

Jan. 18, 1966

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3,230,374

RADIOACTIVE RADIATION EMITTING SOURCES

Filed Dec. 18, 1962

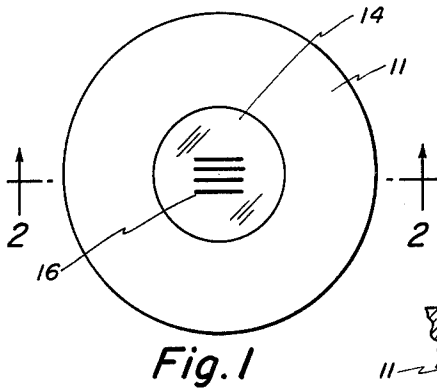


Fig. 1

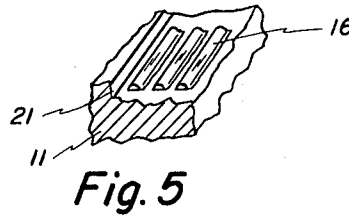


Fig. 5

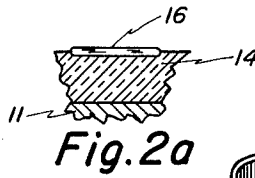


Fig. 2a

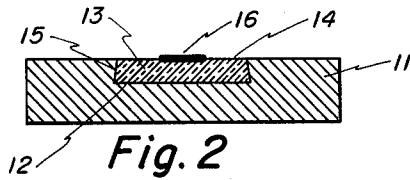


Fig. 2

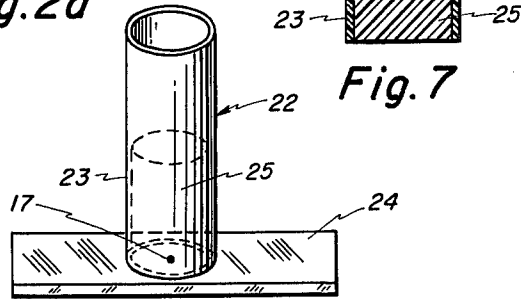


Fig. 6

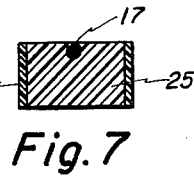


Fig. 7

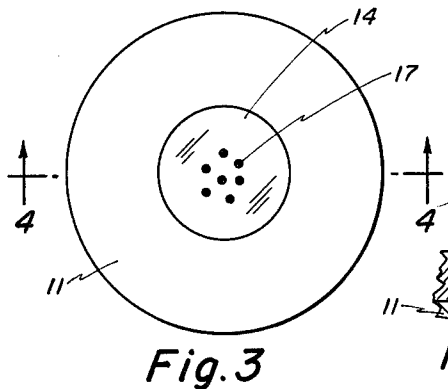


Fig. 3

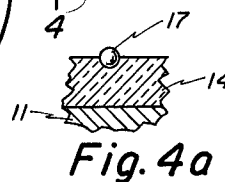


Fig. 4a

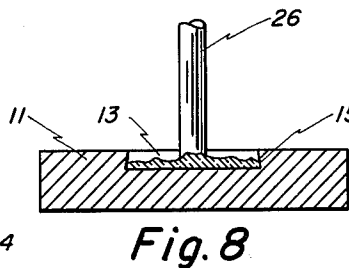


Fig. 8

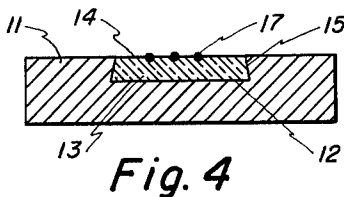


Fig. 4

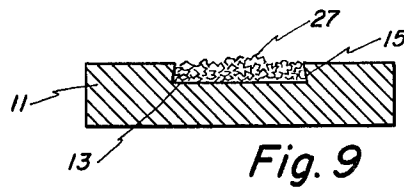


Fig. 9

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RADIOACTIVE RADIATION EMITTING SOURCES

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Filed Dec. 18, 1962, Ser. No. 245,954

2 Claims. (Cl. 250—106)

The present invention relates generally to sources for emitting radiation and more particularly to contamination-free alpha, beta, gamma and neutron sources that are capable of being fabricated into various configurations for providing preselected quantities of radioactive emissions per unit of time.

