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Complete Specification entitled (54) RADON DETECTION.

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Related Art (56) 259,315(19,305/62) 85.2; 00.8.

The following statement is a full description of this invention, including the best method of performing it known to us:

19682/71—L

W. G. Murray, Government Printer, Canberra

X401-98-2D-15P.C.

Radon-222 is a radioactive gas which emanates from uranium ore. It is a member of the radioactive series which begins with uranium and ends with stable lead-206. Radon-220 or thoron is another form of radon gas which arises from thorium-232. Radon-222 has a half-life of 3.82 days, and radon-220 has a half-life of 56 seconds. Both of these gases emit alpha particles as they decay into daughter products in their respective radioactive series. The invention is applicable to any form of alpha-emitting gas, but is of primary interest with radon-222 because the radon-222 half-life is sufficient to permit this gas to diffuse and travel substantial distances through ore bodies.

Radon-222 (hereinafter "radon") decays to a chain of daughter products which are members of the uranium radioactive series. The radioactive daughter products of immediate interest are radium-A (polonium-218, half-life 3.05 minutes), radium-B (lead-214, half-life 26.8 minutes), radium-C (bismuth-214, half-life 19.7 minutes), and radium-C' (polonium-214, half-life 164 microseconds). Radon, radium-A and radium-C' emit alpha particles when they decay, and radium-B and radium-C emit beta and gamma radiation upon decaying. Radon and its alpha-emitting daughters radium-A and radium-C' are of specific interest as possible health hazards to miners and other personnel who work in atmospheres containing above-normal concentrations of radon.

The problem of radiation exposure arising from breathing a radon-containing atmosphere is extensively discussed in a booklet titled "Control of Radon and Daughters in Uranium Mines and Calculations on Biologic Effects" published in 1957 by the U. S. Department of Health, Education and Welfare (Public Health Service Publication No. 494). Recent investigations supplementing the disclosures in this booklet suggest that an observed higher-than-normal incidence of lung cancer among uranium-ore miners may be due to alpha radiation from radon inhalation and the retention of alpha-emitting radon daughters in the lungs and other tissues. It is thus important to monitor the atmosphere in work spaces such as uranium mines which may have an above-normal concentration of radon (and hence alpha radiation) due to the presence of large quantities of the uranium parent material.

Detection and monitoring is also of interest in prospecting for bodies of uranium ore. Radon is created in such ores, and (because it is a gas) has the ability to diffuse through the soil away from the ore toward the surface of the earth. If the ore deposit is fairly shallow (say within 100 feet -- 1 foot = 12 x 2. 54 cm -- of the surface), and the soil overburden is sufficiently porous to permit reasonably rapid gas diffusion, radon emanated from the ore has a sufficient half-life to move to the surface where it can be detected. Uranium prospectors can thus locate subsurface ore bodies by searching for surface areas which show a radon concentration higher than the normal background level (a small amount of uranium is present in normal soil, and a background level of radon is therefore also present in normal soil and the atmosphere).

In the past, various kinds of electronic instrumentation have been used to detect radon. Such equipment is expensive, bulky, usually requires trained operators, and may require special sample-collection techniques. Photographic film has also been used to detect radon because the light-sensitive emulsion of the film will record tracks of alpha particles emitted by radon and some of its daughters. Such film, however, must be packaged in light-tight containers, and must be processed using darkroom techniques. Photographic film is thus inconvenient and relatively expensive to use. There thus exists a need for a simple and inexpensive way to detect radon and to monitor its concentration. This need is met by the method and apparatus of this invention.

Summary of the Invention

We have found that the presence and concentration of radon and its alpha-emitting daughters can be effectively detected and monitored using a process usally called "track etching". This process employs a homogeneous track-registration material which can be a crystalline solid such as mica, or a non-crystalline substance such as inorganic glass or a polymeric plastic. If the detector material is irradiated with charged particles, minute damage "tracks" are created in the material by local alteration of the material structure along the particle trajectories. The damage tracks can be enlarged and made visible by application of a reagent which preferentially attacks the altered material (forming the damage track) at a faster rate than it attacks the unaltered material around the track.

