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CALIFORNIUM-252 NEUTRON SOURCE AND METHOD OF MAKING SAME

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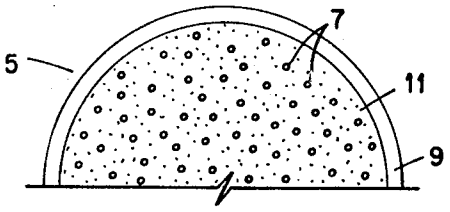


Fig. 1



Fig. 2

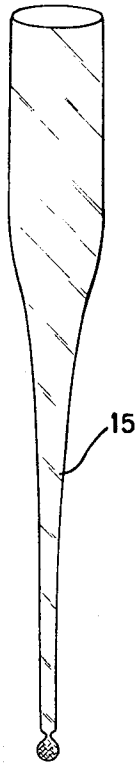


Fig. 3

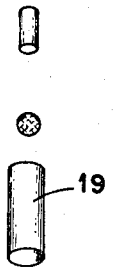


Fig. 4

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CALIFORNIUM-252 NEUTRON SOURCE AND METHOD OF MAKING SAME

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ABSTRACT OF THE DISCLOSURE

A radiation source has been provided which is free of radioactive surface contamination comprising a solid quartz bead containing a spontaneously fissionable element distributed substantially evenly within the confines of an outer glass shell. A method has been provided for preparing the source comprising sorbing a spontaneously fissionable element on quartz powder in a quartz capillary and subsequently fusing the powder and capillary to contain the neutron-emitting material.

BACKGROUND OF THE INVENTION

The present invention was made during the course of, or under, a contract with the U.S. Atomic Energy Commission.

The present invention relates to radiation sources and more specifically to a glass encapsulated radiation source and method for making the same.

In the art of preparing radiation sources, it is well known to suspend certain radioactive particles in vitreous fused materials. Generally, the methods for production of such sources take the form of mixing the source isotopes into a glass forming matrix, subsequently fusing the matrix material at elevated glass making temperatures to bond the isotope in and about the formed glass body. For further information concerning these prior methods reference is made to U.S. Pat. 3,114,716 to Quinby, issued Dec. 17, 1963 and U.S. Pat. 3,147,225 to Ryan, issued Sept. 1, 1964.

Because of its high probability for spontaneous fission, californium-252 is a unique neutron source ($(2.3\times10^{12}$ n./sec./gram). It has outstanding potential in providing compact, portable and reproducible neutron sources with intensities ranging from a few neutrons per second up to greater than 10^{12} neutrons per second. Of utmost necessity in any source preparation involving ^{252}Cf is complete encapsulation, resulting in an external surface completely free of radioactive contamination. The above described prior art methods of glass entrapped sources all suffer from the disadvantage of a certain amount of surface contamination. It has been the practice in the art to wash the surface of glass formed sources with an acid wash to remove radiocative particles which cling to the outer surface. Since californium-252 is so highly radioactive, it is imperative that there be no surface contamination at all in order to provide a safe source which does not require a secondary or double encapsulation to eliminate external contamination.

Further, in the art of fabricating ^{252}Cf sources it has been the practice to electroplate the ^{252}Cf onto a platinum needle, to form a mixed oxide pellet of ^{252}Cf, and to incorporate ^{252}Cf into a rare earth sol which is subsequently cast into gel microspheres and fired to a mixed oxide.

These techniques suffer from lengthy preparation times, dilution of the ^{252}Cf, multiple mechanical manipulation, and the necessity for ultimate double encapsulation of the source to eliminate external contamination.

Further, it has been found that it is very difficult to control the size of the source prepared by the prior art methods as opposed to the present method.

SUMMARY OF THE INVENTION

It is, therefore, an object of the present invention to provide a radiation source embodying a bead comprising vitreous material with a spontaneously fissionable element distributed therein.

It is another object of this invention to provide a neutron source as set forth above having a non-contaminating surface formed by an outer shell of vitreous material.