Radiation sources capable of providing alpha, beta, gamma, and neutron emissions have been known in the art for some time. However, each of these prior art sources for various reasons suffers certain drawbacks or shortcomings which may render it incapable of or unsuitable for performing certain tasks. For example, alpha sources prepared by depositing a radioactive alpha emitter on a metal substrate such as copper, over a period of time tend to oxidize and cause some of the radioactive material to "flake" and lose its adherence to the substrate. Thus, such "flaking" allows the radioactive material to be readily "wiped" from an aged alpha source and, as a consequence, creates a health hazard while effecting deterioration of the source which cannot thereafter be suitably used as a standard. Other shortcomings of the known sources lies in the inability of readily fabricating them to provide a predetermined number of radioactive disintegrations per minute and in preparing sources with unusually large numbers of radioactive disintegrations per minute; for example, an alpha source capable of 10^7 disintegrations per minute from a relatively long half-lived isotope such as Pu_{239} was heretofore very difficult to fabricate by known methods.

The present invention aims to provide new and improved sources and methods of making the same which overcome or substantially minimize the above mentioned or other difficulties or disadvantages of sources heretofore known in the art.

An object of the present invention is to provide new and improved sources and methods of making the same which overcome or substantially minimize the above mentioned or other difficulties or disadvantages of sources heretofore known in the art.

An object of the present invention is to provide new and improved methods of fabricating radiation emitting sources which are substantially safer and less costly than the previously known methods.

Another object of the present invention is to provide new and improved fabrication methods which may be used to individually prepare alpha, beta, gamma and neutron sources.

Another object of the present invention is to provide radiation emitting sources capable of being fabricated to emit a desired number of radioactive disintegrations or counts per minute.

A further object of the present invention is to provide contamination-free radiation emitting sources capable of functioning as stable standards.

A still further object of the present invention is to provide radiation emitting sources which are chemically inert and can withstand temperatures up to about 900°C .

Other and further objects of the invention will be obvious upon an understanding of the illustrative embodiments about to be described, or will be indicated in the appended claims, and various advantages not referred to herein will occur to one skilled in the art upon employment of the invention in practice.

Preferred embodiments of the invention have been chosen for purposes of illustration and description. The preferred embodiments illustrated are not intended to be exhaustive nor to limit the invention to the precise forms disclosed. They are chosen and described in order to best explain the principles of the invention and their application in practical use to thereby enable others skilled in the art to best utilize the invention in various embodiments and modifications as are best adapted to the particular use contemplated.

In the accompanying drawing:

FIG. 1 is a plan view showing a source embodying a plurality of radiation emitting fibers bonded to a glass-filled metal receptacle;

FIG. 2 is an elevational sectional view taken along line 2—2 of FIG. 1;

FIG. 2a is an enlarged fragmentary view of the section illustrated in FIG. 2;

FIG. 3 is a plan view showing a source embodying a plurality of radiation emitting microspheres bonded to a glass-filled metal receptacle;

FIG. 4 is an elevational sectional view taken along line 4—4 of FIG. 3;

FIG. 4a is an enlarged fragmentary view of the section illustrated in FIG. 4;

FIG. 5 is a fragmentary perspective view showing the addition of individual radiation emitting fibers to a metal substrate;

FIG. 6 is a perspective view of a form of the present invention showing a radiation emitting sphere immersed in liquid resin;

FIG. 7 is an enlarged sectional view showing the source produced by the method of FIG. 6;

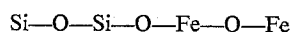
FIG. 8 is a sectional view showing a radiation emitting substance being melted into or bonded to a metal substrate; and

FIG. 9 is a sectional view showing fragments of a radiation emitting substance prior to their direct bonding to a metal substrate.

