Oct. 10, 1961 F. SULLIVAN ET AL 3,003,349

WARNING SYSTEM

Filed March 9, 1959

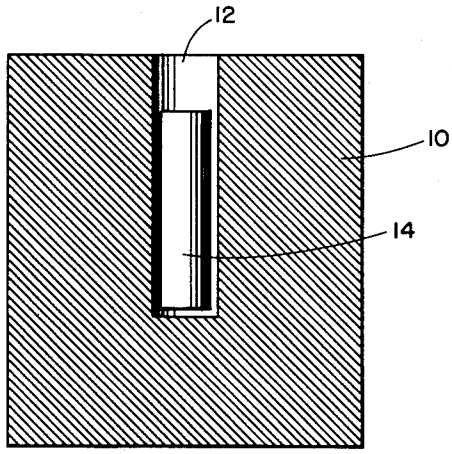


Fig. 1

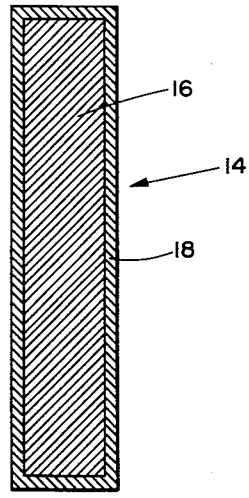


Fig. 2

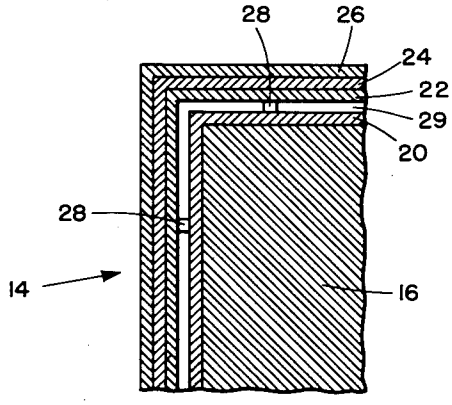


Fig. 3

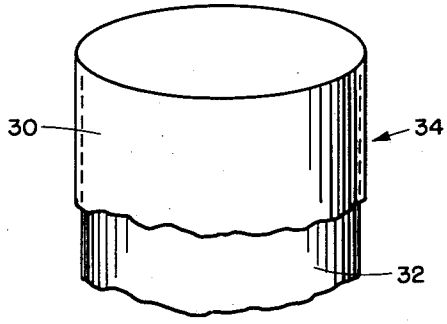


Fig. 4

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WARNING SYSTEM

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This invention relates to warning or alarm systems adapted to provide to the human senses a warning of leakage of radioactive materials, especially from closed systems, or of the hazardous presence of such materials.

Radioactive materials such as cesium 137, cobalt 60, strontium 90, and the like are customarily encased within one or a plurality of hermetically sealed covers or laminae of stainless steel, glass, noble metal, or other suitable material, in order that they may be contained safely and without danger to personnel or contamination of surrounding materials by direct contact. The radioactive material, which may be solid, liquid or gaseous, may be formed and covered or protected in one or another of several ways, although in present practice it is frequently a solid in the shape of a long, narrow rod and the covering or cladding is in the form of a thin-walled cylindrical container, or of a plurality of such containers which nest together closely, one within the other. Each of these is hermetically closed against the escape of the radioactive material. The thus clad or encased radioactive material (hereinafter usually referred to as an "element") when in use is ordinarily placed within shielding means which may be in the form of a suitable solid with an aperture in which the element is placed, or is submerged in water or other suitable liquid. The danger of escaping radioactive material from the elements is, however, a constant problem, especially where instrumentation is not common.

As is well known, radioactive materials have a rapidly increasing use, not only for military purposes, but especially for industrial purposes, such as for fuel elements, in thickness and other gauges, and as radiation processing sources for sterilizing, etc.

Furthermore, there is the problem of safe disposal of radioactive wastes. Such increasing uses and applications of these radioactive materials emphasize the need for the safe handling and for the rapid detection of radiation hazards resulting from leaks through the cladding or containers. Such leaks are presently detected by wipe tests and by monitoring counters which are installed in most laboratories which now handle these materials. However, the absence of such instrumentation, failure of detection equipment, or the handling of the radioactive elements by untrained personnel emphasizes the need for a simple, rapid warning system capable of triggering a sensory response which will warn personnel of possible danger from or leakage of radioactive material from an element containing it. Although the cladding is ordinarily designed and arranged to be hermetically sealed so that no radioactive material will escape, there is always danger due, for example, to the highly active and/or corrosive nature of the radioactive material, or to any error, hazards or damage in handling, that leaks will develop through cladding whereby portions of the radioactive material will escape. This is especially true in the case of radioactive materials which are not in the form of a massive solid, but are rather in gaseous, liquid, or powder form.

