

July 18, 1972

F. D. LONADIER ET AL

3,677,958

RADIOACTIVE HEAT SOURCE

Filed Nov. 6, 1968

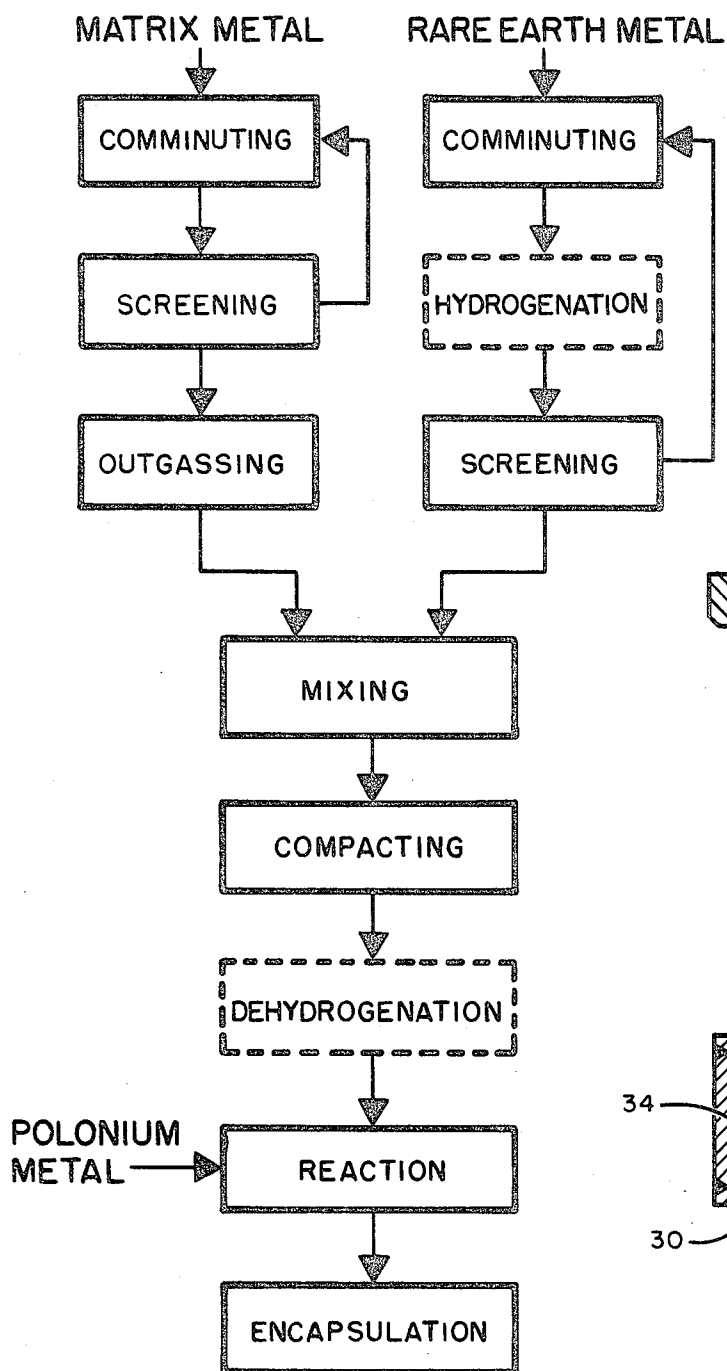


FIG. 1

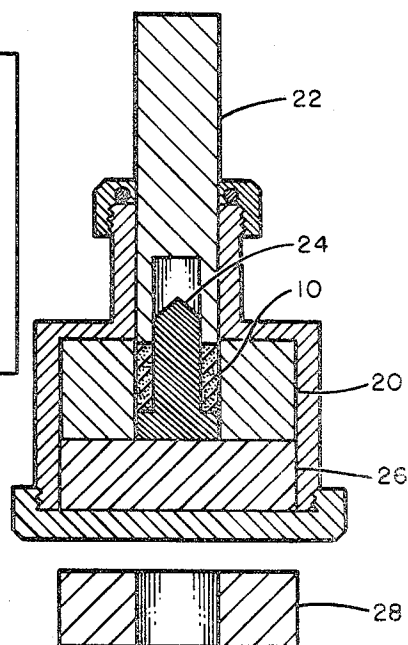


FIG. 2

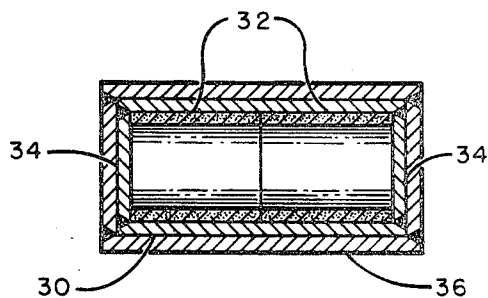


FIG. 3

INVENTORS

FRANK D. LONADIER
CARL J. KERSHNER

By: *Robert A. Goodenough*

1

2

3,677,958

RADIOACTIVE HEAT SOURCE

Frank D. Lonadier, Miamisburg, and Carl J. Kershner,
Centerville, Ohio, assignors to the United States of
America as represented by the United States Atomic
Energy Commission

Filed Nov. 6, 1968, Ser. No. 774,597

Int. Cl. C09k 3/02

U.S. Cl. 252—301.1 R

9 Claims

ABSTRACT OF THE DISCLOSURE

A radioactive heat source comprising a porous element formed by compacting intermixed ratios of matrix and rare earth type particles, and impregnating with polonium radioisotope, with pores or interstices of the element providing for escape of helium which may result from radioisotope decay.

BACKGROUND OF INVENTION

The present invention constitutes improvements upon heat sources such as referred to in U.S. Pat. No. 3,154,501. While heat sources as taught by the noted patent are deemed satisfactory, there are situations in which it is desirable to employ more or less rigid heat source elements of optimum heat transfer configuration, which may be handled up to a point during manufacture with complete absence of radioactivity, and which after reaction with radioisotope provide for ready escape or release of helium which may occur due to radioactive decay.

SUMMARY OF INVENTION

It is therefore an object of the present invention to provide new and improved radioactive heat source elements and method of making them.

Another object of the present invention is to provide a method of producing radioactive heat sources with minimum radioactive material handling and consequently minimum hazard of personnel.

Still another object of the present invention is to provide for uniform distribution of radioisotope throughout a porous heat source element for escape of evolving helium from the element via the pores.

