is selected from the group consisting of binders, resins, dyes, polymers, shelf-life extenders, solvents, stabilizers, surfactants, activators, and combinations thereof.

[00129] Non-limiting, yet particular examples of binders include homopolymers, copolymers, graft-copolymers, block copolymers, polymeric alloys, and mixtures thereof. A large number of

monomers and oligomers can be used to make these polymeric binders. Non-limiting, yet particular examples of such monomers include unsaturated monomers such as olefins, vinyls, acrylates, and (meth)acrylates such as methyl methacrylate, methyl acrylate, styrene, acrylic acid, butane diol 1,4-dimethacrylate, diethylene glycol diacrylate, diethylene glycol dimethacrylate, ethylene glycol dimethacrylate, hexanediol-1,6-dimethacrylate, methylstyrene pentaerythritol triacrylate, polyethylene glycol dimethacrylate, polypropylene glycol dimethacrylate, triethylene glycol dimethacrylate, 4-(vinylloxy) butyl benzoate, bis[4-(vinylloxy)butyl] adipate, bis[4-(vinylloxy)butyl] succinate, 4-(vinylloxymethyl)cyclohexylmethyl, bis[4-(vinylloxy)butyl] isophthalate, bis[4-(vinylloxymethyl)cyclohexylmethyl], tris[4-(vinylloxy)butyl] trimellitate, 4-(vinylloxy)butyl stearate, bis[4-(vinylloxy)butyl] hexanediylbiscarbamate, bis[[4-(vinylloxy)methyl]cyclohexyl]methyl, bis[[4-[(vinylloxy)methyl]cyclohexyl]methyl], bis[4-(vinylloxy)butyl] (4-methyl-1,3-phenylene), and combinations thereof.

[00130] Further non-limiting, yet particular examples of binders include latex emulsion polymers. For a latex emulsion polymer, components include polymers which are the reaction products of one or more ethylenically unsaturated monomers. Non-limiting, yet particular examples of useful ethylenically unsaturated monomers include acrylic acid, acrylonitrile, acetoacetoxy ethyl methacrylate, acetoacetoxy ethyl acrylate, butyl acrylate, butadiene, butyl methacrylate, butyl acrylamide, chloromethyl styrene, crotonic acid, ethyl acrylate, ethyl acrylamide, ethylene, ethyl methacrylate, ethylhexyl acrylate, 2-ethylhexyl acrylate, 2-ethylhexyl methacrylate, glycidyl methacrylate, hydroxyethyl (meth)acrylate, hydroxypropyl (meth)acrylate, isobutyl acrylate, isobutyl methacrylate, isoprene, iso-octyl acrylate, iso-octyl methacrylate, itaconic acid, methyl acrylate, octyl acrylate, octyl methacrylate, methyl methacrylate, methacrylic acid, , α -methyl styrene, styrene, vinyl chloride vinyl naphthalene, vinyl toluene, vinylidene chloride, vinyl acetate, and the like.

[00131] Alkyd resins are generally comprised of polybasic acids, polyhydric alcohols, and fatty acids which may be unsaturated. The polybasic acids such as aromatic, aliphatic and alicyclic saturated and unsaturated compounds, such as adipic acid, chlorendic acid, heptanedioic acid, isophthalic acid, maleic acid, naphthalic acid, phthalic acid, sebacic acid, succinic acid, trimellitic acid, terephthalic acid, and tetrahydrophthalic acid. Polyhydric alcohol components include 1,3-butylene glycol, diethylene glycol, dipentaerythritol, dipropylene glycol, ethylene glycol, glycerin,

1,6-hexanediol, neopentyl glycol, pentaerythritol, propylene glycol, sorbitol, trimethylol ethane, trimethylol propane and triethylene glycol. Fatty acids used in the manufacture of alkyds commonly include dehydrated castor oil, coconut oil, cottonseed oil, fish oil, linseed oil, oiticica oil, tung oil, safflower oil, soya oil and tall oil acids.

[00132] Polyurethane resins are formed from polyisocyanate (aliphatic, aromatic, or combinations thereof) compounds. Examples of aliphatic isocyanates include butane diisocyanate, 4,4'-diisocyanatodicyclohexylmethane, hexamethylene diisocyanate, hexahydroxylylene diisocyanate, isophorone diisocyanate, 1-methyl-2,4(2,6)-diisocyanato cyclohexane, norbornane diisocyanate, and tetramethylxylylene diisocyanate. Examples of aliphatic and aromatic isocyanates include 4,4'-biphenylene diisocyanate, , 4-chloro-1,3- phenylene diisocyanate, 1,4-cyclohexylene diisocyanate, 1,10-deca-ethylene diisocyanate, methylene bis-(4-phenyl isocyanate), 4,4- methylene-bis(cyclohexyl isocyanate), 1,5-naphthalene diisocyanate, 1,3-phenylene diisocyanate, 2,4-tolylene diisocyanate, 2,6-tolylene diisocyanate, , 1,4-tetramethylene diisocyanate and 1,5-tetrahydronaphthalene diisocyanate.

[00133] In addition to isocyanates, alcohols and carboxylic acids, which form polyester compositions, can also be used in the preparation of polyurethane resins. The polycarboxylic acids may be of an aliphatic, cycloaliphatic, aromatic and/or heterocyclic nature and may comprise halogen atoms and/or unsaturated moieties. Suitable acids include adipic acid, azeleic acid, bis-glycol terephthalate, dimeric fatty acids, dimethyl terephthalate, endomethylenetetrahydrophthalic anhydride, fumaric acid, glutaric anhydride, hexahydrophthalic anhydride, isophthalic acid, maleic acid, maleic anhydride, phthalic anhydride, phthalic acid, suberic acid, succinic acid, sebacic acid, tetrahydrophthalic anhydride and tetrachlorophthalic anhydride. Polyhydric alcohols examples include 1,4-, 1,3- and 2,3-butylene glycol, cyclohexanedimethanol (1,4-bis-hydroxymethylcyclohexane), diethylene glycol, dipropylene glycol, dibutylene glycol, ethylene glycol, 1,2- and 1,3-propylene glycol, 1,6-hexanediol, 2-methyl-1,3-propanediol, neopentylglycol, 1,8-octanediol, polyethylene glycol, polypropylene glycol, polybutylene glycol, triethylene glycol and tetraethylene glycol. Polyesters comprising carboxyl groups and terminal carboxyl groups are envisioned. Diols comprising carboxyl or carboxylate groups which are suitable to support ionic or potentially ionic groups are envisioned. Such moieties can be constructed by dihydroxysuccinic acid, dimethylolacetic acid, 2,2-dimethylolpropionic acid, 2,2-dimethylolbutyric acid and 2,2-