It is a principal object of this invention to provide a detecting means associated with the radioactive elements whereby any leakage becomes immediately manifested. It is a further object of this invention to provide such detecting means in the form of a substance which is placed either between the respective layers of cladding or within

the innermost such layer, either surrounding or mixed with the radioactive material itself. A further object of this invention is to provide such detecting means which will not be damaged by radiation from the radioactive material and which can be detected by one or more suitable means such as by odor, by appearance, or by the automatic actuating of detecting means. Other objects will appear as the description proceeds.

This invention will now be described in more detail with reference to the accompanying drawings wherein—

FIG. 1 represents a fuel element or other clad radioactive material positioned within a solid shield;

FIG. 2 represents a longitudinal cross-section of a typical element of this invention;

FIG. 3 represents an enlarged cross-section of a portion of an element of this invention; and

FIG. 4 represents another modification of this invention.

The arrangement shown in FIG. 1 of the drawings is that of a shield **10** having a cylindrical aperture or orifice **12** in which is placed a fuel element **14** or other radioactive source. Details of a typical element **14** are shown in FIG. 2 wherein the radioactive material is represented by the numeral **16** and the cladding or claddings by the numeral **18**. The cladding **18** may be a single layer.

FIG. 3 shows an enlarged detail of a portion of an element of the general type of FIG. 2. In this instance, several cladding layers are shown, identified by the numerals **20, 22, 24,** and **26**. A warning material **29**, in accordance with the present invention, is placed between two contiguous layers of the cladding, in this instance between layers **20** and **22**. Alternatively or in addition, the warning material of this invention may be mixed with the radioactive material **16** or may surround the same between it and the inner cladding **20**. It is also possible to use different types of warning materials in the same element, e.g. by placing one of them between any two contiguous layers of cladding and another between any other two contiguous layers.

When the warning material between claddings, as at **29** in FIG. 3 is gaseous, the claddings between which it is placed may be spaced apart by suitable means such as lugs **28**, and the gas may also be under pressure in order to promote its rapid diffusion in the event of a leak.

FIG. 4 shows another modification wherein a warning material **30**, typically a fluorescent warning material, is placed as a coating over the surface **32** of a container **34** within which radioactive material is sealed.

The warning material, if intended to be detected because of its odor, should, of course, be non-hazardous to personnel and environment and should be unique (i.e. not like other odors in the environment concerned), persistent, pungent, not significantly damaged by radiation from the radioactive material (although it may be altered, as indicated below), and not injurious to that material. It must be capable of alerting people to immediate action to correct the hazard. Selection should take into account the consideration that radiation-stable materials usually have low molecular weights or a ring-type aromatic structure. Radiation stability can be ascertained experimentally by exposing the warning materials to intense doses of radiation of the type to be warned against. Suitable warning materials for the purposes of this invention also include those which are not in themselves notably detectable or distinguishable by the senses, but which are converted promptly thereto upon exposure to the type of radiation to be detected hereby. Suitable warning materials also include those which are not radiation-stable, but which may be reformed into suitable form upon leakage out of, and away from, the radioactive element. For example, chlorine gas is not very radiation-stable, being disso-

ciated into chlorine atoms or ions by radiation. Yet, upon removal from a radiation field, chlorine would again be formed; or hydrogen chloride or other chlorine compound from reaction with the surrounding air or moisture might be produced as the warning medium.

Odorants of the class referred to above are the thioalcohols (mercaptans) and thioethers, certain cyclic sulfur-bearing compounds, and various lachrymators and irritants, as illustrated below. The cyclic sulfur compounds are especially stable and resistant to thermal breakdown at temperatures as high as 500° C. and more. Mercaptans have exceedingly low thresholds of detection (approximately 3.3×10^{-5} p.p.m.) and are of such character that any leakage into the surrounding atmosphere is at once detected. Typical of such mercaptans are methyl, ethyl and propyl mercaptan and lower homologues thereof, lower mercapto-alcohols such as 2-mercapto ethanol, and other compounds typically containing the —SH radical. Examples of thioethers, which have the generic formula R—S—R', are diethyl sulfide, allyl sulfide, and mixed sulfides, where R and R' are different radicals. Among the above-mentioned cyclic sulfur compounds are thiophene, while other odorous sulfur compounds within the class are allyl isothiocyanate and dimethyl trithiocarbonate. Choice will depend both upon the factors recited above, and upon whether the warning material is desired in solid, liquid, or gaseous form. Also, as a rule, and particularly with the thioalcohols and thioethers, the odorous principles are much more pronounced with the impure than with the pure compounds. Hence the impure compounds are usually preferred.

Examples of lachrymators are such compounds as ethyl chloro dithioformate (which can also be classed as an odorous sulfur-bearing compound), chloracetophenone, and crotonaldehyde (which is both lachrymator and irritant). Phenyl isothiocyanate is an example of an irritant.

Many other odorous compounds may serve as warning materials, including various nitrogen-containing and/or unsaturated hydrocarbon compounds such as pyridine, dinitrobenzene, indene, indole and vinyl cyclohexane.

In instances where the warning material has inadequate stability in the presence of the radiation to be warned against, a stabilizer may be added. Such stabilizers are for example aromatic compounds such as benzene, diphenyl, and terphenyl.