A further object of the present invention is to provide radioactive heat source elements which facilitate efficient heat transfer.

A still further object of the present invention is to provide rigid radioactive heat source elements having large effective heat transfer areas.

Various other objects and advantages of the invention will become apparent from the description of an embodiment thereof.

As shown in the drawings the invention comprises producing radioactive heat source elements comprising comminuting and thoroughly intermixing particles of matrix metal and rare earth type materials, confining and compacting the intermixed particles into rigid and self-supporting but porous elements of density less than the theoretical solid density of solid matrix metal, and thereafter impregnating the compacted and rigid but porous element with radioactive polonium but retaining the pores of the article for escape of helium as may evolve by radioactive decay of the polonium directly into the pores and then from the element. At an appropriate time the element with polonium impregnated therein is heated to effect reaction between polonium and the rare earth type element so as to form the polonide noted in U.S. Pat. No. 3,154,501.

DESCRIPTION OF DRAWINGS

FIG. 1 is a schematic flow diagram illustrating an embodiment for production of radioactive heat source elements;

FIG. 2 is a cross-sectional view illustrating one means and manner of compacting the particles which are to comprise the elements; and

FIG. 3 is a cross-sectional view showing one arrangement of a plurality of heat source elements as they may be utilized.

DETAILED DESCRIPTION

In some instances it is desirable that there be provided a relatively rigid radioisotopic heat source element, of large exposed area as compared with thickness thereof, embodying radioactive material uniformly distributed or dispersed throughout the element. Such distribution and geometry provides heat source elements for operation at optimum temperatures by distributing radioisotopic material over a large heat transfer area and volume.

