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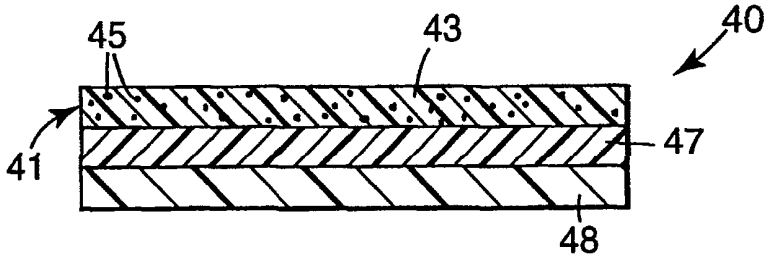
(54) Title: *IN-SITU* RADIOACTIVITY DETECTION



(57) Abstract: Detection materials for the *in-situ* detection of radioactivity. The detection material comprises at least one Scintillator material (45) distributed in adhesive material (48) and is sufficiently flexible to be wound and unwound without cracking or fracturing.

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IN-SITU RADIOACTIVITY DETECTION

Field of the Invention

The present invention relates to detection materials comprising at least one scintillator material for the in-situ detection of radioactivity. In another aspect, the present invention relates to methods of making and using such detection materials.

Background of the Invention

Radioactivity is the emission of energy in the form of alpha, beta, or gamma radiation. Such energy has been harnessed for a number of uses, including scientific research, nuclear power production, and medical procedures. Such radioactive materials, however, are inherently dangerous to living organisms. If accidentally spilled or improperly discarded, radioactive material can negatively impact the ecosystem. Therefore, it is desirable to have methods available to allow for the characterization and quantification of radioactive materials (e.g., contaminants) at a particular location for the proper maintenance, and possible removal, of such biologically hazardous materials.

The use of solid scintillation materials in polymers and paper is known. For example, U.S. patent application having Serial No. 08/975,007, filed November 20, 1997, reports a sheet article including a porous matrix or membrane comprising separators for radioactive species and scintillators which emit light in response to radioactive emissions from the radioactive species. The sheet material is useful in detecting and quantifying the radioactivity associated with liquid samples. U.S. Pat. No. 5,496,502 (Thomson) reports an adhesive plastic scintillator that can be attached to a solid support medium (e.g., sample wells in a microtiter plate). Radioactive samples are then introduced in the sample wells and the microtiter plate is then brought to a detector for radioactive coating.

Other methods for detecting radionuclides include scintillation counting and low background gas proportional counting.

Summary of the Invention

In one aspect, the present invention provides detection material for in-situ detection of radioactive materials (i.e., detection of radioactive materials on a surface and/or in a substrate without removal of the radioactive material or detection material). The detection material comprises scintillator material distributed (typically uniformly distributed) in matrix material (typically an adhesive material), wherein the scintillator material, when excited by radioactivity, emits light (i.e., electromagnetic radiation having at least a portion of its spectrum in the range from 10 to 30,000 nanometers) that is detectable externally to the detection material for in-situ detection of radioactive material. Optionally, the detection material comprises two or more different scintillator materials.

Typically, detection material according to the present invention comprises two or more different scintillator materials that emit light when excited by at least two of, more preferably each of, alpha, beta, or gamma radiation. For example, the detection material may include one scintillator material that emits light when excited by alpha radiation, and a second scintillator material that emits light when excited by beta radiation. Further, for example, the detection material may include a first scintillator material that emits light when excited by gamma radiation, and a second scintillator material that emits light when excited by beta radiation.

Certain preferred detection materials according to the present invention are flexible. For example some preferred detection materials according to the present invention are sufficiently flexible to be wound once around a 2.5 centimeter (preferably, 1 cm, more preferably, 3 mm, or even 1 mm) diameter rod, and then unwound without cracking or fracturing the detection material.

One preferred detection material according to the present invention is a tape comprising a backing having a first major surface and a second opposed major surface, adhesive material attached to the first major surface, and a first scintillator material, wherein the first scintillator material, when excited by radioactivity, emits light that is detectable externally to the tape for in-situ detection of radioactive material. The adhesive material, the backing, or both, may comprise the scintillator material. Alternatively, or in addition, scintillator material may be present as another layer or coating on the tape, and/or as another layer or coating of the tape. In some embodiments, at least a majority, by weight, of the scintillator material is distributed in either the adhesive material or the

backing material. In some embodiments according to the present invention, the backing material comprises a first scintillator material and the adhesive material has distributed therein a second, different scintillator material.

In another aspect, the present invention provides a method of in-situ detection of radioactive material comprising:

applying a detection material comprising scintillator material distributed in matrix material distributed therein to a surface including radioactive material, wherein the scintillator material emits light when excited by radioactivity that is detectable externally to the detection material for in-situ detection of the radioactive material; and

detecting the light emitted from at least a portion of the scintillator material by a light-detecting device for in-situ detection of the radioactive material.

In yet another aspect, the present invention provides an assembly for in-situ detection of radioactive material comprising:

a substrate having a first major surface that includes radioactive material; and

a tape attached to the substrate, the tape comprising a backing having a first major surface and a second, opposed major surface, adhesive material attached to the first major surface, and scintillator material, wherein the scintillator material emits light when excited by radioactivity that is detectable externally to the tape for in-situ detection of the radioactive material.