Certain track-registration materials such as cellulose plastics register damage tracks when subjected to alpha radiation, but do not register beta- and gamma-radiation damage tracks.

This selective-sensitivity property is useful in radon dosimetry because radon alpha radiation is considered to be a possible cause of lung cancer whereas beta and gamma radiation from radon daughters does not present this hazard. Cellulose nitrate is an especially useful material for selective registration of alpha damage tracks, and is inexpensive and readily obtainable. Etching to enlarge the damage tracks is quickly and easily accomplished in daylight conditions using a common reagent such as six-normal sodium hydroxide.

The range of alpha particles in air is relatively short. For example, the alpha particle emitted by radium-C' has an energy of 7.68 MeV, and a range in air of about 6.5 centimeters. If an alpha-track-registering material is isolated by being positioned a distance greater than this range from alpha-emitting solid bodies such as an ore body, any tracks which are formed in the material will arise from the motion (as by convection or diffusion) of a radioactive alpha-emitting gas from a distance source to the material. The half-life of radon is sufficient to permit transportation over a considerable distance before radioactive decay occurs, and radon is the only alpha-emitting gas normally found in appreciable quantities in the neighborhood of uranium ore bodies. Alpha tracks on an isolated track-registration material will thus be formed only by radon or its alpha-emitting daughters, and the material is therefore useful as a radon detector and dosimeter.

The isolated alpha-track-registering material is positioned in a mine to monitor the mine atmosphere (or radon and its daughters), or can be carried by a miner in a badge-type dosimeter. In the uranium-prospecting application of the invention, one or more sheets of track-registering material are buried in the ground in simple containers. After a suitable exposure period, the material is etched to enlarge and make visible the alpha tracks, and the tracks are counted to obtain a measure of the presence and concentration of radon and its alpha-emitting daughters. The entire procedure is carried out in daylight conditions using inexpensive and readily available materials, and sensitive detection of radon is made possible.

Briefly stated, the invention expressed in method terms is a process for detecting radon and alpha-emitting daughter products of radon in an atmosphere. A track-registration material is positioned in the atmosphere at a location spaced from any adjacent alpha-emitting ore bodies a distance greater than a transit range of alpha particles in the atmosphere, whereby the material is exposed to alpha particles emitted by a gaseous source only. The track-registration material is substantially insensitive to light and has a property of forming radiation damage tracks along paths in the material traversed by alpha particles. After an exposure period, the material is etched with a reagent such as sodim hydroxide which selectively attacks and enlarges the damage tracks whereby the tracks are made sufficiently visible to be counted. Preferably, the track-registration material is a cellulose plastic such as cellulose nitrate which forms damage tracks only along paths traversed by alpha particles when irradiated by alpha, beta and gamma radiation.

In terms of apparatus, the invention includes a housing, which is readily penetrable by gases and is substantially impervious to solid matter whereby only gases circulate through the housing. A body of track-registration material is disposed within the housing for exposure to the gases. The material is substantially insensitive to light and has the property of forming damage tracks along paths traversed by alpha particles.

In one form of the invention, a body of calibration material such as uranium glass is secured to and overlays a portion of the track-registration material. The calibration material has the property of emitting alpha particles at a known rate whereby the covered portion of the track-registration material is exposed to a known amount of alpha radiation.

The invention also contemplates the use of a thin cover film secured to and overlaying a portion of the track-registration material. The cover film has the property of impeding alpha particles with energies of about 6.5 MeV or less so that such particles do not form damage tracks. The covered portion of the track-registration material is thereby made selectively sensitive only to high-energy alpha particles as emitted by radium-C'.

Brief Description of the Drawings

The invention will be described in detail with reference to the attached drawings, in which:

FIG. 1 is a sectional view of a mine tunnel, showing a radon detector positioned to monitor radon in the mine atmosphere;

FIG. 2 is an elevation, partly broken away, of a housing suitable for positioning a track-registration material in a mine atmosphere;

FIG. 3 is a plan view of a holder for a sheet of track-registration material;

FIG. 4 is a plan view of a glass slide with a coating of track-registration material and a calibration plate;

FIG. 5 is a side view of the slide shown in FIG. 5;

FIG. 6 is a schematic diagram showing a complete system for continuous monitoring and recording of radon in mine atmospheres;

FIG. 7 is an elevation of a radon dosimeter badge;

FIG. 8 is a side view of the badge shown in FIG. 8; and

FIG. 9 shows the use of the invention in uranium-ore prospecting.

