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(54) **Gaseous contaminant dosimeter**

(57) A personal dosimeter for collecting a quantity of a gaseous contaminant in proportion to its average ambient concentration over the time of collection consists essentially of a closed receptacle containing a gas-collection medium, a channel-containing diffusion device forming a part of the boundary of the receptacle and a porous, hydrophobic film in contact with the interior side of the barrier. After use, the gas collection medium is removed from the receptacle for determination of the contaminant.

FIG. 1

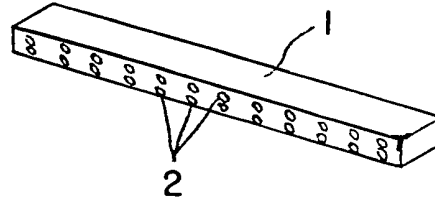


FIG. 2

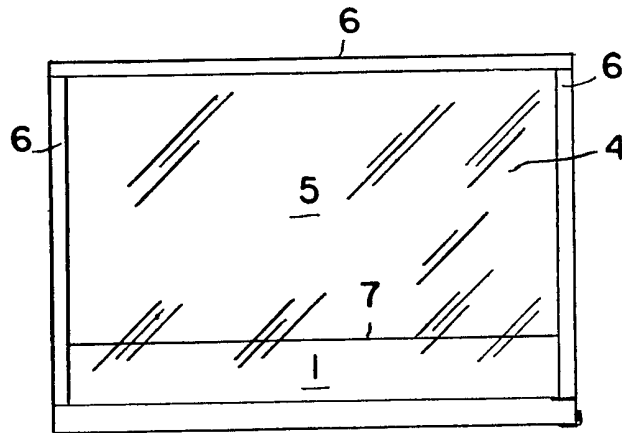


FIG. 3

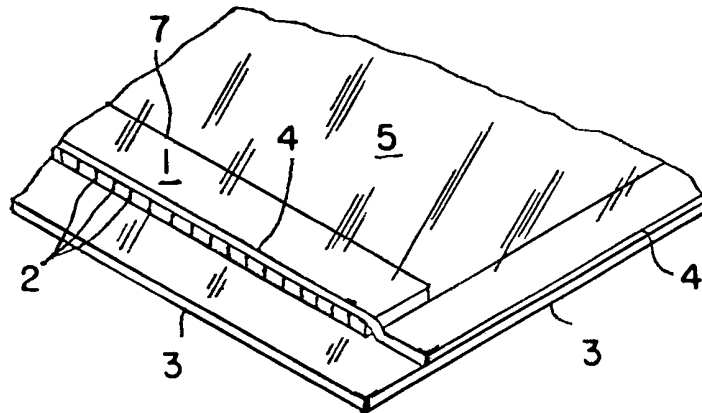


FIG. 4

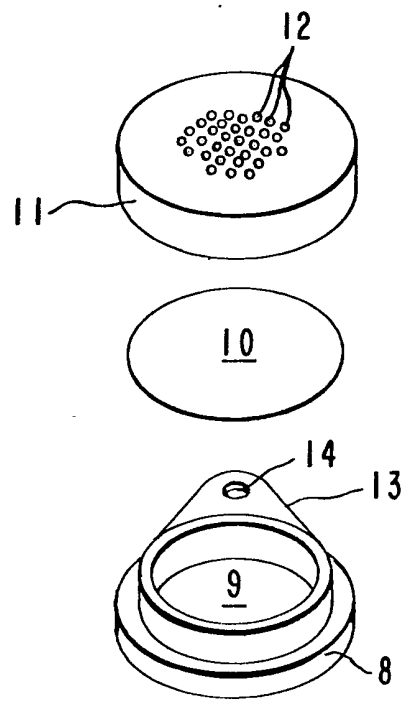
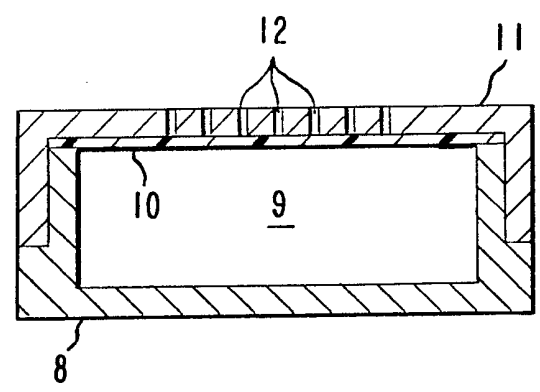