Detection materials according to the present invention and methods of making and using the such materials offer a number of advantages over conventional techniques for identifying radioactive contamination. For example, embodiments of detection materials according to the present invention allow for the detection of radioactive contamination from solid or vapor states in-situ without collection, dispersion, and concentration of samples. Further, embodiments of detection materials according to the present invention (particularly flexible tape and sprayed-on detection materials) allow for the detection of radioactive contamination in and/or on rough surfaces such as concrete or brick.

Brief Description of the Drawing

FIG. 1 is a cross-sectional view of a layer of a detection material according to the present invention;

FIG. 2 is a cross-sectional view of a tape according to the present invention;

FIG. 3 is a cross-sectional view of a tape according to the present invention attached to a flat radioactive surface;

FIG. 4 is a side view of a tape according to the present invention attached to a sneaker; and

FIG. 5 is a perspective view of a canister spraying a detection material composition onto a potentially radioactive surface.

Detailed Description

Detection materials according to the present invention emit light (i.e., electromagnetic radiation having at least a portion of its spectrum in the range from 10 to 30,000 nanometers) when excited by radioactivity such as at least one of alpha, beta, or gamma radiation. The radioactive material from which the radioactivity is emitted may be comprised of one or more different radioactive sources (e.g., one radioactive source may be emitting alpha radiation, and other beta or gamma radiation).

Referring to FIG. 1, layer of detection material 31 comprises scintillator material (e.g., particles 35) distributed in matrix material 33. Optionally, detection material 31 includes other components such as plasticizers, fillers, and rheology modifiers. Preferably, matrix material 33 is a pressure sensitive adhesive material that can be applied to a surface via a spray. Layer of detection material 31 preferably is or can be applied or formed over rough, cracked, curved, or otherwise irregular surfaces, although of course it is also useful on smooth or planar surfaces as well.

Referring to FIG. 2, tape 40 includes detection layer 41, backing 47, and adhesive material 48. Detection layer 41 comprises scintillator material 45 distributed in matrix material 43. FIG. 3 shows another tape according to the present invention 50 applied to flat surface 54. Tape 50 comprises backing 51 and detection material 53. Radioactive particles 55 are shown emitting radioactivity 57. For such particles underneath tape 50, the scintillator material in detection layer 53 is excited by

radioactivity 57 and emits light 56 (which is transmitted through translucent (with respect to the wavelength(s) of light emitted by the scintillator material(s) used), preferably transparent, backing 51).

FIG. 4 illustrates tape 50 may be applied to a curved surface such as the heel of sneaker 61. Certain preferred detection material (including tape and sprayed-on material) according to the present invention are flexible such that they are capable of being wound once around a 2.5 cm (preferably, 1 cm, more preferably, 3 mm, or even 1 mm) diameter rod and then unwound without cracking or fracturing the detection material.

Although tapes 40 and 50 are shown to have the scintillator material in one layer of the construction, it should be understood that one or more scintillator materials (including the same or different) scintillator materials can be present in a different layer and/or multiple layers of a tape construction. For example, the scintillator material may be present in the backing itself. It is typically preferred, however, that the tape backings are substantially free of scintillator material, and that an adhesive layer of the tape comprises at least one scintillator material. Optionally, a tape according to the present invention includes two or more adhesive layers.

The particular construction of detection material according to the present invention (including the relative amounts of the components of the detection material) may depend, for example, on the particular application, the raw materials used to the detection material and/or the amounts of such raw materials.

Preferably, a tape according to the present invention comprises from about 1 to about 90 weight percent scintillator material, based on the combined weight of the matrix material (and/or adhesive material) and scintillator material.

Matrix material suitable for detection materials according to the present invention include adhesive materials such as pressure sensitive adhesives and hot melt adhesives. Pressure sensitive adhesives are normally tacky at room temperature and can be adhered to a surface by application of light pressure. Preferably, the adhesive material has an average 90° peel adhesion value of at least 3 N/dm.

Examples of suitable adhesives useful in the practice of the present invention generally include compositions of thermoplastic elastomeric block copolymers, natural rubber, butyl rubber, polyisobutylene, silicones, polyalphaolefins, polyacrylates, and combinations thereof. Examples of useful thermoplastic elastomeric block

copolymers include ethylene-propylene-diene polymers and styrene-containing block copolymers such as styrene-isoprene, styrene-isoprene-styrene, styrene-butadiene-styrene, styrene-ethylene/butylene-styrene, and styrene-ethylene/propylene-styrene block copolymers. Other useful adhesive compositions include materials based on polyvinyl ethers; polychloroprenes; butadiene-acrylonitrile polymers; ethylene-containing copolymers such as ethylene vinyl acetate, lower alkyl (meth)acrylates (typically containing 1-8 carbons) homo, co- and ter-polymers; polyurethanes; polyamides; epoxies; polyvinylpyrrolidone and vinylpyrrolidone copolymers; polyesters; and mixtures of the above.

Optionally, the matrix material may comprise optional additives, such as coupling agents, fillers, expanding agents, fibers, antistatic agents, tackifiers, plasticizers, fillers, curing agents, suspending agents, photosensitizers, lubricants, wetting agents, surfactants, pigments, dyes, ultraviolet light stabilizers, anti-oxidants , and low boiling solvents (e.g., ethyl acetate). The amount of these materials is selected to provide the desired application properties, adhesive properties, and stability.