dimethylolpentanoic acid. Polyesters constructed from lactones are also envisioned. Polycarbonates comprising hydroxyl groups are useful and are prepared by reacting diols with dicarbonates such as diphenyl carbonate or phosgene. Polyethers comprising diols, formed from polymers derived from ethylene oxide, propylene oxide and/or tetrahydrofuran are also useful. An amine functionality can be employed to introduce terminal hydroxyl functionality, with compounds such as diethanolamine, ethanolamine, N-methylethanolamine, propanolamine, N,N,N'-tris-2-hydroxyethyl-ethylendiamine.

[00134] Epoxy resins are comprised primarily of linear chain molecules. These molecules are formed from the reaction of bisphenols with halohydrins to yield epoxy resins containing epoxy groups. Common bisphenols include bisphenol-A, bisphenol-F, bisphenol-S, and 4,4' dihydroxy bisphenol. Common halohydrins include epichlorohydrin, dichlorohydrin, and 1,2-dichloro-3-hydroxypropane.

[00135] Non-limiting, yet particular examples of commercially available epoxy resins include Dow Chemical epoxy resins DER 333, DER 661 and Shell Chemical epoxy resins EPON 828, EPON 836, and EPON 1001. Ciba-Geigy epoxy resins GT-7013, GT-7014, GT-7074, GT-259 and Air Products Ancarez® AR 555.

[00136] Non-limiting, yet particular examples of commercial latex emulsion polymers, available from Air Products, BASF, Bayer, Celanese, Chemtura, Dow, Hexion, Styron and Wacker, include Ancarez®, Acclaim®, Acronal 296D, Aquamac 705, Aquamac 588, Arcol®, Avanse™, Avicor®, Baybond®, Bayhydrol®, Bayhydur®, Britecoat®, Celvaset®, Desmodur®, Dur-o-coat®, Dur-o-set®, Ecovae®, EVEREST™ Latex Technology, ENVERSA™, Flexbond®, Formashield™, FOUNDATIONS™ Latex, Hiloft®, HPL™ Latex, LOMAX™ Latex, Nacrylic®, Resyn®, Rhoplex SG-30, Rhoplex HG-74P, Rhoplex SG-10M, Rhoplex AC2508, Trumoda®, Tufcor®, UCAR 313, UCAR 626, UCAR 379G, Vinac®, Vinamul®, Vinnapas®, Witcobond®, X-Link® and the like. Combinations of latex emulsions are also envisioned. Latex can also be functionalized and further cross-linked as required.

[00137] Non-limiting, yet particular examples of rheology modifiers include cellulosics, modified ureas, polyurethane thickeners and associative thickeners, alkali swellable emulsions (ASE), hydrophobically modified alkali swellable emulsions (HASE), hydrophobically modified

polyurethanes (HEURS), hydrophobically modified polyethers (HMPE), attapulgites, acrylate thickeners, amides and organic derivatives, fumed silicas, synthetic layered silicates, organoclays, mixed minerals, thixotropy boosters, polyalkylene ether derivatives, starches, polyacrylates, surfactants, and hydrophobically modified polymers and copolymers.

[00138] Non-limiting, yet particular examples of functional binders, including rheological modification, are polyvinyl pyrrolidone, poly(vinyl pyrrolidone-co-vinyl acetate), polyvinylcaprolactam, poly(vinyl pyrrolidone-co-vinyl caprolactam), poly(vinyl pyrrolidone-co-dimethylaminopropylmethacrylamide), poly(vinyl pyrrolidone-co-vinyl caprolactam-co-dimethylaminopropylmethacrylamide), poly(isobutylene-co-ethylmaleimide-co-hydroxyethylmaleimide), poly(vinyl pyrrolidone-co-dimethylaminoethylmethacrylate), quaternized poly(vinyl pyrrolidone-co-dimethylaminoethylmethacrylate), poly(vinyl pyrrolidone-co-vinyl caprolactam-co-dimethylaminoethylmethacrylate), poly(vinyl acetate-co-crotonates-co-vinylneodecanoate), poly(2-ethyl oxazoline), polyethylene imine, poly(2-alkyl oxazoline), poly(octylacrylamide-co-acrylate-co-butylamino ethylmethacrylate), polyvinyl alcohol, partially hydrolyzed polyvinyl alcohol, acetoacetylated polyvinyl alcohol, polyalkylene imines, polydi allyldialkylammonium chloride, starches, modified starches, polyurethanes, acrylated oligomers of polyurethanes, acrylated glycidyl oligomers, polyfunctional acrylates, cellulosics, silane or silicone based polymers, anionic polyacrylates such as polyacrylic acid, poly(maleic anhydride-alt-methyl vinyl ether), poly(methylvinylether-alt-maleic acid), poly(methylvinylether-alt-maleic acid)ester, poly(methylvinylether-alt-maleic anhydride)ester copolymer, poly(styrene-co-maleic acid), poly(styrene-co-maleic acid)ester, poly(styrene-co-maleic anhydride), poly(styrene-co-maleic anhydride)ester, styrene acrylic acid copolymer, styrene acrylic acid ester copolymer, hydrophobically modified polyacetal polyether, maleimide/maleic acid copolymer, urethane acrylate methacrylate, acrylic polymer, polyoxyethylene-polyoxypropylene block copolymer, and hydrophobically modified ethoxylated urethane. Cellulose and derivatives include ethyl cellulose (EC), hydroxyethyl cellulose (HEC), hydroxypropyl cellulose (HPC), ethylhydroxyethyl cellulose (EHEC), carboxymethyl cellulose (CMC), carboxymethylhydroxyethyl cellulose (CMHEC), hydroxypropylhydroxyethyl cellulose (HMHEC), methylcellulose (MC), methylhydroxypropyl cellulose (MHPC), methylhydroxyethyl cellulose (MHEC), carboxymethylmethylcellulose (CMMC), hydrophobically modified carboxymethylcellulose (HMC MC), hydrophobically modified hydroxyethyl cellulose (HMHEC), hydrophobically modified hydroxylpropyl cellulose