FIG. 5



SPECIFICATION

Gaseous contaminant dosimeter**5³ Background of the invention** 5*Field of the invention*

This invention is related to a personal dosimeter for registering gaseous contaminants in the atmosphere. More particularly, it is related to a dosimeter capable of collecting a quantity of contaminant in proportion to its average atmospheric concentration to aid in determining the integrated exposure level to the
10 contaminant. 10

Description of the prior art

In response to the increasing concern about the health of workers who are exposed to harmful pollutants in the air, it has become necessary to monitor the concentration of the air-borne contaminants. One
15 development for this purpose involved a rather large air pump which would force air to be sampled through a filter, trapping particulate contaminants. This obviously is unavailing for the monitoring of gaseous
contaminants and, even for particles, is not accurate to determine concentration of the particles in the
sampled atmosphere. 15

Personal sampling devices which are worn by individual workers and which passively collect the
20 contaminants have also been used. For example, a device which utilized the molecular diffusion of the gas to be monitored to collect the sample has been described in American Industrial Hygiene Association Journal, Volume 34, pages 78-81 (1973). This device and others like it, called impinging tubes, are often cumbersome
to use since their design and delicate construction necessitate that they always be oriented properly to
accurately sample the atmosphere and to prevent dislocation of the sampling mechanisms within the tube.
25 Therefore, there remains a need for a personal dosimeter that simply but accurately collects gaseous
contaminants in proportion to their average atmospheric concentration. 25

Summary of the invention

According to the present invention, there is provided a personal dosimeter for collecting a gaseous
30 contaminant in proportion to its average ambient concentration during the collection time where the
dosimeter consists essentially of 30

a closed receptacle;

a collecting medium for the gaseous contaminant within the receptacle;

a diffusion device, forming a part of the boundary of said receptacle, the device containing a plurality of
35 through-and-through channels adapted for the gaseous contaminant to diffuse therethrough from the
atmosphere to the interior of the receptacle, said channels each having a length-to-diameter ratio of at least 3
and said channel providing the only communication between the atmosphere and the interior of the
receptacle; and 35

a porous, hydrophobic, inert film covering the interior openings of said channels. 40

40

Brief description of the drawings

Figure 1 is a magnified perspective view of a diffusion device usable in the present invention.

Figure 2 is a top view of a gaseous contaminant dosimeter utilizing the diffusion device of *Figure 1*.

Figure 3 is a partial perspective view of the dosimeter of *Figure 2*.

45 *Figure 4* is an exploded view of another dosimeter of the present invention. 45

Figure 5 is a cross-sectional view of the assembled dosimeter of *Figure 4*.

Detailed description of the invention

The dosimeters of this invention collect a gaseous contaminant in proportion to its average concentration
50 in the atmosphere during the collection period and provide for the expedient determination of this
concentration. This is achieved by passively sampling the gaseous contaminant in ambient air in proportion
to its concentration therein by allowing the contaminant to diffuse into an interior portion of the dosimeter
where it is maintained, by a collecting medium situated therein, until it is analyzed. 50

The collecting medium holds the gaseous contaminant or its ions in a form that is more readily analyzable
55 than is the gaseous form. After collection, the medium is removed from the dosimeter and treated with
appropriate reagents to product color, the intensity of which is dependent upon the amount of gaseous
contaminant collected. The time-average ambient concentration can then be determined, as later explained,
with a previously-calibrated colorimeter or spectrophotometer. Alternatively, the contaminant can be
separated from the collecting medium and its quantity determined, for example, by gas chromatography
60 wherein the results of the gas chromatography analysis have been previously calibrated against known
time-average ambient concentrations of the contaminant. The preferred method of determination is
colorimetric. 60