Yet another object of this invention is to provide a method for the preparation of a radiation source as set forth in the above objects.

Still another object of this invention is to provide a method for the preparation of a radiation source wherein an open-ended impermeable vitreous tube is packed with a porous powder of a refractory material consisting primarily of silicon dioxide having a water-immiscible organic liquid ion exchange material sorbed thereon, an aqueous solution containing cations of a spontaneously fissionable material passed through the thus-treated powder pack and the powder pack is subsequently dried and heated in air to oxidize and volatilize the organic material to provide a non-volatile oxide of the fissionable material; and then fusing the tip and the remaining powder filled portion of the tube to form a vitreous body sealably containing the radioactive oxide.

Other objects and many of the attendant advantages of the present invention will be apparent from the following detailed description of the invention.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a cross sectional view of a radioative source article of the invention;

FIG. 2 is a pictorial view of a capillary tube containing refractory quartz powder and the fissionable source material prior to heating;

FIG. 3 shows the tube of FIG. 2 subsequent to heating and forming of a sphere encapsulating the source material; and

FIG. 4 shows the spherical source taken from the capillary end and a closable secondary container for the source.

TECHNICAL DISCLOSURE

Referring to FIG. 1, an enlarged cross sectional view of the present radioactive source article **5** is shown. The article is preferably a small sphere or spheroid in shape although the shape need not be restricted to a sphere-like shape. The spherical shape and size of the preferred source article is a very close approach to a true point source which is significant in many applications where small portable, maintenance free, efficient neutron sources are desired. Such a source may be very useful in cancer therapy, mineral exploration, neutron radiography, oil well logging, small-scale radioisotope production, moisture gauges, on-line analysis of flow process streams, etc.

The sphere or bead is a vitreous material with spontaneously fissionable elements **7**, such as ^{252}Cf, evenly distributed within a sealing outer layer containment **9** of vitreous material.

The fusion of the quartz capillary into a ball and melting the ball up the length of the packed bed provide the outer spherical shell of quartz **9** which prevents surface exposure of the fissionable material contained within. The core **11** of the source article is composed of a matrix of vitreous material having dispersed therein, in oxide form, radioactive isotopes of the source material **7**. These radioactive isotopes are held within the core matrix by physical

forces exerted on the particles by the vitreous core material, thereby further reducing the extent of contamination in the event of accidental fracturing or breaking of the source articles.

In the process of making the source articles of this invention the following steps are employed: as shown in FIG. 2, an open-ended impermeable quartz capillary **15** is packed with a porous dry powder, fusible refractory material **17** consisting primarily of silicon dioxide, such as powdered quartz. The powder is first contacted with a water immiscible organic liquid ion exchange material such as 2-ethylhexylphenylphosphoric acid or di(2-ethylhexyl) phosphoric acid by adsorbing the liquid onto the porous powder which is then packed to a given depth depending on the final source size desired. An aqueous solution containing cations of ^{252}Cf is caused to flow downward through the packed capillary whereby more than 99.9 percent of the ^{252}Cf is sorbed onto the packing material. Dilute acid is passed through the capillary to wash non-adsorbed activity from the walls. The capillary is then dried at an elevated temperature of approximately 125° C. and then further heated to a temperature ranging from 350° C.–400° C. to decompose the organic phase and to convert the ^{252}Cf to a non-volatile oxide. By using an organic ion exchange medium the bulk of the exchange medium is vaporized or burned and the volume of the packing material is not affected by entrapped particles of the exchange material.

Once the fissionable material is sorbed on the powder pack and the organic material is decomposed by heating, a small torch is then used to first melt the lower tip, sealing the bottom end of the capillary and progressively fusing the remainder of the packed portion of the capillary and the fusible powder material into a sphere to completely encapsulate the ^{252}Cf, as shown in FIG. 3. This step is performed within a temperature range of approximately 1600°–1800° C., well below the boiling point of quartz, in order to fuse the powder pack thereby bonding the ^{252}Cf within the fused powder, and to prevent surface contamination by the diffusion of the source particles to the surface of the sphere. The sphere can then be cut off and stored in a suitable secondary container **19**, as shown in FIG. 4.