Description of the Preferred Embodiments

Referring to FIG. 1, a radon detection and monitoring assembly 10 is shown positioned on the floor of a tunnel 11 in a uranium mine. Assembly 10 is shown in detail in FIGS. 2 and 3, and includes a cylindrical screen or wire-mesh housing or cage 12 having a wire-mesh bottom portion 13 and being open at its upper end. Assembly 10 is supported by a plurality of legs 14 secured to bottom portion 13.

A C-shaped bracket 16 is positioned in cage 12, and has a lower arm 17 secured to bottom portion 13 by a bolt and nut 18.

The bracket has an upper arm 19 which extends horizontally at the open upper end of the cage. A lid or cover 20 closes the open end of the cage, and is secured to the upper arm of the bracket by a flanged bolt 21 and a nut 22 fastened to the upper arm.

A clamp member 24 extends from bracket 16 midway between the upper and lower arms, and includes a slot 25. A holder 26 for a sheet of track-registration material 27 is secured in the slot by a clamp screw 28. The track-registration material is secured to holder 26 by a pair of clips 29. The holder includes a central aperture 30 so the mine atmosphere penetrating wire-mesh cage 12 will reach both faces of the track-registration material.

Another form of a track-registration material holder 33 is shown in FIGS. 4 and 5, and includes a baseplate 34 such as a conventional glass microscope slide. A film of track-registration material 35 such as cellulose nitrate is coated on one face of the baseplate, or may be in the form of a separate sheet of material which is cemented or otherwise secured to the baseplate by any convenient means. A body of calibration material 36 such as a small sheet of uranium glass is secured to one end of holder 33 by a U-shaped spring clip 37. Holder 33 is positioned in assembly 10 in the same manner as holder 26 described above.

In use, assembly 10 is positioned at any convenient location in a mine shaft or tunnel, and the atmosphere in the mine readily penetrates wire-mesh cage 12. Radon gas present in the mine atmosphere will be manifested by the formation of damage tracks in the track-registration material positioned inside the cage. As discussed above, these damage tracks are formed by alpha particles emitted during the radioactive disintegration of radon and its alpha-emitting daughter products radium-A and radium-C'.

After a suitable exposure period of say an eight-hour working shift, the track-registration material is removed from assembly 10 and processed with an etchant such as six-normal sodium hydroxide to make visible the damage tracks. The material can be immersed in the etchant, or the etchant can be swabbed on the surface of the material, and either process is carried out in daylight conditions as the material is not sensitive to light. Etching time is related to etchant temperature, and typically ranges from less than one minute at 70°C., to perhaps several hours at room temperature.

The etched tracks are then counted under a microscope, and the number of tracks per unit area on the track-registration material is a measure of the concentration of radon and its alpha-emitting daughters in the mine atmosphere. The track-registration material serves as a dosimeter or total-dose monitor because it integrates or accumulates the total alpha radiation to which it is subjected during the exposure period.

Calibration of the track-registration material is readily accomplished during the exposure period by covering a portion of the material with a known source of alpha-emitting material. This technique is incorporated in holder 33 shown in FIGS. 4 and 5. Calibration material 36 is preferably a uranium glass having a known content of uranium of perhaps 0.03 percent. The alpha emission from such a uranium glass is known, and that portion of track-registration material 35 covered by the calibration material is thus subjected to a known amount of alpha radiation during the exposure period. The tracks in this calibration zone are counted to provide a standard against which the number of tracks in the uncovered area of the track-registration material can be compared.

The range of alpha particles in a track-registration material such as cellulose nitrate is in the range of about thirty to sixty microns. The track is not visible under normal optical magnification before etching. However, after the track is enlarged by etching with sodium hydroxide or some other suitable reagent, the track is greatly enlarged and is readily visible under a microscope.