Specific examples of useful pressure sensitive adhesives include tackified thermoplastic elastomeric block copolymers, tackified silicones, tackified natural rubber, and inherently tacky (meth) acrylate copolymers. General descriptions of useful pressure-sensitive adhesives can be found, for example, in the Encyclopedia of Polymer Science and Engineering, Vol. 13, Wiley-Interscience Publishers (New York, 1988) and the Handbook of Pressure-Sensitive Adhesive Technology, 2nd ed., D. Satas, Ed., Van Nostrand Reinhold Co. (New York, 1982).

Such adhesives can be prepared by curing or polymerizing at least one monomer by a variety of polymerization methods to form a material having adhesive characteristics. Heat or radiation in combination with appropriate catalysts and/or photo-initiators may be used to cure such precursors. Radiation curing methods include visible light, ultraviolet radiation, and electron beam. Preferably, the pressure sensitive adhesives are polymerized by using ultraviolet light, such as those described in U.S. Pat. No. 4,181,752 (Martens).

Scintillator materials produce light when exposed to radioactivity (e.g., when near a source of radioactive particles that is emitting radioactivity). Such scintillator materials may be organic or inorganic in composition. The emitted light includes at least a

portion of its spectrum in at least one of the ultraviolet range (i.e., 10-390 nanometers), the visible range (i.e., 390-780 nanometers), or the infrared range (i.e., 780-30,000 nanometers). Scintillator materials may be present in the detection material as a solute and/or a particulate.

Examples of organic scintillator materials include benzoxazoles (e.g., 1,1'-biphenyl 4-yl-6-phenyl-benzoxazole, derivatives of the 2-phenylbenzoxazole such as 2-phenylbenzoxazole, 2-(4'-methylphenyl)-benzoxazole, 2-(4'-methylphenyl)-5-methylbenzoxazole, 2-(4'-methylphenyl)-5-t-butylbenzoxazole, 2-(4'-t-butylphenyl)-benzoxazole, 2-phenyl-5-t-butyl-benzoxazole, 2-(4'-t-butylphenyl)-5-t-butylbenzoxazole, 2-(4'-biphenylyl)-benzoxazole, 2-(4'-biphenylyl)-5-t-butylbenzoxazole, 2-(4'-biphenylyl)-6-phenyl-benzoxale, derivatives of 2-phenylbenzoxazole such as those disclosed in Donald L. Horrocks, Organic Scintillators, Proceedings of the International Symposium on Organic Scintillators, Argonne National Laboratory, p. 406, (1966); oxazoles (e.g., 2-p-biphenylyl-5-phenyloxazole (CAS Registry No., 852-37-9, Aldrich Chemical Co., Milwaukee, WI); 2,2' -p-phenylenebis (5-phenyloxazole) (CAS Registry No.: 1806-34-4, Aldrich Chemical Co.), 2,5-diphenyloxazole (CAS Registry No.: 92-71-7); oxadiazoles (e.g., 2,5-diphenyloxadiazole) (CAS Registry No.: 725-12-2), as well as derivatives from the 1,3,4- oxadiazole (e.g., 2,5-diphenyl-1,3,4-oxadiazole, 2-(4'-t-butylphenyl)-5-phenyl-1,3,4-oxadiazole, 2,5-di-(4'-t-butylphenyl)-1,3,4-oxadiazole, 2-phenyl-5-(4"-biphenylyl)-1,3,4-oxadiazole, and 2-(4'-t-butylphenyl)-5-(4"-biphenylyl)-1,3,4-oxadiazole; terphenyls (e.g., 4,4"-di-tert-amyl-p-terphenyl (CAS Registry No.: 121838-04-8); polynuclear aromatics (e.g., 4,4'-bis (2,5-dimethylstyryl) diphenyl (CAS Registry No.: 72814-85-8) and p-terphenyl (CAS Registry No.: 92-94-4); pyrozolines (e.g., 1-phenyl-3-mesityl-2-pyrazoline (CAS Registry No.: 60078-97-9), 1,5-diphenyl-3-(4-phenyl-1,3-butadienyl)-2-pyrazoline (CAS Registry No.: 59715-47-8), 1,5-diphenyl-beta-styrylpyrazoline (CAS Registry No.: 2515-62-0); phosphoramides (e.g., anilinobis (1-aziridinyl) phosphine oxide (CAS Registry No.: 6784-53-8); and thiophenes (e.g., derivatives of the benzoxazalyl-thiophene series such as: 2,5-bis-benzoxazolyl(2')-thiophene; 2,5-bis-[5'-methylbenzoxazolyl(2')]-thiophene, 2,5-bis-[4', 5'-dimethylbenzoxazolyl (2')]-thiophene, 2,5-bis-[4', 5'-dimethylbenzoxazolyl (2')]-3,4-dimethylthiopene, 2,5-bis-[5'-isopropylbenzoxazolyl (2')]-3,4-dimethylthiophene, 2-benzoxazolyl (2')-5-[7'-secbutyl-benzoxazolyl(2')]-thiophene, 2-benzoxazolyl(2')-5-[5'-t-butyl-benzoxazolyl(2')]-thiophene,

2,5-bis-[5'-t-butylbenzoxazolyl(2')]-thiophene; and combinations thereof. Other suitable organic scintillator materials for use in the practice of the present invention are disclosed in Donald L. Horrocks, Organic Scintillators, Proceedings of the International Symposium on Organic Scintillators, Argonne National Laboratory, p. 485, (1966).