While radioactive substances which emit gamma rays are considered true radiation emitters, the expression "radiation emitter" is used herein with the intent of encompassing various radioactive substances which in addition to gamma radiation are individually capable of emitting alpha particles, beta particles or neutrons.

As shown in the embodiment of FIGS. 1—4a the present invention comprises a generally circular metal disk or container 11 which may have at one side thereof a recessed surface 12 defining a cavity or well 13. A single piece of silicate glass 14 or fragments thereof may be placed in the well 13 and then by a suitable heating means, such as, for example, a furnace or an induction coil (not shown), the container may be heated to a temperature of about 900°C to 1000°C so as to melt the glass 14 and substantially, if not completely, fill the well 13 with molten glass. The heating means may then be inactivated and the glass filled container cooled to about room temperature. While silicate glass is preferable because of its chemical and thermal stability with the isotope oxide or compound and its resistance to chemical attack, other glass, such as, for example, phosphate glass or borate glass, may be used.

The support member or container 11 may be preferably made of stainless steel with a slightly oxidized surface, since the molten silicate glass 14 readily adheres or fuses to the oxidized surface by forming



bonds therebetween. In addition to the excellent bonding properties provided between the glass and the steel container the latter also provides a favorable coefficient of thermal expansion for use with silicate glass. However, containers made of other material, such as, for example,

nickel, platinum or tantalum, may be suitably used. Side walls of the well 13 are preferably slightly undercut 15 to furnish a mechanical bond for supplementing the chemical bond between glass and the container in retaining the glass within the well 13. The angle of the side wall undercut 15 is not normally significant as long as it provides a slight inward slope towards the upper surface of the container 11.

A radiation emitting substance which may be in the form of one or more beads, microspheres, fibers or sheets, as will be set forth in detail below, may be placed upon the exposed surface of the glass 14 and the container 11 and its contents thereafter reheated to suitable temperature, e.g., about 900° to 1000° C. for fibers, to return the glass to a molten state. In some instances (particularly where the radiation emitting substance is disposed in parent glass and spun into a plutonium bearing glass fiber) as the glass 14 nears the molten state, the radiation emitting fiber 16, which may be of any appropriate length and of any desired diameter (FIGS. 1-2a) softens and fuses into the surface of the glass 14 (FIG. 2a), whereupon subsequent cooling of the container and its contents effects a secure wipe-free bond between the glass 14 and the fiber 16 for providing a source integral in structure. The fiber 16 may be as small as one micron in diameter, but normally fibers between fifteen and thirty microns are used. Also, the fiber 16 may be fused into the glass to a depth where a minor portion of the fiber 16 remains above the surface of the glass (FIG. 2a) or, if desired, the fiber 16 may be fused into the glass 16 so as to have its upper surface substantially level with the surface of the glass 14. However, in another instance, as will be brought out below, the glass 14 may be heated to about 1200° C. for causing a differently prepared radiation emitting substance in the form of a microsphere 17 (FIGS. 3-4a) to partially or entirely "sink" into the molten glass 14 (FIG. 4a). As the glass 14 cools to about room temperature it tightly "grips" the contiguous surfaces of each radiation emitting microsphere and rigidly secures it. If the radiation emitting substance provides alpha or beta emissions, care should be exercised when attaching it to the glass to assure that it does not submerge too far into the glass 14, since the latter acts as a shield which serves to attenuate and hence control alpha or beta particle emission.

The radiation emitting substances may be produced by utilizing various novel processes. For example, an alpha emitting substance may comprise plutonium-bearing glass in any suitable configuration which may be prepared by dissolving plutonium oxide in modified silicate glasses such as by utilizing the procedure set forth below.