In some monitoring applications, it may be desirable to measure only the high-energy alpha particles emitted by radium-C'. This kind of selective sensitivity is easily accomplished by securing a thin plastic cover film 38 (see FIGS. 4-5) over a portion of the exposed surface of track-registration material 35. The thickness and other characteristics of the cover film are chosen such that lower-energy alpha particles (say less than about 6 or 6.5 MeV) emitted by radon and radium-A do not have quite enough energy to pass through the cover film and form damage tracks in material 35. The higher-energy particles (7.68 MeV) emitted by radium-C', however, have sufficient energy to penetrate the cover film and to form damage tracks in material 35.

Discrimination against lower-energy alpha particles is thus possible on a portion of material 35, and incident alpha particles from radon and radium-A and -C' will be detected on an adjacent uncovered portion of the material. The cover film is cemented or clamped in place during the exposure period, and is stripped off the material before etching. This kind of selective measurement is often useful in circulating atmospheres when secular equilibrium of radon and its daughter products has not been achieved.

An automated radon-detection system 40 for mine atmospheres and the like is shown in schematic form in FIG. 6. A long strip of track-registration material 41 such as cellulose nitrate is coiled on a feed spool 42 disposed in a housing case 43. The strip of material is secured to a takeup spool 44 which is rotated by a spring motor or electric motor (not shown). A pair of spaced-apart partition walls 46 having slots 47 are secured inside case 43, and a fine wire mesh or screen 48 is secured between the walls. The space enclosed by the screen and the walls is an exposure zone, and the strip of material passes through slots 47 to be exposed to the mine atmosphere as it is moved between the feed and takeup spools which form a transport means.

After the track-registration material emerges from the exposure zone, it passes over a guide means such as a set of guide rollers 50 to be immersed in an etching solution 51 such as sodium hydroxide in a tank 52 positioned in case 43. After etching, the strip is transported through a track-detection system such as a photometer having a light source 54 and a photoelectric detector 55 positioned on opposite sides of the strip. 39,974/68

A conventional recorder 56 records the output of the photoelectric detector to provide a permanent record of track density on the etched track-registration material, and thus a record of radon concentration in the mine atmosphere. Preferably, recorder 56 includes an alarm which is triggered automatically as a personnel warning when radon concentration exceeds a predetermined maximum safe level. The various components of the photometric measuring system, strip transport apparatus, recorder and alarm are of conventional design, and, for brevity, will not be described in detail. The system is also useful with the enclosed transport apparatus and strip material alone (without the etching bath and measuring means) for long-term time-correlated monitoring of mine atmospheres, with etching being performed as a separate subsequent step in the process.

The invention is shown in the form of a dosimeter badge 60 in FIGS. 7 and 8, and the badge includes a holder 61 having a spring clip 62 secured thereto for fastening the badge to the user's clothing. Holder 61 has a recessed cavity 63 and a sheet or strip of track-registration material 64 such as cellulose nitrate is positioned in the cavity through a slit 65 in the holder. A fine wire mesh or screen 66 is secured to the holder to cover the cavity whereby dust particles in the mine atmosphere are prevented from reaching the track-registration material. Radon gas passes freely through screen 66, and alpha particles emitted by the gas and its daughter products are detected and recorded on the track-registration material. The material is removed after a suitable exposure period and processed by etching as described above.

FIG. 9 shows the use of the invention in exploratory prospecting for uranium ore. A plurality of housings or cannisters 70 are buried in soil 71 at horizontally spaced-apart locations. A rod 72 is secured to each cannister, and extends above the surface to mark the locations of the cannisters. A flag 73 is secured to the upper end of each rod to make it readily visible. The "planting" process can be partially automated by using equipment such as a power-driven post-hole digger, to cut the cannister holes.

Each cannister 70 includes a simple spring clip 75 or a similar conventional clamp, and a sheet of track-registration material 76 is positioned in the clip. Holder 33 described above and illustrated in FIGS. 4 and 5 is also convenient for use in these cannisters, and can be mounted in clip 75. Cannisters 70 are perforated, or can include screened portions 77 to permit free diffusion of soil gases into the cannisters to contact the track-registration material.