Examples of suitable inorganic scintillator materials include doped crystalline scintillators (e.g., NaI(Tl), CsI(Tl), and undoped crystalline scintillators (e.g., Cs, I, BaF_2, CeF_3, yttrium aluminate, and $Bi_4Ge_3O_{12}$). Preferred inorganic scintillator materials include yittrium aluminate, bismuth germinate, and combinations thereof. Other inorganic scintillator materials suitable for use in the present invention are disclosed in Scintillator and Phosphor Materials, Material Research Society Symposium Proceedings, Materials Research Society, Pittsburgh, PA, vol. 348, p. 5 (1994). Many of the above-mentioned organic and inorganic scintillator materials are available from Aldrich Chemical Co., Milwaukee, WI.

Detection materials according to the present invention can be made by any of a number of techniques, including techniques disclosed herein, as well as by techniques, which may be apparent to those skilled in the art, after they review the disclosure of the present invention.

Detection material according to the present invention can be made, for example, by combining curable material(s) with scintillator material(s), applying the resulting mixture to a substrate as desired (e.g., coating the mixture into a tape backing), and then curing the curable material. Typically a curing agent (e.g., a photo-initiator or a catalyst) is added to the curable material before the scintillator material is added. Certain preferred methods for making detection material according to the present invention may comprise the steps of partially curing the curable material(s) to form a pre-polymer syrup, uniformly dispersing the scintillator material in the syrup, and then completing the cure of the matrix material. Although the preferred amount of scintillator material present in a detection material may vary, depending, for example, on the material, the scintillator material, other components of the detection material, and the application (e.g., the type of radioactivity to be detected), typically, the detection material comprises in the range of about 1 to about 90 percent by weight scintillator material, based on the combined weight of the matrix and scintillator materials.

Tapes according to the present invention (which include the detection material or layer) can be made using the general teachings in the art for making tapes except the tapes of the present invention also include scintillator materials present in the tape to allow for the in-situ detection of radioactive material.

A layer(s) of detection material according to the present invention can also be made in-situ by, for example, spraying or brushing a sprayable or brushable composition comprising curable material and scintillator material onto a desired surface and then curing the curable material. The sprayable or brushable composition can be sprayed, for example, using an aerosol canister (including a can or a bottle) system or a mechanical pump (e.g., a finger pump) canister system. Referring to FIG. 5, sprayable composition 71 is shown being sprayed from canister 77 onto surface 73. The material sprayed from canister 71 includes curable matrix material (e.g., an adhesive material), scintillator material, and propellant. Suitable propellants should be apparent to those skilled in the art and may include fluorocholorohydrocarbons; chlorofluorocarbons; a mixed system of propane and butane (liquefied natural gas); dimethyl ether and the like. Propellants that avoid environmental problems are preferred.

It is also within the scope of the present invention to make the matrix material by preparing a liquid mixture that when dried or the liquid therein is evaporated provides the matrix material. The liquid mixture can be a solution or dispersion. Optionally, such a liquid mixture can be utilized in preparing a sprayed-on detection material.

Optionally, a light enhancing material (e.g., light enhancement film) may be used to enhance the detectability of light emitted from scintillator material. For example, a light enhancement film may be placed on top of, or incorporated into, a detection material to focus the light emitted from scintillator material present in the detection material. In one embodiment, for example, a tape according to the present invention may utilize light enhancing film as a backing material.

Examples of light enhancement materials or devices include light redirecting sheet material such as that available under the trade designations "3M IMAGE DIRECTING FILM (IDF) II", "3M TRANSMISSIVE RIGHT ANGLE FILM (TRAF) II", and "3M BRIGHTNESS ENHANCEMENT FILM (BEF) II" from the 3M Company of St. Paul, MN.

Other suitable light redirecting sheet material may be apparent to those skilled in the art after reviewing the disclosure of the present application.

Optionally, detection materials according to the present invention may be used in conjunction with sorptive and/or containment media suitable for handling radioactive materials. For example, sorptive powder materials may be incorporated directly into matrix materials along with the scintillator materials to provide a containment constructions capable of also detecting the presence of radioactive material (e.g., radioactive contamination). Alternatively, or in addition, higher sorptive capacities can be provided by laminating sheets of sorptive media to detection materials according to the present invention. Suitable sorptive media sheets can comprise, for example, fibrous cellulosic materials, fibrous nonwoven polymeric webs of polyolefins, (e.g. polyethylene, polypropylene, or copolymers thereof) polyacrylonitrile, fibrillated polymer webs comprising polytetrafluoroethylene, nonwoven inorganic fibrous webs of glass or ceramic materials, or fibrous pulps comprising poly(p- or m-phenyleneterphthalamide) or chemical modifications thereof, optionally further comprising sorptive or reactive particles enmeshed within the fibrous webs. Examples of sorptive fibrous materials suitable for use in the practice of the present invention are further described in U.S. Pat. No. 5,637,506 (Goken, et. al.). The use of apertured or discontinuous sorptive sheets in such laminate constructions can facilitate observation of scintillation events associated with the presence of radioactive material (e.g., radioactive contamination) which might otherwise be obscured from one side of the construction by an opaque sorptive sheet.