(HMHPC), hydrophobically modified ethylhydroxyethyl cellulose (HMEHEC), hydrophobically modified carboxymethylhydroxyethyl cellulose (HMCMHEC), hydrophobically modified hydroxypropylhydroxyethyl cellulose (HMHPHEC), hydrophobically modified methyl cellulose (HMMC), hydrophobically modified methylhydroxypropyl cellulose (HMMHPC), hydrophobically modified methylhydroxyethyl cellulose (HMMHEC), hydrophobically modified carboxymethylmethyl cellulose (HMCMMC), cationic hydroxyethyl cellulose (cationic HEC), cationic hydrophobically modified hydroxyethyl cellulose (cationic HMHEC), nano fibrillated celluloses (NFC), and microfibrillated celluloses (MFC). Guar and guar derivatives include carboxymethyl guar, carboxymethylhydroxypropyl guar, cationic hydroxypropyl guar, hydroxyalkyl guar, including hydroxyethyl guar, hydroxypropyl guar, hydroxybutyl guar and higher hydroxylalkyl guar, carboxylalkyl guar, including carboxymethyl guar, carboxylpropyl guar, carboxybutyl guar, and higher alkyl carboxy guar, the hydroxyethylated, hydroxypropylated and carboxymethylated derivative of guaran, the hydroxethylated and carboxymethylated derivatives of Carubin and the hydroxypropylated and carboxymethylated derivatives of *Cassia-Gum*.

[00139] Non-limiting, yet particular examples of solvents include water, high boiling solvents such as butoxy-2-ethylstearate, butyrolactone, diethyl fumarate, dimethyl maleate, dimethylcarbonate, dioctyl phthalate, ethylene glycol dimethyl ether ethyl salicylate, polyethylene glycol dimethylether, propylene carbonate, triacetin, benzyl ether, dodecyl-1,2-methyl pyrrolidone, ethoxyethylacetate, ethylene glycol diacetate, ethyltrichloroacetate, methylpyrrolidone, methyl sulfoxide, polyethylene glycols of different molecular weight, dimethylformamide, cyclohexane, p-dioxane, tetrahydrofuran, and p-xylene.

[00140] Non-limiting, yet particular examples of dyes include new Sunset Yellow, fuschin cyanide, hexahydroxy ethyl violet cyanide, pararose aniline cyanide, leuco crystal violet, leuco malachite green, carbinol dyes such as malachite green base and p-roseaniline base, and those described in U.S. Pat. Nos. 2,877,169 ; 3,079,955 ; and 4,377,751, each of which disclosure is herein incorporated by reference in its entirety. Other examples of dyes can be found in the patent EP1529089 B1 that is herein incorporated by reference in its entirety.

[00141] Non-limiting, yet particular examples of activators include a halocarbon, a halonium, a sulfonium, ethyl trichloroacetate, heptachloropropane, ethyltrichloroacetate, chloroacetic acid,

chloropropionic acid, hexachlorocyclohexane, methyltrichloroacetimidate, trichloroacetic acid, trichloroacetamide, trichloro ethanol, trichloro methyl benzyl acetate, trichloro methyl propanol hydrate, trichloro propane, chlorinated polymers, diphenyliodonium iodide, diphenyliodonium hexafluoroarsenate, diphenyliodonium chloride, trimethylsulfonium iodide and triphenylsulfonium hexafluoroantimonate.

[00142] In a fifth aspect, the disclosed and/or claimed inventive concept(s) provides a process for preparing a multilayer radiation dosage indicator comprising: (a) selecting a base substrate comprising a visible mark; (b) coating on the base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain a coated base substrate; and (c) coating on the coated base substrate at least one second layer of an ultraviolet radiation filtering composition wherein the second layer comprises a viewing zone through which the visible mark on the base substrate is viewable depending on opacity of the first layer prior to exposure to the ultraviolet radiation; and (d) optionally providing a protective layer on top of the second layer that is optionally transparent to the ultraviolet radiation.

[00143] In a sixth aspect, the disclosed and/or claimed inventive concept(s) provides a process for preparing a multilayer radiation dosage indicator comprising: (a) selecting a base substrate comprising a visible mark; (b) printing on the base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain a coated base substrate; and (c) printing on the printed base substrate at least one second layer of an ultraviolet radiation filtering composition wherein the second layer comprises a viewing zone through which the visible mark on the base substrate is viewable depending on opacity of the first layer prior to exposure to the ultraviolet radiation; and (d) optionally providing a protective layer on top of the second layer that is optionally transparent to the ultraviolet radiation.

[00144] In a seventh aspect, the disclosed and/or claimed inventive concept(s) provides a process for preparing a multilayer radiation dosage indicator comprising: (a) selecting a base substrate; (b) coating on the base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain a coated base substrate; and (c) coating on the coated base

substrate at least one second layer of an ultraviolet radiation filtering composition; and (d) optionally providing a protective layer on top of the second layer that is optionally transparent to the ultraviolet radiation.

[00145] In an eighth aspect, the disclosed and/or claimed inventive concept(s) provides a process for preparing a multilayer radiation dosage indicator comprising: (a) selecting a base substrate; (b) printing on the base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain a coated base substrate; and (c) printing on the printed base substrate at least one second layer of an ultraviolet radiation filtering composition; and (d) optionally providing a protective layer on top of the second layer that is optionally transparent to the ultraviolet radiation.

[00146] The general process for preparing multilayer radiation dosage indicators is described in the U.S. Pat. No. 5,051,597, the disclosure of which is herein incorporated by reference in its entirety.

[00147] In one non-limiting embodiment, the multilayer radiation dosage indicators according to the claimed and/or disclosed inventive concept(s) are used for detection and/or measurement of ultraviolet radiation in dental, non-destructive testing, oncological, radiological or radiotherapeutic applications.