Generally, the collecting medium is a material that absorbs, adsorbs, reacts or otherwise combines with
the gaseous contaminant being measured. Regardless of the manner in which the medium interacts, as
65 above, with the contaminant, the quantity or strength of the collecting medium in the dosimeter should be 65

sufficient to interact completely with the total quantity of gaseous contaminant which is anticipated to be collected. The collecting medium will often be specific to the particular gaseous contaminant being monitored. Examples, meant to be representative but not limiting, include aqueous solutions of oxidizing agents or triethanol amine to absorb nitrogen dioxide, solutions of potassium or sodium tetrachloromercurate to absorb sulfur dioxide, solutions of sulfuric or other acids to absorb ammonia, and distilled water or a solution of sodium bisulfite to absorb formaldehyde. Charcoal or powdered carbon of high surface area, powders of metals, or metal salts can be used to adsorb many other organic contaminants.

Methods for colorimetric analysis, for example, for sulfur dioxide, nitrogen dioxide, ammonia, or formaldehyde, in air, are described in National Institute for Occupational Safety and Health method numbers 160 (publication 121, 1975), 108 (publication 136, 1974), 205 (publication 121, 1975) and 125 (publication 136, 1974), respectively. The techniques therein described are readily adaptable with respect to absorbing solution and color-forming reagents for use in connection with collection by the dosimeter of the present invention.

One preferred embodiment of the present invention is shown in Figures 2 and 3 and is described and can be formed as follows. A base sheet 3 of impermeable polymeric material is provided to form one side of the receptacle portion of the dosimeter. The sheet is preferably transparent and thermoplastic and can be made of polymers of olefin, halogenated polymers, polyesters, or ionomer resins. Preferred materials are the ionomer resins shown in U.S. Patent No. 3,264,272 issued August 2, 1966 to R. W. Rees. They are the ionic copolymers of alpha-olefins and alpha, beta-ethylenically unsaturated carboxylic acids of 3-8 carbon atoms having 10-90% of the carboxylic acid groups neutralized with metal ions.

The size of sheet 3 is not critical but is preferably a size easily adaptable for use in a personal dosimeter which is to be worn or readily carried. The collecting medium is placed in the central portion of sheet 3. When the medium is a liquid, this can be accomplished by first forming a depression in the central portion of the sheet by applying pressure thereto with an appropriate die, heated or otherwise. The depressed area of sheet 3 is the central portion of the interior, designated 5, of the dosimeter.

After the collecting medium has been placed on sheet 3, a second top-sheet 4 corresponding to sheet 3 in composition and substantially in size is placed over sheet 3. Heat and pressure are then applied to the three areas 6 to provide permanent, fluid-tight bonding at the three corresponding edges of sheets 3 and 4. Adhesives or other forms of bonding can also be used provided the bonds are permanent and fluid-tight and the adhesive is inert to the collecting medium.

An elongate gas diffusion device 1 having a plurality of through-and-through channels 2 is positioned parallel and proximate to the fourth, unbonded edge of base sheet 3 and parallel and flush with the fourth, unbonded edge of top sheet 4. The open channels 2 of device 1 are thus oriented horizontally with respect to the plane of sheet 3 and perpendicularly with respect to the fourth edges of sheets 3 and 4. On the interior side 7 of device 1, covering the channel openings of this interior side, is a porous, hydrophobic film, described in more detail below. Diffusion device 1, thus placed between sheets 3 and 4, is bonded to the sheets by the application of heat and pressure or by use of adhesives which should be impermeable and chemically inert to the collecting medium.

The bond between diffusion device 1 and each of sheets 3 and 4 should be liquid-tight and air-tight, thus completely enclosing the interior 5 of the receptacle formed by sheets 3 and 4. The relative positions of diffusion device 1 and sheets 7 and 8 are such that the channels 2 provide the only communication between the atmosphere and the interior 5 of the receptacle.

It is also possible to form the dosimeter of Figures 2 and 3 saving the placement of the collecting medium, when it is a liquid, for last. In such a case, the dosimeter is otherwise formed as described above. The collecting medium can be placed by piercing top sheet 4 at an appropriate spot with a hypodermic needle and injecting a measured amount of the collecting medium into the interior. The hole made by the hypodermic needle can then be thermally sealed.