The invention is further illustrated by the following specific example.

EXAMPLE

A quartz loading tube about 8 cm. long having a tapered portion along about 5 cm. of its length and an internal diameter of about 0.5 mm. was packed with a bed of organic impregnated quartz powder to a depth of approximately 2 cm. The organic impregnated quartz powder was prepared by contacting approximately 5 grams of 200–280 mesh quartz powder with 1 milliliter of an acetone solution containing di(2-ethylhexyl)phosphoric acid followed by vacuum evaporation of the acetone to obtain an organic concentration in the powdered quartz of about 200 mg. of sorber per gram of quartz powder. About 2 milliliters of 0.05 molar HCl containing up to 65 millicuries of ^{252}Cf was passed through the packed column at a flow rate of about 1 milliliter per 10 minutes. The column was then washed by passing 1 ml. of 0.05 molar hydrochloric acid through the column. The column was then heated to a temperature of 125° C. for about 10 minutes to drive off the water and then heated to a temperature of 400° C. for about 30 minutes to decompose the organic material. The column was then fused with a propane-oxygen torch to fuse the quartz powder and tube, allowing the ball of molten quartz to roll up the length of the column. The fused quartz ball was then cut off from the remaining tube and checked for surface contamination.

Although the method completely encloses the californium, surface contamination from handling in the contaminated atmospheres of a hot cell may inadvertently contaminate the surface of the source. This may be readily removed by appropriate contact with decontamination solution.

Depending on the desired source configuration, the bead can be reshaped by appropriate glass forming techniques to form any shape desired.

The immediate advantages of this method are: (1) source fabrication times are short requiring approximately two hours; (2) the necessary mechanical manipulations are suited for hot cell application; (3) the resulting source can be in a nearly true point source configuration; and (4) no further encapsulation steps are necessary although advisable, thus minimizing source bulkiness.

It will be seen that a novel source article and method of preparing the same have been provided which has a surface area free of radiation particle contamination and is easier and quicker to prepare than by other methods provided in the art. Obviously many modifications may be made within the teachings as set forth above; therefore, it is intended that the invention be limited only in accordance with the following claims.

What is claimed is:

1. A neutron source comprising: an oxide of californium-252 essentially evenly dispersed in a vitreous quartz matrix and an outer shell of vitreous quartz integral with said inner core, said outer shell being depleted of californium-252.

2. A method of preparing a neutron source, comprising the steps of:

(a) sorbing an organic cation-exchange material contained in an acetone solution onto powdered quartz, said exchange material selected from the group consisting of 2-ethylhexylphenylphosphoric acid or di(2-ethylhexyl)phosphoric acid;

(b) evaporating the acetone to obtain an organic concentration in the powdered quartz of about 200 mg. of sorber per gram of quartz powder;

(c) packing said powdered quartz treated according to step (a) in an open ended impermeable quartz capillary to a predetermined depth so as to form a packed column;

(d) passing an aqueous solution containing HCl and a predetermined amount of californium-252 through said packed column at a rate so as to allow even sorption of substantially all of the californium-252 onto said packed column;

(e) heating said capillary and packed column in air to a temperature ranging from 350° C.–400° C. for approximately 30 minutes to remove the water, decompose the organic phase and convert the californium-252 to a non-volatile oxide; and

(f) fusing said packed column at a temperature ranging from 1600° C.–1800° C. to form a solid vitreous spheroid containing said californium-252 in a vitreous quartz matrix within the confines of an outer vitreous quartz shell depleted of californium-252.

3. The method of claim **2** wherein said organic cation-exchange material is di(2-ethylhexyl)phosphoric acid.

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