A quantity of silicate or other suitable glass may be ground or crushed in any suitable manner to a size less than about 100 mesh, i.e., less than 0.01 of an inch in diameter. A weighed amount of plutonium oxide powder may be placed into a crucible comprised of platinum alloy or the like and then a weighed amount of the ground glass may be placed over or mixed with the plutonium oxide. The glass and the plutonium oxide may be thereafter heated by an induction coil or any other suitable heating means to about 1700° C. to melt the glass and substantially, if not completely, dissolve the plutonium oxide. In order to completely homogenize the mixture and thereby uniformly distribute the plutonium oxide alpha emitter throughout the solution to form the plutonium-bearing glass, the heating means may be turned off after the glass is initially liquified to allow the mixture to solidify. The crucible may then be inverted and again heated until the glass runs down the side of the crucible. The heating means may be turned off again to allow the mixture to solidify and then the crucible is inverted and the process repeated.

As briefly mentioned above the radiation emitting substance may be in the form of thin rods or fibers of plutonium-bearing glass or, if desired, in the form of small

beads or microspheres of the plutonium-bearing glass. These various forms may be produced in any suitable manner.

Microspheres of the plutonium-bearing glass normally having diameters smaller than the above mentioned beads, may, for example, be prepared by crushing the plutonium-bearing glass to a particle size less than about 200 mesh which are thereafter passed through an induction coupled plasma torch with an argon feed which provides a temperature of about 14,000°-19,000° K. and causes the particles to fluidize and take on a generally spherical shape. After the spheres pass through the torch they may be collected in a water-filled beaker placed below the torch.

Radiation emitting substances in the form of sheets may be conveniently formed by melting plutonium-bearing glass on a metal surface and then smoothing it out.

While plutonium oxide is generally preferred as the source of alpha radiation it may be desired to use other alpha emitting metals or oxides, for example, americium, neptunium, uranium, curium and thorium.

Beta emitting sources and gamma emitting sources may be provided by substituting the oxide or other form of a suitable beta or gamma emitting substance for the alpha emitting substance in the fabrication method above described. A beta emitter which may be homogeneously mixed with glass may comprise, for example, strontium-90 while a gamma emitter which may be similarly used may comprise cobalt-60.

Neutron emitting sources may be prepared in a contamination-free manner by using a fluoride glass composition containing dissolved plutonium tetrafluoride. For example, a neutron source weighing about three grams and emitting about 2400 neutrons per second may be prepared by adding about two and one fourth grams of plutonium tetrafluoride powder to about five and one third grams of a fluoride glass of the following composition:

	Wt. percent
MgF ₂ -----	12.63
CaF ₂ -----	13.69
SrF ₂ -----	8.42
BaF ₂ -----	11.58
CeF ₄ -----	4.21
ThF ₄ -----	5.26
AlF ₃ -----	21.05
BeF ₂ -----	23.16

The above plutonium fluoride and fluoride glass mixture may be placed in a suitable crucible and heated to about 900° C. in the presence of ammonium bifluoride which functions to form a thick fog for blanketing the mixture to prevent reaction with atmospheric oxygen and moisture. The ammonium bifluoride subsequently completely decomposes. Upon decomposition of the ammonium bifluoride, the homogeneous glass mixture may be formed and cooled in any convenient shape such as beads, fibers, etc. by utilizing methods similar to those above described.

If desired, neutron emitting sources may also be prepared by dissolving plutonium oxide and beryllium oxide in a parent glass containing essentially silicon dioxide, sodium oxide, potassium oxide, calcium oxide and aluminum oxide. Such a mixture which may have about seven and one-half weight percent plutonium oxide and about seven weight percent beryllium oxide and weighs about six grams is capable of emitting about 1760 neutrons per second. Other parent glasses, e.g., borate glass or phosphate glass, may be utilized, if desired, in the preparation of a neutron source.

Instead of making the alpha radiation emitter contamination-free by dissolving the plutonium oxide in a parent glass it has been found that by passing plutonium oxide in the form of crushed particles ranging in size from about 400 mesh to 35 mesh through a plasma torch employing an oxygen enriched argon plasma, similar to the one de-

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scribed above, small microspheres of plutonium oxide, such as indicated by numeral 17 in FIGS. 3-4a, are formed. These microspheres are contamination-free and provide a greater number of alpha disintegrations per minute than similarly sized microspheres of the plutonium-bearing glass.