Diffusion device 1 allows the gaseous contaminant to diffuse through each of channels 2 according to Fick's Law, which is expressed in relevant form as

$$M = D \cdot C \cdot t \cdot A / L$$

where

M = quantity of gaseous contaminant transferred (mg)

D = diffusion coefficient of the gaseous contaminant through air (cm²/min)

C = concentration of the contaminant in the atmosphere (mg/cm³)

t = time of exposure (minutes)

A = cross-sectional area of the channel (cm²)

L = distance in direction of diffusion, herein channel length (cm)

Values of D for various gaseous contaminants are readily available from the literature. The purely

diffusional nature of the transfer of the gaseous contaminant through the channels, at a rate in linear proportion to its atmospheric concentration, provides the integrating character of the dosimeter.

Gas diffusion device 1 is preferably made from materials that are non-hygroscopic and both chemically and physically inert to the gaseous contaminant and to the collecting medium. Examples are polyethylene, polypropylene, polymers or copolymers of tetrafluoroethylene and hexafluoropropylene, and stainless steel. The above-named polymers are preferred since they can be easily injection-molded.

As can be seen from Fick's Law, the number and diameter of the channels affect the quantity of gaseous contaminant collected since they affect the total cross-sectional area available for transfer. The quantity of contaminant collected is also inversely proportional to the length of the channels. Although these parameters are not necessarily critical to the integrating operation of the diffusion device, it has been found that when each of the channels has a length-to-diameter ratio of at least 3, preferably at least 4.5, the dosimeter attains the desired insensitivity to relative atmospheric motion caused by wind or movement of the wearer. It has further been found that the use of about 5-500 channels, preferably 10-100 channels, each having a diameter of about 50-1000 microns and a length of about 1.0-25.0 mm, preferably 3.0-8.0 mm, provides a device that is sufficiently sensitive to low ambient contaminant concentrations but is still of a conveniently small size.

A porous, hydrophobic film of 15-1000 micron thickness is placed over the channel openings on the interior side of diffusion device 1, the side communicating with the interior of the dosimeter. The film can be made, for example, of polymers or copolymers of tetrafluoroethylene and hexafluoropropylene. The function of the film is to prevent the absorbing solution, if that form of collecting medium is used, from flowing into the channels of diffusion device 1 and to further reduce sensitivity to atmospheric motion. Accordingly, the porosity of the film and the size of its pores should be selected so that these functions are performed without interfering with the passage of the gaseous contaminant from the interior ends of the channels to the absorbing solution. That is, the diffusion of gaseous contaminant through this film should be significantly greater than the diffusion through the channels so that the overall rate of diffusion is essentially controlled only by the channels. It has been found that a film that is 50-80% porous with a pore size of 0.1-3.0 microns is sufficient for this purpose when channels as previously-described are used.

The diffusion device 1 of Figure 1 and the dosimeter of Figures 2 and 3 are an example of preferred embodiments of the present invention but the invention is not limited thereto. The diffusion device, for example, can be in the shape of a plug sealed into the face of either sheet 3 or sheet 4. Similarly, the receptacle of the dosimeter need not be pouch-like as hereinbefore described but, for example, could be in the form of a rigid cuvette.

Such an embodiment of the present invention is shown in Figures 4 and 5. With respect to these Figures, there is shown a personal dosimeter composed of a circular base cup 8 having cylindrical walls which define an open cavity 9. Base cup 8 is provided with flange 13, having circular opening 14, which permits the assembled dosimeter to be fastened easily to a person's clothing. A friction-fitting circular cap 11 tightly engages base cup 8 in a manner forming closure of cavity 9. Cap 11 has a plurality of channels 12 of circular cross-section extending therethrough towards cavity 9. The channel-containing cap 11 functions as a gas diffusion device following Fick's Law and therefore the number, length, and diameter of channels 12 of cap 11 are selected as previously described for channels 2 of diffusion device 1.

Cap 11 and base cup 8 are preferably made from materials that are non-hygroscopic and both chemically and physically inert to the gaseous contaminant and to the collecting medium. The materials described for construction of diffusion device 1 are equally preferable for use in making cap 11 and base cup 8.