Radon gas present in the soil gas will cause alpha tracks to be formed in the track-registration material, and the presence and concentration of radon in the soil is thus detected. Higher-than-normal concentrations of radon are indicative of an underlying deposit of uranium ore. The use of a plurality of cannisters permits construction of a radon-concentration map of the prospected area.

Cannisters 70 can be of inexpensive construction, and are conveniently formed as metal or plastic cans. The track-registration material is positioned far enough from screen portions 77 such that alpha particles (typically having a range less than seven centimeters in air) from the soil around the cannister will not have sufficient range to reach the material. Tracks are thus formed in the material only by gas emanating from the soil and passing through the screen portions to reach the track-registration material, and a true measure of radon gas in the soil is thus obtained.

The track-etching material is a sensitive detector of radon and its alpha-emitting daughters. For example, a safe working level in mine atmospheres is presently thought to be about 300 pico-curies of radon daughter products per liter of air. This concentration will result in the formation of about 15,000 alpha tracks per square centimeter of track-registration material in a one-month exposure period. Much shorter exposure periods are of course suitable, as individual tracks can be observed during microscopic examination of the etched material.

The invention is not restricted to the use of cellulose nitrate as a track-registration material, and other materials such as cellulose acetate or cellulose acetate butyrate which register alpha tracks can also be used. Cellulose nitrate is a presently preferred material as it appears to have the greatest sensitivity to alpha radiation. Any alpha tracks which may have been formed in the material (prior to the intentional exposure period) from background radon normally present in the air can be "erased" by heating the material to its softening point whereby the latent tracks are removed. For example, heating a sheet of cellulose-nitrate track-registration material to about 100° Centigrade is adequate to remove unetched latent tracks. The thickness of the track-registration material is preferably at least twenty microns such that the full length of the alpha trajectory is recorded in the material.

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The Claims Defining the Invention are as follows:-

1. A method for detecting radon and alpha-emitting daughter products of radon in an atmosphere, characterized in that it comprises the steps of:

positioning a track-registration material in the atmosphere at a location spaced from any adjacent alpha-emitting ore bodies a distance greater than a transit range of alpha particles in the atmosphere, whereby the material is exposed to alpha particles emitted by a gaseous source only, the material being substantially insensitive to light and having a property of forming damage tracks along paths in the material traversed by alpha particles; and

etching the material after exposure with a reagent which selectively attacks and enlarges the damage tracks whereby the tracks are made sufficiently visible to be counted.

2. The method defined in claim 1 characterized in that the track-registration material has a selective-sensitivity property of forming damage tracks only along paths in the material traversed by alpha particles when irradiated by alpha, beta and gamma radiation.

3. The method defined in claim 2 characterized in that track-registration material is cellulos nitrate.

4. The method of claims 1 to 3 are applied to prospecting for uranium ore in earth, characterized by the steps of:

burying a track-registration material in the earth for exposure to radon gas emanating from the ore, the material having property of forming damage tracks along paths in the material traversed by alpha particles emitted by the radon;

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recovering the track-registration material after exposure; and etching the exposed material with a reagent which selectively attacks and enlarges the damage tracks whereby the tracks are made sufficiently visible to be counted.

5. The method defined in claim 4 characterized in that the track-registration material is substantially insensitive to light.

6. The method defined in claim 5 characterized in that the reagent is sodium hydroxide.

7. The method defined in claim 4 characterized in that a plurality of track-registration materials are buried at spaced-apart locations in a region and subsequently recovered and etched, whereby a map of radon distribution in the region can be constructed.

8. Apparatus for detecting radon and alpha-emitting radon daughter products, in accordance with the method of claims 1 to 7 characterized by a housing defining an enclosed space, the housing being readily penetrable by atmospheric gases and substantially impervious to solid matter whereby only gases circulate through the enclosed space; and a body of track-registration material disposed in the enclosed space of the housing for exposure to the gases, the material being substantially insensitive to light and having the property of forming damage tracks along paths in the material traversed by alpha particles.