Typically, detection materials according to the present invention are attached to, or otherwise placed near, an area suspected to contain radioactivity. A device capable of measuring and/or quantifying light emissions can then be placed over the tape and the amount of light emitted from the detection material or tape can then be determined. Suitable light measuring devices preferably contain an optical radiation detector such as a photomultiplier tube, charge coupled device, or a photodiode array. Other types of photoelectric cells or light-sensing devices may also be used to detect and quantify scintillator material light output.

This invention is further illustrated by the following examples, but the particular materials and amounts thereof recited in these examples, as well as other conditions and details, should not be construed to unduly limit this invention. Various

modifications and alterations of the invention will become apparent to those skilled in the art. All parts and percentages are by weight unless otherwise indicated.

Examples

Test Procedures

90° Peel Adhesion Test

90° Peel adhesion properties of pressure sensitive adhesive (PSA) tape constructions according to the present invention were determined using a Sintech Instron (Model Renew 1122, available from Sintech, Research Triangle Park, NC). Test samples were prepared according to specifications included in the 90° Peel test procedure description included in the Sintech Instron operating manual. The operating crosshead speed was 305 mm/minute, with a full scale load of 400 grams. The 90° peel adhesion data is reported in Newtons per decimeter (N/dm).

Radioactivity Detection

The ability of scintillator material containing PSA tape constructions according to the present invention to detect radiation was evaluated by subjecting tape samples to a variety of different radioactive sources, and measuring the resulting luminescence. A Ludlum Model 182 Sample Counter with a photomultiplier tube, (available from Ludlum Measurements, Inc., Sweetwater, TX) was used to detect light emitted from the tape. A Ludlum Model 2000 scaler (available from Ludlum Measurements, Inc.), operating at 820 volts, was used to count the detected light events. Detected light count events were corrected for a background luminescence of about 400 counts per minute.

Samples of adhesive scintillator constructions (4 cm X 4 cm) were attached to a radioactive source and the laminate placed in a Ludlum Model 182 counter. The counter was closed for approximately 5 minutes to allow for dark adaptation, and 5 readings, each of 0.1 minute duration, were taken. The count data reported is an average of five readings, which have been corrected for background counts. The uncertainty associated with a given measurement was estimated to be less than 10%.

Radioactive isotopes used in this evaluation included a sealed Tc-99 source (technetium, 11.89 nCi, obtained from Isotope Products Laboratory, Burbank, CA), and

three laboratory-prepared sources. These sources were prepared by applying a 0.5 ml, 0.2 ml, and 0.1 ml aliqouts of a diluted aqueous Tc-99 standard (obtained from Isotope Products Laboratory, Burbank, CA) to stainless steel planchets (5.08 cm X 5.08 cm, available from A.F. Murphy Die & Machine Co., Inc., North Quincy, MA) and air drying the coating. The dried sources respectively displayed activities of 47.1 nCi of Tc-99 (942-1), 18.9 nCi of Tc-99 (942-2), and 9.43 nCi of Tc-99 (942-3).

Pre-Polymer Syrup Preparation

A pre-polymer syrup was prepared from a mixture of 94 parts isooctyl acrylate, 6 parts acrylic acid (obtained from Aldrich Chemical, Milwaukee, WI), and 0.04 part benzil dimethyl ketal (a photoinitator obtained under the designation "KB-1" from Sartomer Co., Exton, PA). The mixture was placed in a container, purged with nitrogen, sealed in the container under a nitrogen atmosphere, and then irradiated under an ultraviolet black light (obtained under the trade designation "GE F15T8 BL" from General Electric Co., Farfield, CT) until the resulting pre-polymer syrup had a visually estimated viscosity of approximately 3,000 centipoise. 0.1 part benzil dimethyl ketal was added to the syrup. The mixture was stirred to produce a uniform solution. The resulting solution was stored in the dark under nitrogen atmosphere.

Examples 1-10

Scintillating PSA tape constructions according to the present invention were prepared by coating scintillator containing pre-polymer mixtures on a polyester backing and then curing the resulting coating.

Coatable, curable scintillator containing compositions were prepared by adding scintillator material to the pre-polymer syrup (described above) in the proportions indicated in Table 1, below, and stirring to produce a uniform mixture.

Table 1

Example	Scintillator Material	Parts Scintillator Material	Parts Pre-Polymer Syrup	Adhesion, N/dm
1	Yttrium aluminate[1]	5	95	30.8
2	Yttrium aluminate[1]	10	90	29.1
3	Yttrium aluminate[1]	15	85	26.3
4	Yttrium aluminate[1]	20	80	23.8
5	2,5-diphenyloxazole[2]	20	80	Not tested[4]
6	2,5-diphenyloxazole[2]	40	60	Not tested[4]
7	2,5-diphenyloxazole[2]	60	40	Not tested[4]
8	Bismuth germinate[3]	20	80	18.6
9	Bismuth germinate[3]	40	60	10.5
10	Bismuth germinate[3]	80	20	7.0

1. Obtained from Rexon Components, Inc., Beachwood, OH.
2. Obtained from Aldrich Chemical Company, Milwaukee, WI.
3. Obtained from Rexon Components, Inc.
4. Adhesion of sample was so low that sample was not tested.