Another type of warning material which is particularly useful in instances where the radioactive material is in submerged position is one which provides a visual alarm such as a fluorescent compound or a visible dye either of which would be released into the liquid medium surrounding the radioactive material, e.g. as in a swimming pool type of system. The fluorescent compound may be one which would be detectable in the presence of a suitable light source arranged to shine continuously upon the liquid medium. Alternatively, the fluorescent compound may be one which fluoresces when activated by the radiation from the source itself.

The visual warning material may also be used in instances where the radioactive material is not submerged, e.g. in the form of a colored liquid material such as a non-volatile dye dissolved in a suitable solvent. Development of a leak in the container would allow the colored warning material to seep out and remain visible on the element's surface as positive evidence of leakage. A slight pressure to force the warning agent out of the leak may be maintained in the element by incorporating a gas such as air with the agent, or selecting an agent which exhibits a slight decomposition to a gas upon irradiation.

A coating **30** (FIG. 4) may advantageously serve to warn of the dangers of radiation from the radioactive material enclosed within an element, even though there may be no leakage. Thus, such an element unless properly shielded (e.g. by shield **10**, FIG. 1) can cause severe if not fatal radiation damage to any individual who is near it or who handles it without proper precautions. However, such an element does not ordinarily look any different from any other non-radiating object of the same size and external structure. But, by providing a suitable coating **30** which glows in the presence of radiation from the contained radioactive material, any individual who sees the resulting glowing object is effectively warned that the object is not a conventional and safe one. Suitable fluorescent materials for this purpose are the television tube and fluorescent light phosphors, such as calcium lead silicate and magnesium tungstate. Organic fluorescent materials such as the sodium salt of fluorescein may also be used.

The fluorescent coating **30** may advantageously be made up of the above-described fluorescent warning material in a vehicle which does not affect the action of either the warning material or the radioactivity, and which is unaffected thereby, and which is adherent to the surface **32** of the object coated. Many ordinary lacquers, e.g. of the phenolic type, are suitable for this purpose. The warning material is dissolved or dispersed in the vehicle and the resulting composition applied in the manner appropriate for the vehicle used.

Alternatively, in connection with the arrangement shown in FIG. 4, the coating **30** may be a fluorescent material which serves to detect the presence of dangerous radiation from sources adjacent to, but outside of, container **34**.

Obviously, the warning materials of this invention may be used in conjunction with the Geiger counters and other like detecting means presently used for instrumental warning purposes. It is also clear that the warning material of this invention must not interfere with the action or effectiveness of the radioactive material. The radiation from the latter may in some uses be relatively weak, and obviously the warning material employed should be chosen accordingly, with respect to characteristics, quantity and location.

It should also be noted that the placement of the warning material of this invention outside of the innermost cladding, as at **29** of FIG. 2, will serve to warn of potential leakage of radioactive material before active leakage occurs, in instances where the outer claddings have been ruptured or broken, but the innermost cladding is still intact.

Other warning materials detectable by the human senses, and other means for detecting radioactivity thereby, will occur to those skilled in this art, within the scope of the appended claims.

What is claimed is:

1. A sealed radioactive element consisting of a radioactive material and a casing completely surrounding said material, and a warning medium associated with said element and positioned within said casing, said medium being adapted to move outwardly from within said casing through any break therein and thereupon to provide a warning, directly detachable by the human senses, of the presence or potential leakage of radioactivity, said medium being confined within said casing in the absence of such break.

2. A sealed radioactive element according to claim 1 wherein said medium is under superatmospheric pressure.

3. A sealed radioactive element according to claim 1 wherein said medium contains a substance adapted to stabilize it against deleterious effects of said radioactivity.

4. A sealed radioactive element according to claim 1 wherein said medium is a fluorescent compound.

5. A sealed radioactive element according to claim 1 wherein said medium is a lachrymator.

6. A sealed radioactive element according to claim 1 wherein said medium is a dye.

7. A sealed radioactive element according to claim 1

wherein said medium is an odorant which is non-toxic, persistent or pungent.

8. A sealed radioactive element according to claim 7 wherein said odorant is a mercaptan.

9. A sealed radioactive element according to claim 7 wherein said odorant is a cyclic sulfur compound.

10. A sealed radioactive element according to claim 7 wherein said odorant is an irritant.

11. A sealed radioactive element consisting of radioactive material encased within a plurality of successive superposed layers of cladding, and a warning medium positioned in said element within at least the outer of said layers, said medium being adapted to diffuse outwardly from said element through any break in said outer layer, but being otherwise confined within said

layer, and to be readily detectable by the human senses upon so diffusing.

12. A sealed radioactive element according to claim 11 wherein two of said layers are spaced apart to define a space wherein said warning material is contained.

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CERTIFICATE OF CORRECTION

Patent No. 3,003,349 October 10, 1961

Frederick Sullivan et al.

It is hereby certified that error appears in the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 4, line 58, for "detachable" read -- detectable --.

Signed and sealed this 27th day of March 1962.

(SEAL)
Attest:

ERNEST W. SWIDER
Attesting Officer

DAVID L. LADD
Commissioner of Patents