9. The apparatus defined in claim 8 further characterized by transport means in the housing and including a feed spool and a takeup

spool for transporting the track-registration material through the enclosed space, the material being in the form of an elongated strip coupled to an extending between the spools.

10. The apparatus defined in claim 9 further characterized by a tank in the housing and adapted to hold an etching reagent, and means for guiding the material through the tank for etching of the damage tracks after the material is exposed to gases in the enclosed space.

11. The apparatus defined in claim 10 further characterized by comprising measuring means in the housing between the tank and the takeup spool for detecting and measuring the pressence of etched damage tracks in the track-registration material.

DATED this 28th day of June, 1968.
GENERAL ELECTRIC COMPANY.

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Fellow Institute of Patent Attorneys of Australia
of EDWD. WATERS & SONS
SYDNEY

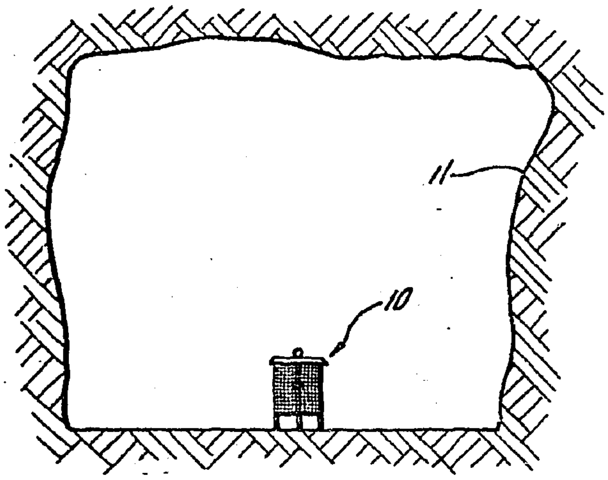


FIG_1

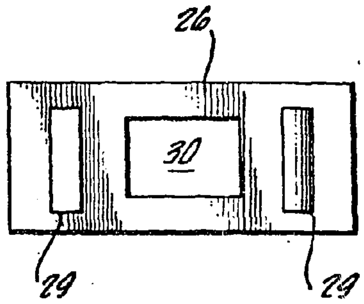


FIG_3

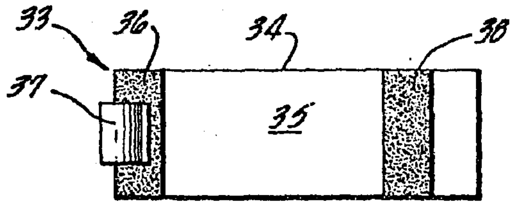
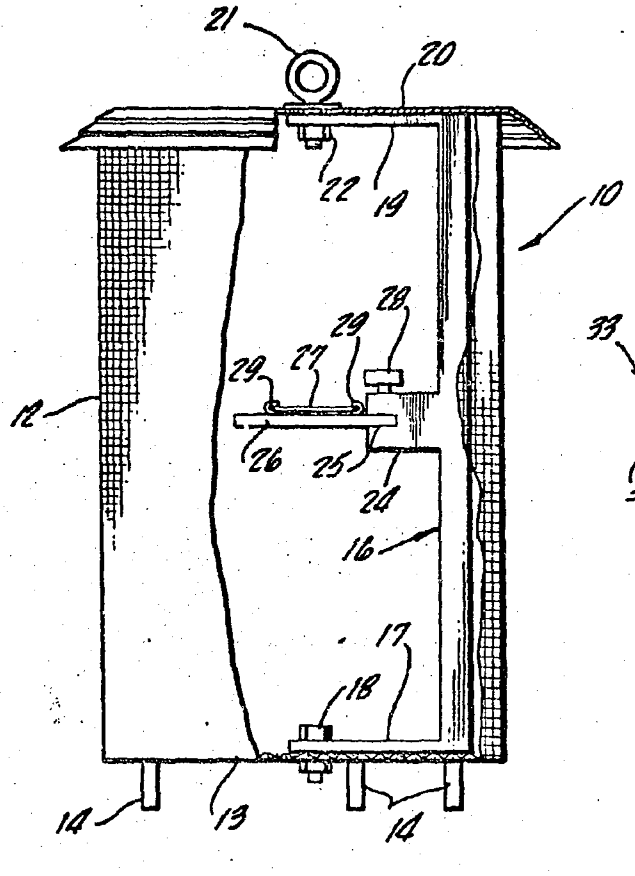