[00148] Non-limiting examples of oncological, radiological or radiotherapeutic applications include radiation therapy, surgery, chemotherapy, immunotherapy, and hormonal therapy. Non-limiting examples of cancers curable with radiation therapy either alone or in combination with other modalities include skin cancer, prostate carcinomas, lung carcinomas, cervix carcinomas, lymphomas (Hodgkin's and low grade Non-Hodgkin's), head and neck carcinomas, breast carcinomas, rectal and anal carcinomas, local advanced cervix carcinomas, bladder carcinomas, endometrial carcinomas, CNS tumors, soft tissue sarcomas, and pediatric tumors. More information on cancer and radiation therapy and its current advances and future directions can be found in Baskar et al., *Int J Med Sci*, **2012** (9), 193-199 that is herein incorporated in its entirety by reference. Monitoring of the oral cavity and dental health is required during radiation therapy, particularly of the head and neck, to decrease the severity of the side effects.

[00149] In one non-limiting embodiment, the method of measurement and/or detection of ultraviolet radiation using the multilayer radiation dosage indicator according to the claimed and/or disclosed inventive concept(s) is non-destructive in nature. In general, in the fields of radiology and radiography, non-destructive detection, testing and/or measuring methods are those that help to maintain the integrity and properties of materials or components that are exposed to radiation without causing undue damage to the tested object.

[00150] Details of the fabrication, mechanism of operation, and use of bandpass filters can be found in the following references: book chapter - Op Amps for Everyone (Third Edition, 2009), Eds. Ron Mancini, Bruce Carter, Chapter 20: 'Active Filter Design Techniques', Thomas Kugelstadt, Newnes, book - Handbook of Optical Filters for Fluorescence Microscopy (2000), Jay Reichman, Chroma Technology Corp. USA, and book - Handbook of Optics: Fundamentals, Techniques, and Design (1995), Ed. Michael Bass, Optical Society of America, McGraw-Hill, the disclosure of each of which is herein incorporated by reference in its entirety.

[00151] FIG. 1 is a chart of percentage transmittance versus wavelength (in nanometer) of different materials such as a colored film #1, a colored film #2, a film of cellulose acetate #1, a film of PET, a film of controlled sample, a film of an adhesive tape, and a film of cellulose acetate #2. As seen in FIG. 1, these materials exhibit significantly different UV transmission properties.

[00152] The multilayer radiation dosage indicators according to the disclosed and/or claimed inventive concept(s) may be prepared according to the examples set out below. These examples are presented herein for purposes of illustration of the disclosed and/or claimed inventive concept(s) and are not intended to be limiting, for example, the preparations of the indicators.

[00153] FIG. 2 shows the side view of an exemplary, non-limiting embodiment of the multilayer radiation dosage indicators according to the disclosed and/or claimed inventive concept(s). The base substrate with a visible mark is the bottom-most layer of the multilayer assembly. On the top of the base layer, a layer of a UV radiation sensitive composition is appropriately positioned (labelled as the first layer). On top of the first layer, a layer of a UV radiation filtering composition is positioned (labelled as the second layer). On top of the second layer, a protective layer is positioned.

[00154] FIG. 3 shows the side view of an exemplary, non-limiting embodiment of the multilayer radiation dosage indicators according to the disclosed and/or claimed inventive concept(s). The base substrate is the bottom-most layer of the multilayer assembly. On the top of the base layer, a layer of a UV radiation sensitive composition is appropriately positioned (labelled as the first layer). On top of the first layer, a layer of a band pass filter is positioned.

[00155] FIG. 4 shows the side view of an exemplary, non-limiting embodiment of the multilayer radiation dosage indicators according to the disclosed and/or claimed inventive concept(s). The base substrate is a polyester film layer present as the bottom-most layer of the multilayer assembly. On the top of the base substrate, a layer of a radiation sensitive composition is appropriately positioned (labelled as the first layer). On top of the first layer, a layer of a cellulose-based film is appropriately positioned (labelled as the second layer). On top of the second layer, a protective layer of a discontinuous polyester film is positioned.

What we claim is:

1. A multilayer radiation dosage indicator comprising:
 - (a) a base substrate comprising a visible mark;
 - (b) at least one first layer on top of said base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold;
 - (c) at least one second layer on top of said first layer of ultraviolet radiation filtering composition wherein said second layer comprises a viewing zone through which said visible mark on said base substrate is viewable depending on opacity of said first layer prior to exposure to said ultraviolet radiation; and
 - (d) an optional protective layer that's optionally transparent to said ultraviolet radiation.
2. A multilayer radiation dosage indicator comprising:
 - (a) a base substrate comprising a visible mark;
 - (b) at least one first layer on top of said base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; and
 - (c) at least one second layer on top of said first layer of an ultraviolet radiation bandpass filter wherein said second layer comprises a viewing zone through which said visible mark on said base substrate is viewable depending on opacity of said first layer prior to exposure to said ultraviolet radiation.
3. A multilayer radiation dosage indicator comprising:
 - (a) a base substrate;
 - (b) at least one first layer on top of said base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold;
 - (c) at least one second layer on top of said first layer of ultraviolet radiation filtering composition; and
 - (d) an optional protective layer that's optionally transparent to said ultraviolet radiation.
4. A multilayer radiation dosage indicator comprising:

- (a) a base substrate;
 - (b) at least one first layer on top of said base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; and
 - (c) at least one second layer on top of said first layer of an ultraviolet radiation bandpass filter.
5. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 wherein said first layer is optically transparent prior to exposure to said ultraviolet radiation such that said visible mark is viewable.
 6. The multilayer radiation dosage indicator according to claim 5 wherein said first layer becomes opaque after exposure to said ultraviolet radiation such that said visible mark is nonviewable.
 7. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 wherein said first layer is opaque prior to exposure to said ultraviolet radiation such that said visible mark is nonviewable.
 8. The multilayer radiation dosage indicator according to claim 7 wherein said first layer becomes optically transparent after exposure to said ultraviolet radiation such that said visible mark is viewable.
 9. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 wherein said radiation sensitive composition comprises at least one acetylenic compound.
 10. The multilayer radiation dosage indicator according to claim 9 wherein said acetylenic compound comprises at least one acetylene moiety and at least one non-acetylenic functional group.
 11. The multilayer radiation dosage indicator according to claim 10 wherein said non-acetylenic functional group is selected from the group consisting of carboxyl, carboxylate, hydroxy, hydroxide, alkoxy, alkoxide, epoxy, amino, ammonium, aldehyde, keto, amide, ester, nitrile, urethane, ether, and combinations thereof.