A porous, hydrophobic, inert film 10 is placed over the channel openings on the interior face of cap 11. This film performs the same functions as earlier described with respect to the dosimeter of Figures 2 and 3. Film 10 preferably is 15-1000 microns in thickness and has a porosity of 50-80% with a pore size of 0.1-3.0 microns. It is preferably made of polymers or copolymers of tetrafluoroethylene or hexafluoropropylene, although any hydrophobic, inert material of the above physical characteristics is sufficient.

In assembled form, the friction-fit between base cup 8 and cap 11 is liquid-tight and air-tight, thus completely enclosing cavity 9 of cup 8. Channels 12 of cap 11 thus provide the only communication between the atmosphere and cavity 9 and the collecting medium therein. The collecting medium preferably completely fills cavity 9 and is retained therein, against seepage through channels 12, by film 10.

In use, a dosimeter of this invention is exposed to the air containing the gaseous contaminant for a period of time for which the average contaminant concentration is sought. When the collecting medium is an absorbing solution, for example, a measured amount of the solution is then withdrawn from the dosimeter by, for example, a hypodermic syringe.

When the analysis is to be made photometrically, the withdrawn absorbing solution is mixed with appropriate color forming reagents which change the color of the absorbing solution. The intensity of color so formed is dependent upon the amount of gaseous contaminant collected. Although it is often desirable to have a self-contained dosimeter, as shown in U.S. Patent 4,208,371, in which the reagents are contained in the dosimeter and no withdrawal of material is necessary, this is sometimes impractical. An example of this is where the reagents are highly acidic, as in the color-production for formaldehyde where the reagents are chromotropic acid and sulfuric acid. In such cases, it is difficult to package the reagents in a stable and safe form, and the simple dosimeter of the present invention is well suited for these applications.

The dosimeter of this invention can be calibrated to give a direct relationship between colorimetric or

spectrophotometric readings and average ambient concentration of the gaseous contaminant. This can be accomplished by following a calibration procedure similar to that described in U. S. Patent 4,208,371. In such a procedure, several dosimeters are exposed over a given period of time to various known concentrations of a contaminant for which calibration is sought. The dosimeters contain the same kinds and amounts of collecting medium. Spectrophotometric readings, for example, are determined for at least two dosimeters at each of several known concentrations, and a straight-line is plotted, using a least-squares analysis, through the data points thus obtained.

CLAIMS

- 10 1. A personal dosimeter for collecting a gaseous contaminant in proportion to its average ambient concentration during the collection time, the dosimeter consisting essentially of a closed receptacle; a collecting medium for the gaseous contaminant within the receptacle;
- 15 a diffusion device, forming a part of the boundary of said receptacle, the device containing a plurality of through-and-through channels adapted for the gaseous contaminant to diffus therethrough from the atmosphere to the interior of the receptacle, said channels each having a length-to-diameter ratio of at least 3 and said channels providing the only communication between the atmosphere and the interior of the receptacle; and
- 20 a porous, hydrophobic, inert film covering the interior openings of said channels.
2. The dosimeter of claim 1 in which said receptacle is a pliable pouch of polymeric material.
3. The dosimeter of claim 1 in which said receptacle consists essentially of a cup-like container having an open end and a cap engaging the container and covering the open end, and in which said diffusion is embodied by at least a portion of the cap.
- 25 4. The dosimeter of claim 1, 2, or 3 in which there are 5-500 channels, each having a diameter of 50-1000 microns and a length of 1.0-25.0 mm.
5. The dosimeter of claim 4 in which the channel length is 3.0-8.0 mm and there are 10-100 channels.
6. The dosimeter of any of claims 1 to 5 in which the film has a thickness of 15-1000 microns, a porosity of 50-80%, and a pore size of 0.1-3.0 microns.
- 30 7. The dosimeter of claim 6 in which the collecting medium is an absorbing solution.
8. The dosimeter of claim 7 in which the absorbing solution is for formaldehyde.
9. A dosimeter as claimed in claim 1, substantially as described with reference to Figures 1-3 or Figures 4 and 5 of the accompanying drawings.