FIG_4

FIG_2



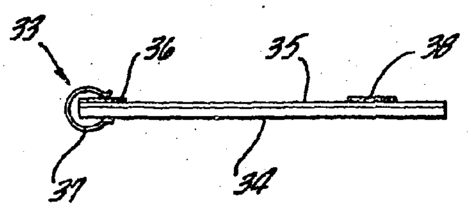


FIG_5

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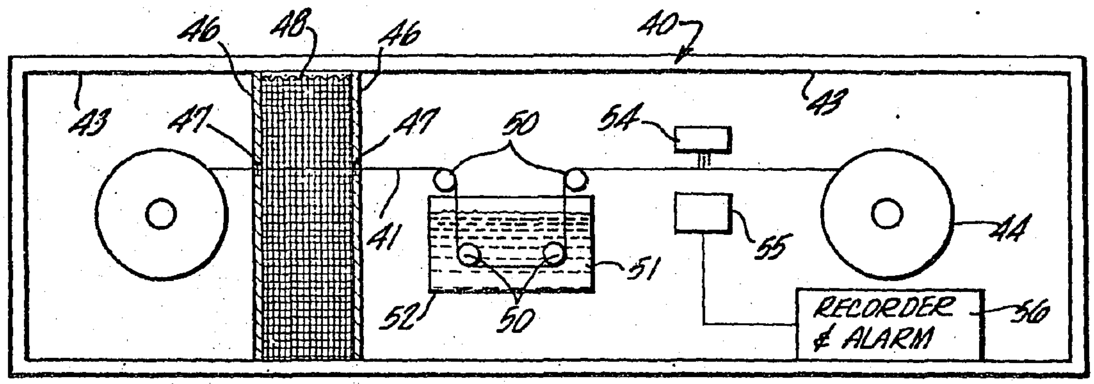

FIG. 6

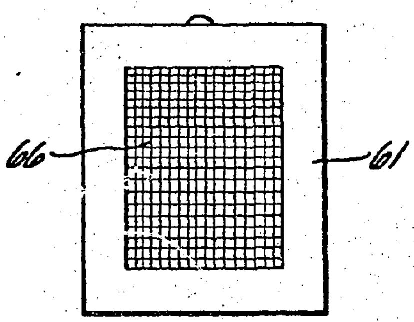

FIG. 7

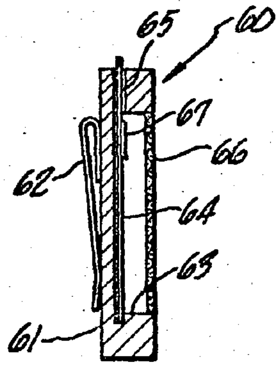

FIG. 8

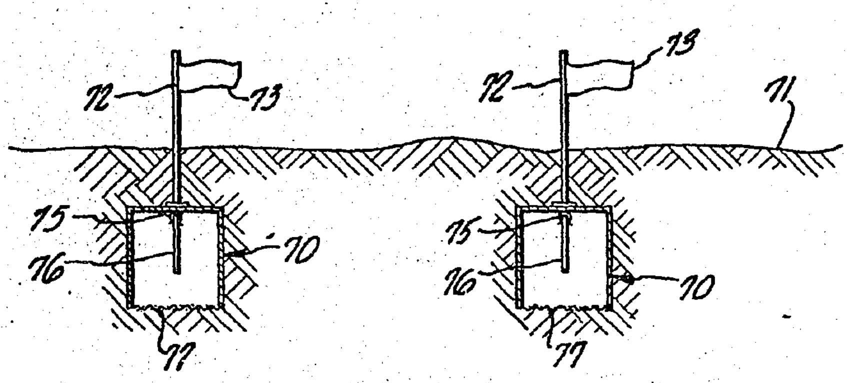

FIG. 9

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