12. The multilayer radiation dosage indicator according to claim 11 wherein said non-acetylenic functional group is selected from the group consisting of carboxyl, carboxylate, and combinations thereof.
13. The multilayer radiation dosage indicator according to claim 10 wherein said acetylenic compound selected from the group consisting of decadiynoic acids, undecadiynoic acids, dodecadiynoic acids, tridecadiynoic acids, tetradecadiynoic acids, pentadecadiynoic acids, hexadecadiynoic acids, heptadecadiynoic acids, octadecadiynoic acids, nonadecadiynoic acids, icosadiynoic acids, heneicosadiynoic acids, docosadiynoic acids, tricosadiynoic acids, tetracosadiynoic acids, pentacosadiynoic acids, hexacosadiynoic acids, heptacosadiynoic acids, octacosadiynoic acids, nonacosadiynoic acids, triacontadiynoic acids, salts thereof, and combinations thereof.
14. The multilayer radiation dosage indicator according to claim 13 wherein said acetylenic compound is selected from the group consisting of 10,12-pentacosadiynoic acid, a salt thereof, and combinations thereof.
15. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 wherein said radiation sensitive composition comprises at least one radiation sensitive dye.
16. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 wherein said base substrate is a material selected from the group consisting of paper, polymer, plastic, textile, metal, alloy, cloth, wood, leather, ceramic, glass, stone, and combinations thereof.
17. The multilayer radiation dosage indicator according to claim 1 or 3 wherein said second layer is a continuous film or a discontinuous coating on said first layer.
18. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 wherein said ultraviolet radiation filtering composition comprises at least one compound selected from the group consisting of metal compounds, metalloid compounds, organic compounds, organometallic compounds, and combinations thereof.
19. The multilayer radiation dosage indicator according to claim 18 wherein said metal compound is selected from the group consisting of metal oxides, metal sulfides, metal

sulfates, metal hydroxides, metal carbonates, metal silicates, metal chlorides, metal fluorides, metal bromides, metal iodides, metal salts, metal complexes, metal alloys, metal nitrates, and combinations thereof.

20. The multilayer radiation dosage indicator according to claim 19 wherein said metal compound is selected from the group of titanium monoxide, titanium dioxide, titanium trioxide, aluminum oxide, aluminum sulfate, aluminum potassium sulfate, lead sulfate, lead chloride, lead bromide, lead oxide, lead iodide, barium chloride, barium sulfate, barium oxide, tungsten carbide, tungsten oxide, cesium chloride, cesium bitartrate, cesium halide, cesium oxide, cesium sulfide, cesium carbonate, zinc oxide, and combinations thereof.
21. The multilayer radiation dosage indicator according to claim 20 wherein said metal compound is titanium dioxide.
22. The multilayer radiation dosage indicator according to claim 18 wherein said metalloid compound is selected from the group consisting of boron oxides, boron carbides, boron nitrides, boron halides, boric acid, organoboron compounds, silicon oxides, organosilicon compounds, silicon halides, silicon carbides, silica, silicates, silicic acid, and combinations thereof.
23. The multilayer radiation dosage indicator according to claim 18 wherein said organic compound is selected from the group consisting of dibenzoyl methanes, diphenyl acrylic esters, benzophenones, triazines, benzotriazoles, benzimidazoles, benzalmalonates, tetrazolium salts, spiranes, spiropyrans, naphthopyrans, spirooxazines, pH-sensitive dyes, bipyridiliums, biquinolyls, bis(pyridinium) ethylenes, diazapyriniums, tropeolins, nitrophenols, phthaleins, naphthalenes, benzeins, indophenols, quinolines, anthraquinones, indamines, thiazines, diazoniums, bisimidazoles, bispyrroles, phenothiazines, triphenylmethanes, and combinations thereof.
24. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 wherein said first layer is a continuous film or a discontinuous coating on said base substrate.

25. The multilayer radiation dosage indicator according to claim 16 wherein said polymer is selected from the group consisting of carbohydrates, polysaccharides, polyethers, polyesters, polyamides, polyethylene terephthalates, polyolefins, polyurethanes, polycarbonates, polycarbamates, polylactides, polyglycolides, copolymers of lactides and glycolides, polymers derived from vinylic monomers, polymers derived from (meth)acrylic monomers, polyvinyl alcohols, polyvinyl acetates, polyvinyl butyrals, and combinations thereof.
26. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 wherein said ultraviolet radiation has a wavelength in range from about 100 nanometer to about 400 nanometer.
27. The multilayer radiation dosage indicator according to claim 26 wherein said ultraviolet radiation is ultraviolet-C radiation having a wavelength in range from about 100 nanometer to 280 nanometer.
28. The multilayer radiation dosage indicator according to claim 26 wherein said ultraviolet radiation is ultraviolet-B radiation having a wavelength in range from 281 nanometer to 310 nanometer.
29. The multilayer radiation dosage indicator according to claim 26 wherein said ultraviolet radiation is ultraviolet-A radiation having a wavelength in range from 311 nanometer to about 400 nanometer.
30. The multilayer radiation dosage indicator according to claim 26 wherein said ultraviolet radiation has wavelength in range from about 100 nanometer to about 310 nanometer.
31. The multilayer radiation dosage indicator according to claim 26 wherein said ultraviolet radiation has wavelength in range from about 280 nanometer to about 400 nanometer.
32. The multilayer radiation dosage indicator according to claim 26 wherein said ultraviolet radiation has wavelength in range from about 100 nanometer to about 280 nanometer or from about 311 nanometer to about 400 nanometer.