The pre-polymer mixture was knife coated (0.10 millimeter thick coating) onto a biaxially oriented poly(ethylene terephthalate) film (0.05 millimeter thick) and covered with a silicon release agent coated poly(ethylene terephthalate) film. The coated syrup was cured by passing the film sandwich between two banks of fluorescent black lamps (obtained from General Electric Co., Farfield, CT), one above and one below the film sandwich. The black lamps had 90% of their emission spectra between 300 and 400 nm, with a maximum at about 350 nm. In addition, the black lamps had a light intensity of 2.2 mW/cm^2 for 3.5 minute exposure.

Adhesive and scintillation performance of the Example 1-10 tape constructions were determined, as described above. The results are reported in Table 1, above.

The data in Table 1 illustrates that scintillator PSA tape constructions can be prepared that retain good adhesive properties with up to 80 weight percent scintillator material in the adhesive. It is believed, for example, that scintillator materials having densities greater than that of bismuth germinate can be used to construct PSA tape compositions having greater than 80 weight percent scintillator material.

The ability of PSA scintillator compositions according to the present invention to detect radiation is believed to be influenced, for example, by the amount of scintillator material in the adhesive composition, as is illustrated, for example, by the average net count data presented in Table 2, below, for Examples 1-4.

Table 2

Example	Scintillating Material	Radioactive Source	Source Activity, nCi	Average Net, CPM
1	5% Yttrium aluminate	Tc-99 sealed source	11.89	60
2	10% Yttrium aluminate	Tc-99 sealed source	11.89	330
3	15% Yttrium aluminate	Tc-99 sealed source	11.89	550
4	20% Yttrium aluminate	Tc-99 sealed source	11.89	790

In this series (shown in Table 2, above), the yttrium aluminate content of the tape samples increased from 5 to 20 weight percent, while the activity of the radiation source remained constant.

A second set of Examples 1-4 tape samples was subsequently tested against three different radiation sources of decreasing source activity. The results are reported in Table 3, below.

Table 3

Example	Scintillating Material	Radioactive Source	Source Activity, nCi	Average Net, CPM
1	5% Yttrium aluminate	942-1	47.1	554
1	5% Yttrium aluminate	942-2	18.9	200
1	5% Yttrium aluminate	942-3	9.43	142
2	10% Yttrium aluminate	942-1	47.1	1210
2	10% Yttrium aluminate	942-2	18.9	506
2	10% Yttrium aluminate	942-3	9.43	244
3	15% Yttrium aluminate	942-1	47.1	2316
3	15% Yttrium aluminate	942-2	18.9	954
3	15% Yttrium aluminate	942-3	9.43	576
4	20% Yttrium aluminate	942-1	47.1	3170
4	20% Yttrium aluminate	942-2	18.9	1264
4	20% Yttrium aluminate	942-3	9.43	634

The data in Table 3 shows that increasing the amount of inorganic scintillator material in an adhesive layer of the tapes resulted in an increase in the measured counts detected by the counting device. As the source activity of the radioactivity decreased, the measured counts detected by the counting device also decreased for each tape. The data suggests a correlation between the concentration of radioactive particles within a radioactive source and light detected by the detection device.

The data in Table 4 (below) show that scintillator tape constructions (Examples 5-10) incorporating 2,5-diphenyloxazole or bismuth germinate exhibited scintillator material content/count measurement correlations similar to those exhibited by the yttrium aluminate-based constructions (Examples 1-4).

Table 4

Example	Scintillating Material	Radioactive Source	Source Activity, nCi	Average Net, CPM
5	20% 2,5-Diphenyloxazole	942-1	47.1	4134
5	20% 2,5-Diphenyloxazole	942-2	18.9	1636
5	20% 2,5-Diphenyloxazole	942-3	9.43	830
6	40% 2,5-Diphenyloxazole	942-1	47.1	17348
6	40% 2,5-Diphenyloxazole	942-2	18.9	7162
6	40% 2,5-Diphenyloxazole	942-3	9.43	3530
7	60% 2,5-Diphenyloxazole	942-1	47.1	19952
7	60% 2,5-Diphenyloxazole	942-2	18.9	7942
7	60% 2,5-Diphenyloxazole	942-3	9.43	3934
8	20% Bismuth germinate	942-1	47.1	1938
8	20% Bismuth germinate	942-2	18.9	804
8	20% Bismuth germinate	942-3	9.43	338
9	40% Bismuth germinate	942-1	47.1	4778
9	40% Bismuth germinate	942-2	18.9	1806
9	40% Bismuth germinate	942-3	9.43	904
10	80% Bismuth germinate	942-1	47.1	11966
10	80% Bismuth germinate	942-2	18.9	4644
10	80% Bismuth germinate	942-3	9.43	2474

Various modifications and alterations of this invention will become apparent to those skilled in the art without departing from the scope and spirit of this invention, and it should be understood that this invention is not to be unduly limited to the illustrative embodiments set forth herein.

What is claimed is:

1. A detection material for in-situ detection of radioactive materials, said detection material comprising scintillator material distributed in adhesive material, wherein said scintillator material, when excited by radioactivity, emits light that is detectable externally to said detection material for in-situ detection of radioactive material, and wherein said detection material is sufficiently flexible to be wound once around a 2.5 centimeter diameter rod, and then unwound without cracking or fracturing the detection material.

2. The detection material according to claim 1, wherein said adhesive material is pressure sensitive adhesive material.

3. The detection material according to claim 1, wherein said scintillator material is uniformly distributed in said adhesive material.