Again referring to FIGS. 1-4a, the radiation emitting sources shown may be "built up" to provide a desired number of radioactive disintegrations or counts per minute by preselecting the size and quantity of the radiation emitting substance to be placed upon the glass substrate 14 within the container well 13. For example, when a single microsphere of plutonium oxide 17, which normally emits about 153,000 counts per minute, is submerged about halfway into the glass 14 it emits about 80,000 counts per minute while on the other hand, four such microspheres submerged about halfway into the glass emit about 308,000 counts per minute. Thus, by placing a desired number of plutonium oxide microspheres, which number may be one or more, on the glass surface of a glass-filled container prior to the reheating step, a radiation emitting source capable of emitting a preselected number of counts per minute may be readily fabricated. However, as briefly mentioned above, the penetration of alpha and beta radiation emitting substances into the glass substrate should be carefully controlled since the deeper the penetration the less the count due to the shielding effect of the glass; for example, the plutonium oxide microsphere capable of initially emitting about 153,000 counts per minute will only emit about 20,000 counts per minute when it is almost but not quite completely submerged in the glass 14.

In the form of the invention shown in FIG. 5, a desired quantity of radiation emitting substances, which preferably have a glass base, are placed in an empty well 13 of a metal container 11 which is thereafter heated to about 900° C. for a short time period sufficient to cause a slight melting of the substances and effectively bond the latter to the metal of the container. The radiation emitting substances may be in any desired shape or size, such as, for example, elongated glass fibers 16 as shown or "shorter" beads or microspheres. These may be arranged within the well 13 in a contiguous or spaced apart side-by-side relationship or in any other suitable arrangement. Also there may be only a single fiber, bead, or microsphere adjacent the center of the well or the entire well surface may be substantially covered with the fibers depending, of course, upon the source strength desired.

By laying the elements, especially the alpha and beta emitters, in the well 13 in such a manner that the well side wall 21 provides an upright enclosure about the elements sets forth a desirable feature in that the upright side wall 21 functions as a radiation shield and minimizes "edge" effects or spurious radiation readings as to source strength.

FIGS. 6 and 7 show a modified form of the invention which may provide a point source useful as a calibrated checking means for various instruments. This form of the invention utilizes a plastic mount for maintaining the radioactive substances in their proper position. The plastic mount 22 may comprise a tube 23 made of a suitable plastic, such as Plexiglas or the like, with an end thereof ground or cut substantially flat. This flat end of the tube 23 may be placed in an upright position upon a clean flat surface of a material such as a glass microscope slide 24 or the like, which is preferably not capable of accepting a bond with plastic. A small volume of any suitable commercially available liquid plastic resin 25, e.g., that known by the trade name "Kold Mount," made by Vernon-Benshoff Co. of Pittsburgh, Pennsylvania which normally cures and hardens within a short time after formulation may be poured into the open or the upper end of the tube 23. A single radiation emitting substance, which may be in the form of a microsphere 17, may then be dropped into the resin at the center of the

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tube 23. Because the microsphere 17 normally has a density greater than the resin it drops or passes through the resin 25 until it eventually comes to rest against the flat surface of the glass slide 24. A convenient method which may be used in properly placing the microsphere 17 into the liquid resin 25 is to pick up the microsphere 17 on the end of a pinpoint which has been previously dipped into the liquid resin 25 and thereafter placing the microsphere 17 into the liquid resin within the tube 23, which causes the microsphere 17 to dislodge itself from the pinpoint and drop through the resin 25.

After the plastic 25 hardens (approximately 30 minutes when using the "Kold Mount" plastic) the glass slide may be pulled free from the tube 23 leaving a smooth surface at the interface with the microsphere 17 permanently imbedded in the plastic 25 in a position tangent thereto yet substantially completely surrounded by the plastic. The tube 23 may then be cut off to any desired length so as to provide a source of a convenient and compact size (FIG. 7).