33. The multilayer radiation dosage indicator according to claim 26 that is selectively responsive to exposure to ultraviolet-C radiation having a wavelength in range from about 100 nanometer to 280 nanometer.
34. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 wherein said first layer becomes opaque in response to exposure to ultraviolet radiation dosage exceeding about 0.5 mJ/cm^2 .
35. The multilayer radiation dosage indicator according to claim 34 wherein said first layer becomes opaque in response to exposure to said ultraviolet radiation dosage in range from about 1 mJ/cm^2 to about 100 mJ/cm^2 .
36. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 wherein said first layer becomes opaque in response to exposure to said ultraviolet radiation dosage exceeding about 150 mJ/cm^2 .
37. The multilayer radiation dosage indicator according to claim 36 wherein said dosage is in range from about 200 mJ/cm^2 to 300 mJ/cm^2 .
38. The multilayer radiation dosage indicator according to claim 36 wherein said ultraviolet radiation is ultraviolet-C radiation having a wavelength in range from about 100 nanometer to 280 nanometer.
39. The multilayer radiation dosage indicator according to claim 36 wherein said dosage is in range from 301 mJ/cm^2 to 1999 mJ/cm^2 .
40. The multilayer radiation dosage indicator according to claim 36 wherein said ultraviolet radiation is ultraviolet-B radiation having a wavelength in range from 281 nanometer to 310 nanometer.
41. The multilayer radiation dosage indicator according to claim 36 wherein said dosage is in range from 2000 mJ/cm^2 to about 10000 mJ/cm^2 .

42. The multilayer radiation dosage indicator according to claim 36 wherein said ultraviolet radiation is ultraviolet-A radiation having a wavelength in range from 311 nanometer to about 400 nm.
43. The multilayer radiation dosage indicator according to claim 36 wherein said first layer becomes opaque in response to exposure to said ultraviolet radiation dosage in range from about 2000 mJ/cm² to about 10000 mJ/cm².
44. The multilayer radiation dosage indicator according to claim 1, 2, 3, or 4 comprising means to attach said indicator to an object.
45. The multilayer radiation dosage indicator according to claim 44 wherein said means is an adhesive layer.
46. The multilayer radiation dosage indicator according to claim 44 wherein said object requires exposure to said ultraviolet radiation dosage for reduction in concentration of microbes.
47. The multilayer radiation dosage indicator according to claim 46 wherein said object is located in a hospital, laboratory, clinic, shop, warehouse, factory, vehicle, hotel, restaurant, or residential property.
48. The multilayer radiation dosage indicator according to claim 47 wherein said object is a blood bag, plasma bag, biological fluid bag, catheter, surgical instrument, laboratory equipment, hospital equipment, hospital surface, personnel protective equipment, apron, gloves, gown, or surgical attire.
49. The multilayer radiation dosage indicator according to claim 46 wherein said microbe is selected from the group consisting of bacterium, fungus, virus, pathogen, and combinations thereof.
50. The multilayer radiation dosage indicator according to claim 46 wherein said reduction in concentration is reduction from unsafe concentration to safe concentration for human or animal use.

51. The multilayer radiation dosage indicator according to claim 46 wherein said reduction in concentration is selected from the group consisting of reduction in bulk concentration, reduction in surface concentration, and combinations thereof.
52. The multilayer radiation dosage indicator according to claim 1 or 3 wherein said protective layer comprises a continuous film or discontinuous coating on said second layer.
53. The multilayer radiation dosage indicator according to claim 52 wherein said continuous film or discontinuous coating comprises a polymer.
54. The multilayer radiation dosage indicator according to claim 53 wherein said polymer is selected from the group consisting of carbohydrates, polysaccharides, polyethers, polyesters, polyethylene terephthalates, polyolefins, polyurethanes, polycarbonates, polycarbamates, polylactides, polyglycolides, copolymers of lactides and glycolides, polymers derived from vinylic monomers, polymers derived from (meth)acrylic monomers, polyvinyl alcohols, polyvinyl acetates, polyvinyl butyrals, and combinations thereof.
55. The multilayer radiation dosage indicator according to claim 2 or 4 wherein said ultraviolet radiation bandpass filter enables ultraviolet-C radiation having a wavelength in range from about 100 nanometer to 280 nanometer to pass.
56. The multilayer radiation dosage indicator according to claim 2 or 4 wherein said ultraviolet radiation bandpass filter enables ultraviolet-B radiation having a wavelength in range from 281 nanometer to 310 nanometer to pass.
57. The multilayer radiation dosage indicator according to claim 2 or 4 wherein said ultraviolet radiation bandpass filter enables ultraviolet-A radiation having a wavelength in range from 311 nanometer to about 400 nanometer to pass.
58. A process for making a multilayer radiation dosage indicator comprising: (a) selecting a base substrate comprising a visible mark; (b) coating on said base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain

a coated base substrate; and (c) coating on said coated base substrate at least one second layer of an ultraviolet radiation filtering composition wherein said second layer comprises a viewing zone through which said visible mark on said base substrate is viewable depending on opacity of said first layer prior to exposure to said ultraviolet radiation; and (d) optionally providing a protective layer on top of said second layer that's optionally transparent to said ultraviolet radiation.

59. A process for making a multilayer radiation dosage indicator comprising: (a) selecting a base substrate comprising a visible mark; (b) printing on said base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain a printed base substrate; and (c) printing on said printed base substrate at least one second layer of an ultraviolet radiation filtering composition wherein said second layer comprises a viewing zone through which said visible mark on said base substrate is viewable depending on opacity of said first layer prior to exposure to said ultraviolet radiation; and (d) optionally providing a protective layer on top of said second layer that's optionally transparent to said ultraviolet radiation.
60. A process for making a multilayer radiation dosage indicator comprising: (a) selecting a base substrate; (b) coating on said base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain a coated base substrate; and (c) coating on said coated base substrate at least one second layer of an ultraviolet radiation filtering composition; and (d) optionally providing a protective layer on top of said second layer that's optionally transparent to said ultraviolet radiation.
61. A process for making a multilayer radiation dosage indicator comprising: (a) selecting a base substrate; (b) printing on said base substrate at least one first layer of a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold to obtain a printed base substrate; and (c) printing on said printed base substrate at least one second layer of an ultraviolet

radiation filtering composition; and (d) optionally providing a protective layer on top of said second layer that's optionally transparent to said ultraviolet radiation.