4. A tape for in-situ detection of radioactive material comprising a backing having a first major surface and a second opposed major surface, adhesive material attached to said first major surface, and a first scintillator material, wherein said first scintillator material, when excited by radioactivity, emits light that is detectable externally to said tape for in-situ detection of radioactive material.

5. The tape according to claim 4, comprising at least two different scintillator materials.

6. The tape according to claim 4 further comprising a second scintillator material, wherein said first scintillator material emits light when excited by alpha radiation from said radioactive material that is detectable externally to said tape; and wherein said second scintillator material emits light when excited by beta radiation from said radioactive material that is detectable externally to said tape, for in-situ detection of said radioactive material.

7. The tape according to claim 4 further comprising a second scintillator material, wherein said first scintillator material emits light when excited by gamma radiation from said radioactive material that is detectable externally to said tape, and wherein said second scintillator material emits light when excited by beta radiation from said radioactive material that is detectable externally to said tape, for in-situ detection of said radioactive material.

8. The tape according to claim 4 further comprising a second scintillator material, wherein said first scintillator material emits light when excited by alpha radiation from said radioactive material that is detectable externally to said tape; and wherein said second scintillator material emits light when excited by gamma radiation from said radioactive material that is detectable externally to said tape, for in-situ detection of said radioactive material.

9. The tape according to claim 4 further comprising a second scintillator material and a third scintillator material, wherein said first scintillator material emits light when excited by alpha radiation from said radioactive material that is detectable externally to said tape; wherein said second scintillator material emits light when excited by beta radiation from said radioactive material that is detectable externally to said tape; and wherein said third scintillator material emits light when excited by gamma radiation from said radioactive material that is detectable externally to said tape, for in-situ detection of said radioactive material.

10. The according to claim 4, wherein said scintillator material is uniformly distributed in said adhesive material.

11. The tape according to claim 4, wherein at least a majority, by weight, of said scintillator material is distributed in said adhesive material.

12. The tape according to claim 4, wherein at least a majority, by weight of said scintillator material is distributed in said backing.

13. The tape according to claim 4, said scintillator material is present in each of said backing material and said adhesive material.

14. The tape according to claim 4, wherein said backing material comprises a first scintillator material, and wherein said adhesive material comprises a second, different scintillator material.

15. The tape according to claim 4, further comprising a light-enhancing material.

16. The tape according to claim 4, wherein said tape comprises 1 to 90 weight percent of said scintillator material, based on the combined weight of the adhesive and scintillator materials.

17. The tape according to claim 4, which is flexible such that said tape can be wound once around a 2.5 centimeter diameter rod and then unwound without cracking or fracturing the tape.

18. The tape according to claim 4 wherein said adhesive material has an average 90° peel adhesion value of at least about 3 N/dm.

19. The tape according to claim 4 wherein said adhesive material is a pressure sensitive adhesive material.

20. A method of in-situ detection of radioactive material comprising:

applying a detection material comprising scintillator material distributed in a matrix material to a surface including radioactive material, wherein said scintillator material emits light when excited by radioactivity that is detectable externally to said detection material for in-situ detection of said radioactive material; and

detecting the light emitted from at least a portion of said scintillator material by a light-detecting device for in-situ detection of said radioactive material.

21. The method according to claim 20, wherein said scintillator material is uniformly distributed in said matrix material.

22. An assembly for in-situ detection of radioactive material comprising:

a substrate having a first major surface that includes radioactive material; and

a tape attached to said substrate, said tape comprising a backing having a first major surface and a second, opposed surface, adhesive material attached to said first major surface, and scintillator material, wherein said scintillator material emits light when excited by radioactivity that is detectable externally to said tape for in-situ detection of radioactive material.

23. The assembly according to claim 22 wherein at least a majority, by weight, of said scintillator material is distributed in said adhesive material.

24. The assembly according to claim 22, wherein at least a majority, by weight of said scintillator material is distributed in said backing.

25 The assembly according to claim 22, wherein said scintillator material is uniformly distributed in said adhesive material.

26 The assembly according to claim 22, wherein said tape, prior to being attached to said substrate, was sufficiently flexible to be wound once around a 2.5 centimeter diameter rod and then unwound without cracking or fracturing the tape.