Employing a single plutonium oxide microsphere of about 0.15 millimeter in diameter in the plastic mount can provide an alpha emission of about 15,000 counts per minute. However, while only a single microsphere is shown in the plastic mount it will appear clear that any number and size of microspheres or beads may be employed in the same mount.

In fabricating a radiation emitting source capable of yielding a large number of counts or disintegrations per minute, it may be desirable to place the radiation emitting substance directly into an empty container well 13 and thereby provide a source with a solid radiation emitter readily capable of emitting the desired count. In order to facilitate the filling of the well 13 in container 11 with a suitable radiation emitting substance, the container 11 may be first heated to about 1100° C. in an induction coil or the like and then the radiation emitting substance added which, in turn, melts and substantially fills the well. When using this method of source fabrication it may be desired to use a radiation emitting substance homogeneously mixed in a parent glass for facilitating the formation of a strong bond between the radiation emitter and the metal container. As shown in FIG. 8 the radiation emitter is in the shape of a thin rod 26 which may be continuously melted and fed into the well 13 of the heated container 11 until the desired source strength is attained or until the well is full. FIG. 9 illustrates a similar source except that instead of using a rod, a plurality of radiation emitting fragments 27 may be placed in the container well 13 prior to heating the container 11 to about 1000° C., which causes the fragments 27 to melt down and form a solid mass securely bonded to the metal container 11. The well side wall undercut 15 may also be utilized to provide a mechanical bonding advantage in the embodiments shown in FIGS. 8 and 9 and thereby supplement the chemical bond between the emitter and the containers.

In fabricating sources like those shown in FIGS. 8 and 9, the radiation emission is normally very uniform over the whole surface of the emitter. Therefore, it may be desirable to use a relatively shallow container well 13 so as to decrease the thickness of the radiation emitting substance while increasing the area of the exposed surface for a given volume and thereby obtaining maximum source efficiency by exposing the greatest amount of surface with the smallest volume.

The container 11, the well 13 and the tube 22 are shown generally circular; however, it will appear clear that they each may be of any suitable shape such as square, oval, etc.

It will be seen that the present invention sets forth new concepts in radiation source fabrication which are contamination-free, very resistant to deterioration by flaking, oxidizing and the like, and which do not require the use of protective windows over the sources. The con-

tamination-free radiation emitting substances, whether they are alpha, beta, gamma or neutron emitters, may be fabricated into an infinite variety of shapes and strengths which may be best suited for the particular use anticipated. The sources fabricated are wipe-free, resistant to attack by water and weak acids, resistant to relatively high temperatures and very corrosion resistant. The retention of the alpha emitter, e.g., plutonium, in the parent glass reduces the severity of the container requirements in that accidental destruction of the source by breakage or by fire would not result in a spread of loose contamination.

As various changes may be made in the form, construction and arrangement of the parts herein without departing from the spirit and scope of the invention and without sacrificing any of its advantages, it is to be understood that all matter herein is to be interpreted as illustrative and not in a limiting sense.

We claim:

1. A source providing a predetermined number of radioactive disintegrations per minute comprising a metal container having a surface wall and a recess with an inner wall inwardly spaced from said surface wall and connected therewith by a side wall extending generally laterally from said inner wall, a plurality of discrete glass fibers cumulatively providing said predetermined radioactive disintegrations arranged in parallel contiguity with each other within said recess and each bonded directly to and projecting from said inner wall, said side wall laterally projecting from said inner wall a distance greater

than said fibers for shielding radiations emanating from the fibers in directions generally parallel to said inner wall.

2. A source providing a predetermined number of radioactive disintegrations per minute comprising a metal container having a surface wall and a recess with an inner wall inwardly spaced from said surface wall and connected therewith by a side wall extending generally laterally from said inner wall, a plurality of discrete glass fibers cumulatively providing said predetermined radioactive disintegrations arranged in parallel contiguity with each other within said recess and each secured to said inner wall and having major portions of the fibers disposed inwardly of said surface wall.

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