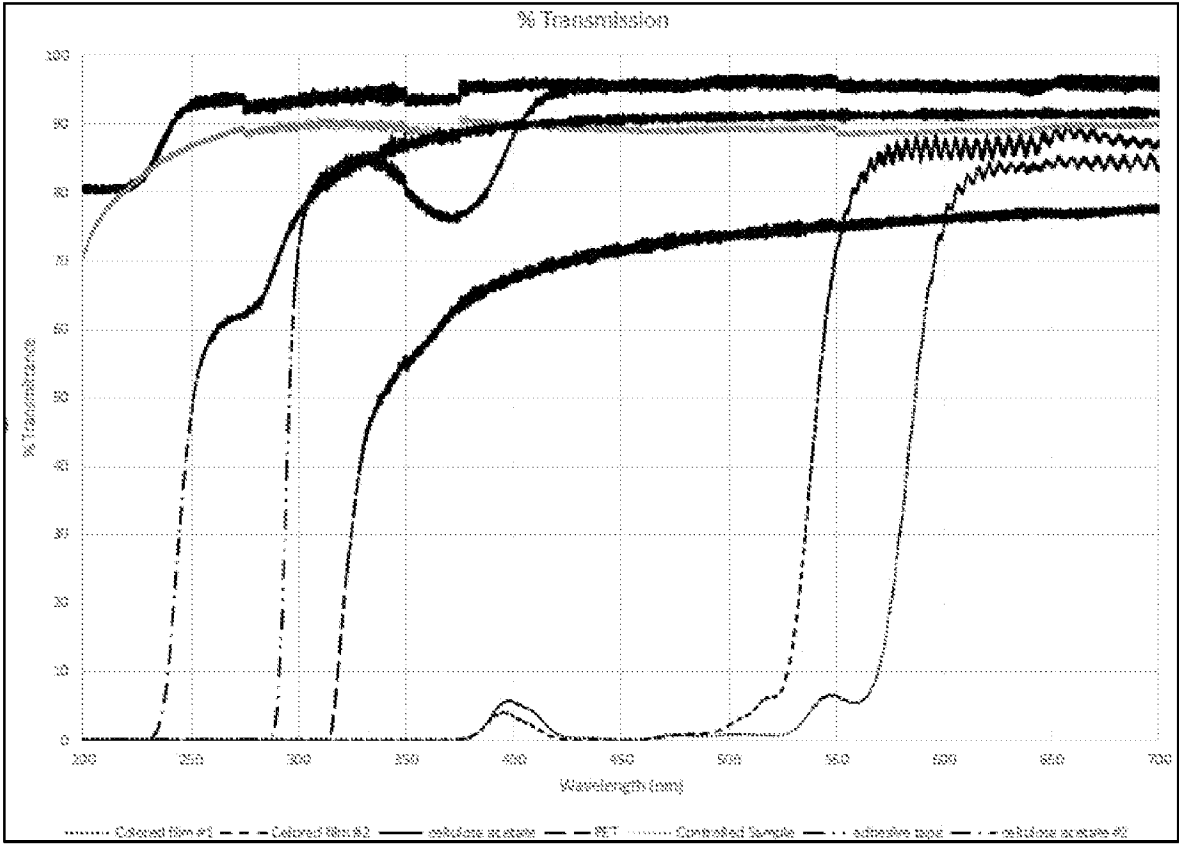
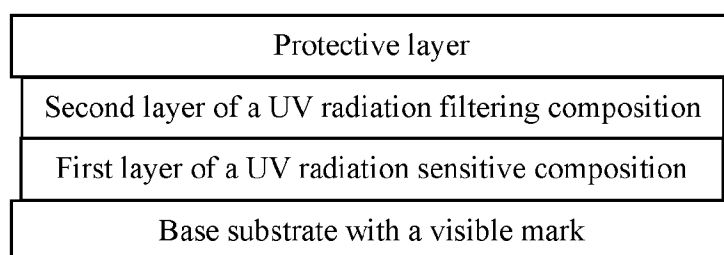


FIG. 1

**FIG. 2**

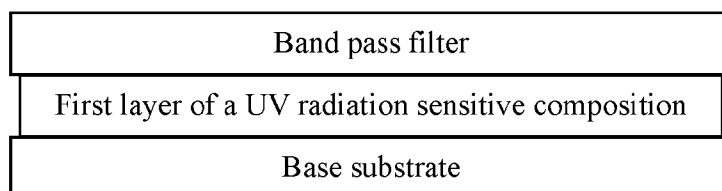
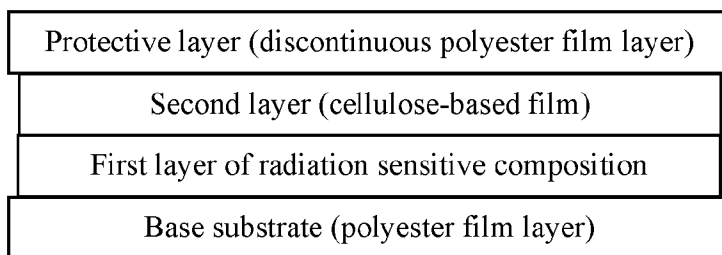


FIG. 3

**FIG. 4**

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 23/17827

A. CLASSIFICATION OF SUBJECT MATTER

IPC - INV. G01T 1/08 (2023.01)

ADD. A61N 5/08, G03C 1/815, G01J 1/02, G01J 1/50 (2023.01)

CPC - INV. G01T 1/08

ADD. A61N 5/06, G03C 1/815, G01J 1/02, G01J 1/50, A61N 2005/0661

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)

See Search History document

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

See Search History document

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

See Search History document

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X --- Y	US 2021/0270978 A1 (ISP INVESTMENTS LLC) 02 September 2021 (02.09.2021) entire document, especially Fig. 1, 2 and 11-13; para [0061], [0062], [0064]-[0066], [0069], [0071], [0072], [0081], [0100], [0103]	1, 3, (5-9,15-17,24,25, 34-37,39,41,43-45,52-54)/(1,3) 2, 4, (5-9,15,16,24,25, 34-37,39,41,43-45, 55-57)/(2,4), 10-14, 18-23, 26-33, 38, 40, 42, 46-51
Y	WO 2021/130155 A1 (AMS SENSORS GERMANY GMBH) 01 July 2021 (01.07.2021) entire document, especially Fig. 1a; pg. 5, ln 32-34; pg. 6, ln 1-6; pg. 8, ln 14-16	2, 4, (5-9,15,16,24,25, 34-37,39,41,43-45,55-57)/(2-4); 26-33, 38, 40, 42
Y	WO 2021/222784 A1 (ISP INVESTMENTS LLC) 04 November 2021 (04.11.2021) entire document, especially para [0050], [0052], [0054], [0055], [0058]	10-14
Y	US 2019/0375948 A1 (ESSILOR INTERNATIONAL) 12 December 2019 (12.12.2019) entire document, especially para [0066], [0067], [0084], [0090]	18-23
Y	US 2022/0001062 A1 (MICROTEK, INC.) 06 January 2020 (06.01.2022) entire document, especially para [0006], [0058], [0061], [0070], [0072], [0104]	46-51