One manner of producing such elements is shown by the flow diagram of FIG. 1. As there indicated, a matrix metal or metals is ground or comminuted to desired particle size, one or more rare earth type materials is comminuted to desired particle size, and the matrix metal particles and rare earth type particles are combined in appropriate proportions and thoroughly intermixed. Intermixing is accomplished either by hand or an appropriate mixing device. All operations are preferably performed in a controlled inert atmosphere such as under argon or helium to prevent contamination of the respective materials.

The matrix metal particles are preferably de-gassed, before combining with other particles, by enclosing them in an appropriate container and subjecting them to suitable high temperatures in a vacuum for sufficient time to permit complete outgassing, for example at temperatures from around 700° C. to 1000° C. for a period of several hours (such as about 8 hours) at pressure of 10⁻⁵ to 10⁻⁴ torr.

Any suitable material may be employed for the matrix metal, provided that when intermixed and compacted or pressed it will provide porosity for helium escape, will not objectionably react with such radioisotope as may be employed, and will withstand and exhibit high strength and good stability at high temperatures present within the heat source. Examples of matrix metals which will give satisfactory results are tantalum, tungsten, chromium and noble metals such as iridium, and mixtures or alloys thereof. Tantalum is very satisfactory, probably because its particles seem to have large surface areas that contribute toward porosity or interstices of the eventual elements. The matrix metal is preferably of high purity, for example around 99.8% if tantalum alone is the metal.

Particle sizes of the matrix metal may be such as are deemed appropriate by the user, for example, such as in the range that they pass through a 60 U.S. standard mesh screen but not through a 140 U.S. standard mesh screen to provide particles between about 250 and 105 microns in size (all mesh screen sizes hereinafter refer to U.S. standard mesh); if desired and 80 mesh screen (particle size about 177 microns) may be substituted for the 60 mesh screen as the larger screen limit. In another instance there may be utilized particles which pass through a 140 mesh screen but not through a 250 mesh screen (particle size about 57 microns); and by way of still further guidance, it may be desired to employ powder which will pass through a 250 mesh screen but not through a 400 mesh screen (particle size about 37 microns). It has been found that it is preferable for

the matrix metal to be ground into a porous powder having high surface area rather than dense spheres or other forms.

The rare earth type material or materials may comprise particles formed by grinding or comminuting shavings or filings obtained for instance from machining bars or other members with a lathe, the shavings being machined from the bar or member by use of a carbon steel cutting bit. The lathe may be enclosed within a glove box or other suitable enclosure filled with argon gas and shavings formed therein. The shavings are then ground into particles to a desired mesh screen size (for example of about 250 to 400 mesh) as deemed appropriate by the user; for example, satisfactory results are obtained with gadolinium powders which pass through a 325 mesh screen (about 44 microns in diameter).

If it is desired to react the rare earth material with polonium at a faster rate and at lower temperatures, the rare earth as shavings or particles or any other convenient form may be reacted with hydrogen to form a rare earth hydride by heating the rare earth in hydrogen at about 300 to 600° C. As the rare earth reacts with hydrogen, the hydride is formed (i.e. comminuted) as a powder which may have the desired small size noted above or may be further comminuted to size. As shown in FIG. 1, the rare earth hydride may later be dehydrogenated just prior to reaction with polonium. The rare earth hydride may then be dehydrogenated or decomposed by heating the porous rare earth hydride-matrix metal element in a vacuum until the hydrogen removed is equal to that initially reacted.

As to rare earth type material, scandium, yttrium, and the lanthanides or rare earths (preferably of high purity, e.g., above 99%) may be employed for reducing to particles or powder and incorporation into the matrix metal. These "rare earth type materials" subsequently combine with the employed radioisotope to form low-volatility compounds. With polonium as the radioisotope they form polonides having substantially lower vapor pressures at given temperatures. The scandium, yttrium or lanthanides material may be outgassed in a manner generally similar to the hereinabove referred to in connection with matrix metal.