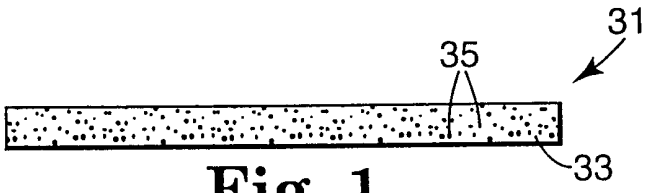

Fig. 1

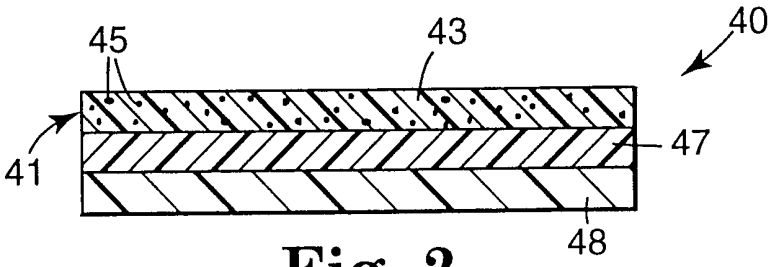

Fig. 2

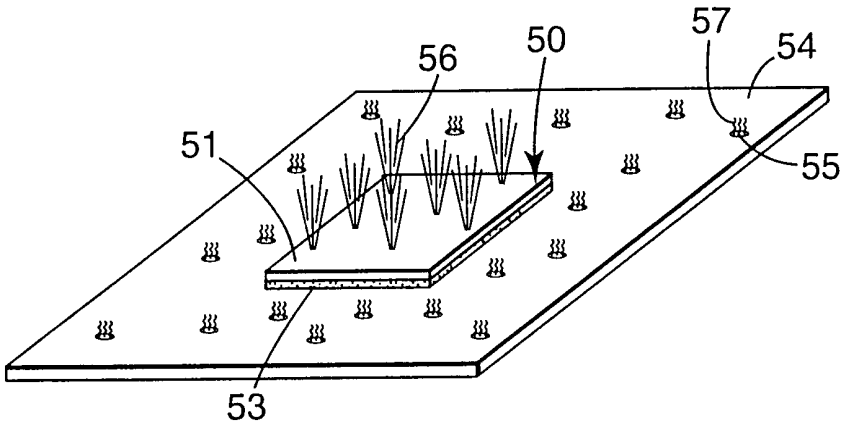

Fig. 3

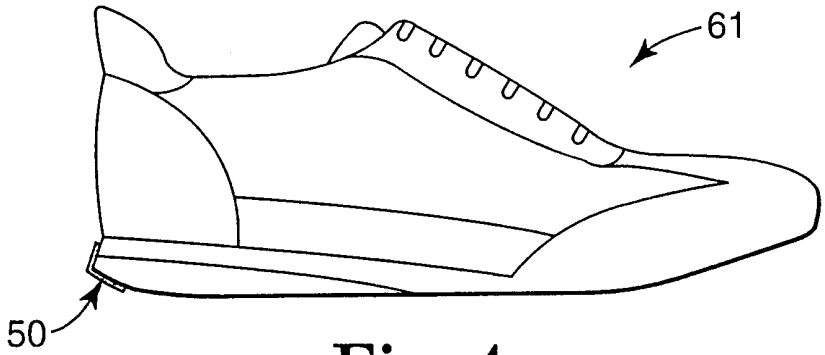

Fig. 4

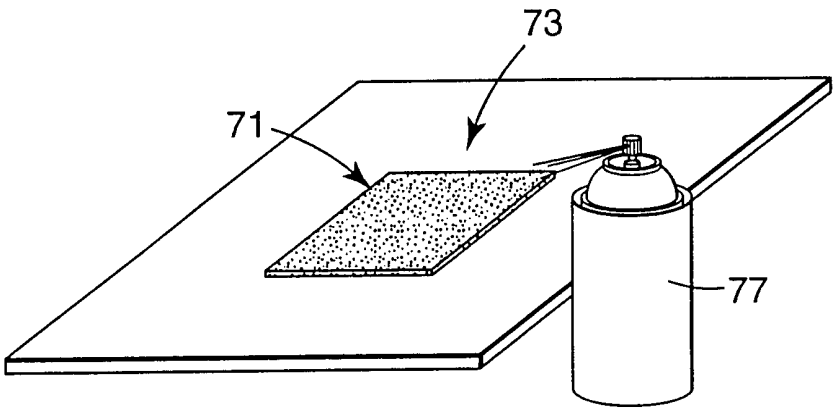

Fig. 5

INTERNATIONAL SEARCH REPORT

Inte ional Application No
PCT/US 99/23701

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 G01T7/00 G01T1/203

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)
IPC 7 G01T

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

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
EPO-Internal, PAJ, WPI Data, INSPEC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 583 343 A (DILMANIAN F AVRAHAM ET AL) 10 December 1996 (1996-12-10) abstract column 2, line 66 -column 3, line 67 figures	1,20,22
A	PATENT ABSTRACTS OF JAPAN vol. 1998, no. 06, 30 April 1998 (1998-04-30) & JP 10 039034 A (FUJII HARUO), 13 February 1998 (1998-02-13) abstract	1,4
A	US 3 678 271 A (GROEZINGER JOHN J) 18 July 1972 (1972-07-18) abstract column 2, line 19 - line 38 column 2, line 61 -column 3, line 50	1

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
15 August 2000	23/08/2000

Name and mailing address of the ISA	Authorized officer
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INTERNATIONAL SEARCH REPORT

Inte ional Application No
PCT/US 99/23701

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
A	US 3 952 204 A (DAVIS EARL V ET AL) 20 April 1976 (1976-04-20) abstract column 2, line 16 - line 29 column 3, line 7 - line 17	1
A	US 5 496 502 A (THOMSON JAMES) 5 March 1996 (1996-03-05) cited in the application page - column 3, line 56 -column 4, line 10 column 6, line 32 - line 52 figures	1,20,22

1

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INTERNATIONAL SEARCH REPORT

Information on patent family members

Inte onal Application No
PCT/US 99/23701

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JP 10039034 A	13-02-1998	NONE	
US 3678271 A	18-07-1972	NONE	
US 3952204 A	20-04-1976	NONE	
US 5496502 A	05-03-1996	NONE	