☐ Further documents are listed in the continuation of Box C.☐ 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

"D" document cited by the applicant in the international application

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

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

04 August 2023

Date of mailing of the international search report

AUG 30 2023

Name and mailing address of the ISA/US

Mail Stop PCT, Attn: ISA/US, Commissioner for Patents

P.O. Box 1450, Alexandria, Virginia 22313-1450

Facsimile No. 571-273-8300

Authorized officer

Kari Rodriguez

Telephone No. PCT Helpdesk: 571-272-4300

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 23/17827

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

This application contains the following inventions or groups of inventions which are not so linked as to form a single general inventive concept under PCT Rule 13.1. In order for all inventions to be examined, the appropriate additional examination fees must be paid.

Group I: Claims 1-57 directed to a multilayer radiation dosage indicator.

Group II: Claims 58-61 directed to a process for making a radiation dosage indicator.

The inventions listed as Groups I and II do not relate to a single general inventive concept under PCT Rule 13.1 because, under PCT Rule 13.2, they lack the same or corresponding special technical features for the following reasons:

--see extra sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:
1-57

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 23/17827

Continuation of Box No. III – Observations where unity of invention is lacking

Special Technical features:

The invention of Group II includes the special technical feature of a process for making a multilayer radiation dosage indicator comprising: selecting a base substrate, not required by the claims of Group I.

Shared Technical features:

The inventions of Groups I-II share the technical features of (a) a base substrate comprising a visible mark; (b) at least one first layer on top of said base substrate comprising a radiation sensitive composition capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold; (c) at least one second layer on top of said first layer of ultraviolet radiation filtering composition wherein said second layer comprises a viewing zone through which said visible mark on said base substrate is viewable depending on opacity of said first layer prior to exposure to said ultraviolet radiation; and (d) an optional protective layer that's optionally transparent to said ultraviolet radiation. Specifically, Groups I and II are related as an apparatus (Group I) and process (methods) for making the apparatus (Group II). The apparatus is known in prior art as shown in US 2021/0270978 A1 to ISP Investments LLC (hereinafter ISP). Therefore, Groups I and II lack unity since the shared technical features do not represent a contribution over ISP:

ISP discloses a multilayer radiation dosage indicator ([abstract]- a multi-ply radiation dosage indicator) comprising: (a) a base substrate (12) comprising a visible mark (14, 14A) (Fig. 1 and 2, para [0061]- Indicator 10 is formed as a multi ply laminate including a first ply 12 having two visible indicia 14 and 14A thereon; para [0072]- each of the component plies of indicator 10 can be made of plastic, coating on substrate, or paper or other suitable sheet materials); (b) at least one first layer (16, 16A) on top of said base substrate comprising a radiation sensitive composition (Fig. 1 and 2, para [0061]- A second ply, which is a combination of 16 and 16A, of transparent material is provided which defines a radiation sensitive ply having two radiation sensitive zone or surface 18 and 18A disposed in overlying relation to the indicia 14 and 14A respectively of first ply 12...Suitable for use as the second ply 16 and 16A are films of a radiation sensitive polyacetylenic system that provides a color change caused by polymerization of a polyacetylene material) capable of changing opacity in response to exposure to an ultraviolet radiation dosage exceeding a predetermined threshold (para [0061]- Radiation sensitive zone 18 and 18A are capable of changing opacity in response to exposure to radiation exceeding a predetermined threshold so as to change the visibility of indicia 14 and 14A; para [0103]- For a radiation sensitive layer which is responsive to ultra-violet and which would be used for measurement of ultra-violet dosage); (c) at least one second layer (26) on top of said first layer of ultraviolet radiation filtering composition (Fig. 1 and 2, para [0066]- if desired, of a fourth ply 26 constituting an optical filter overlying the radiation sensitive zones of 18 and 18A; para [0100]- The inventive indicator can be utilized to measure a wide variety of types of electromagnetic radiation including low energy radiation, which is radiation having wave lengths within the visible range, ultra-violet...ply 26 and 26A must be adapted in terms of their thickness and composition so as to allow the specific type of radiation to which the radiation sensitive zone is sensitive and which it is desired to measure, to penetrate) wherein said second layer (26) comprises a viewing zone through which said visible mark on said base substrate is viewable depending on opacity of said first layer prior to exposure to said ultraviolet radiation (note: element 26 inherently included a viewing zone, since element 20 comprises a viewing zone, para [0062]- The third ply 20 is formed having a viewing zone in the form of a cut-out 22 and 22A positioned to permit exposure of indicia 14 and 14A there through; para [0064]- prior to use...the cut-out portion 22 and 22A of ply 20 so that the combined visible message on indicator 10 reads "NOT IRRADIATED."...as a result of exposing indicator 10 to a radiation treatment, the opacity of radiation sensitive zone 18 or 18A changes...zone 18 or 18A becomes sufficiently opaque to make indicia 14 or 14A non-visible. As a result, the visible message on indicator 10 now constitutes only indicia 24 and 24A to indicate that the indicator, and the substrate to which it is attached, has become "IRRADIATED."); and (d) an optional protective layer (19) that's optionally transparent to said ultraviolet radiation (para [0081]- As shown in FIG. 2 which is a view similar to FIGS. 11, 12, or/and 13, protective transparent sheet 19 covers ply 20).

As the common features were known in the art at the time of the invention, they cannot be considered special technical features that would otherwise unify the groups.

Therefore, Groups I-II lack unity under PCT Rule 13.