The ratios of matrix metal to rare earth type material may be selected as desired, for example, in weight ratios from about 5:1 to about 100:1.

After thorough intermixing of the particles of matrix metal and the rare earth type material, forming may be carried out by confining and compacting the intermixed particle in a press or mold by known techniques, and at appropriate pressures, e.g., from 25,000 to 225,000 pounds per square inch, to provide porous elements of less than the solid density of the particular material or materials, e.g., from about 55% to 80% of the theoretical solid density of tantalum depending on the desired crush strength and reactivity. The element formed may be of any suitable shape but preferably is such as to provide maximum heat transfer area per mass of material. One such shape is a thin-walled annulus or hollow ring of cross section such as circular or rectangular. However, the element may be in the shape of a thin flat plate of relatively large major surfaces.

As shown in FIG. 2, the intermixed powder or particles 10 are confined within an outer ring 20 forming a die and subjected to pressure by a downwardly movable punch member 22, the powder being confined and compacted between inner surfaces of ring 20 and outer surfaces of an upwardly extended bottom punch or projection 24 and between the bottom or base member 26 and the punch member 22. The press parts may be largely of stainless steel. After being compacted into a relatively thin annular configuration the bottom or base member 26 of the press may be removed and a ring base 28 substituted therefore, so that further downward movement of punch 22 serves to eject the bottom punch or projection 24 and compacted

ring 10 through the opening in ring base 28. Thereafter the compacted ring 10 may be removed from the upwardly extended projection of the bottom punch member.

If desirable, a lubricant, e.g., zinc stearate, may be employed to facilitate movement of various parts and separation of the compacted element.

Any other method of forming or shaping the element to desired shape may be employed, for example, isostatic pressing, hot pressing, etc., followed by machining or other shaping if felt necessary toward production of the resulting desired shape.

Density gradients between or within different portions of an element, especially relatively long elements, may be minimized by pressing the intermixed powders in increments. That is, a portion of the required material needed to form a complete element may be placed in the die and compacted by the plunger at the desired pressures noted above. After a period of dwell, such as about 0.5 to 2 minutes, the plunger may be retracted and another portion of material added and pressed in the die. The process may be repeated until an element having the desired dimensions is obtained. The employment of higher pressures of course results in greater compactness or densities of the elements than low pressures. Good results are obtainable at, for example, 50,000 to 60,000 pounds per square inch, with the referred to pressing increments to minimize density gradient, the resulting pressed part achieving a density of around 55% to 80% of the theoretical density of solid tantalum.

Thus far in production of the element there has been no radioisotope or radioactivity involved and hence no radioactivity exposure to personnel. Radioactive material, e.g., polonium²¹⁰, need not go into the process or porous element until just prior to the time at which it is desired so encapsulate the fuel material.

When the time arrives for rendering the porous element radioactive for use as a heat source, it may first be dehydrogenated, where appropriate, by heating at about 1000° C. in a vacuum and then placed in a container or reactor such as a quartz tube containing platinum gauze coated with radioisotope polonium²¹⁰, and the tube appropriately evacuated and sealed. The polonium plated gauze inside the tube may then be heated to vaporize the polonium and cause it to pass to and impregnate the porous element and come into intimate contact with the rare earth or related material that is interspersed throughout the element. Thereafter the now impregnated element may be heated to effect reaction between polonium and the scandium, yttrium or lanthanides material and form the polonides thereof. If desired the quartz tube or reactor may be sealed into sections after initial vaporizing of the polonium so as to separate the resulting element from the polonium gauze. If deemed desirable, after completion of the reaction the unreacted polonium may be distilled to the cooler end of the tube and separated from the resulting element.

Instead of employing the referred-to quartz tube, the desired element and polonium may be enclosed within a "bomb" and the latter heated to initiate the exothermic reaction. The "bomb" or container may thereafter serve as a storage container for the formed and now radioactive element until such time as it is desired to remove and use the element.

By way of example, the rare earth, in porous elements of tantalum and rare earth (previously hydrogenated and dehydrogenated) impregnated with polonium²¹⁰, reacted with substantially all the polonium at temperatures of between about 600° C. to 800° C.

The amounts of scandium, yttrium or lanthanides material selected and employed will, after reaction with the polonium or other radioisotope, determine the power output and operating temperature of the resulting heat source element.

After the radioactive heat source elements have been prepared as above described they may be employed, either

5

singly or in plurality, as a heat source, such as illustrated in FIG. 3. As there shown, an inner capsule 30 houses two annular heat elements 32 and is closed at opposite ends by lids or covers 34 sealed thereto. The inner capsule is indicated as closely fitting and contacting surfaces of the heat source element in order to facilitate heat transfer outwardly. The single capsule may be deemed sufficient or a further outer capsule or cylinder 36 may contact and protect the inner capsule and its element, with surfaces of the two capsules closely fitting against each other for further efficient heat transfer. Evolved helium from radioactive material in elements 32 may pass directly through the pores of the element matrix to the cavity within elements 32. Helium may accumulate within the cavity or it may be permitted to escape from the containers or capsules through any suitable venting mechanism (not shown). A typical heat source may include one or more heat elements having sufficient radioactive material included therein to generate a desired amount of heat. Individual elements may commonly have dimensions of about 0.100 inch wall thickness, one inch inside diameter and one inch long. Such an element may have about 3.14 square inches of exposed heat transfer area. A typical radioactive element incorporating a rare earth to matrix ratio of about 10 to 1 compacted to about 64% of the theoretical density of tantalum may have a power density of about 77.2 watts/cubic centimeter and a thermal conductivity of about 7.2×10^{-2} cal./sec. cm.²/° C./cm. at 290° K.

It will be seen that the present invention provides new and improved radioactive heat source elements which may be produced with minimum danger of radioactivity hazard to personnel, the radioactive isotope being incorporated into a porous element at a late stage of manufacture. The porous nature of the heat source element facilitates ready escape of helium gasses which may evolve during decay of the radioactive isotope, to minimize possible deformation, cracking, or the like, of the element. The element is preferably in such configuration as to provide for maximum heat transfer surface and therefore maximum efficient heating of adjacent elements, heating of operating or thrust gasses, etc.

It will be understood that various changes in details, materials and arrangements of items may be made by those skilled in the art within the presentation and scope of the invention, as expressed in the appended claims.

What is claimed is:

1. The method of making a radioactive heat source element comprising comminuting matrix metal to particles of size not greater than about 250 microns; com-

6

minuting to particles of size not greater than about 57 microns at least one material selected from the group consisting of scandium, yttrium and the lanthanides; combining said matrix metal particles and said material particles in ratio by weight of at least 5 to 1 and thoroughly intermixing them; confining said intermixed particles and compacting them to density in the range of from about 55% to 80% of the normal density of the matrix metal for forming an integral and cohesive but porous element; and thereafter impregnating said porous element with vaporous polonium radioisotope but retaining porosity of the element for escape therefrom directly through said pores of helium resulting from polonium radioisotope decay.

2. The method of claim 1, together with heating the impregnated element to react the polonium and said material and form the polonide of said material.

3. The method of claim 1, together with compacting said confined particles in a plurality of increments for minimizing density gradients in the resulting element.

4. The method of claim 3 wherein said compaction is at pressures in range from about 25,000 pounds per square inch to about 225,000 pounds per square inch.

5. The method of claim 1, together with forming said element into a configuration of thin cross-sectional dimension as compared with exposed area thereof.

6. The method of claim 1 wherein said matrix metal is selected from the group consisting of tantalum, tungsten, chromium and iridium.

7. The method as claimed in claim 1, wherein said matrix metal comprises tantalum.

8. The method of claim 1 including the steps of heating said material before intermixing thereof with said metal particles in a hydrogen atmosphere to hydrogenate said material and heating said porous element in a vacuum to dehydrogenate said material.

9. A radioactive heat source element produced by the method of claim 1.

References Cited

UNITED STATES PATENTS

3,154,501	10/1964	Hertz	252—301.1
3,161,504	12/1964	Black et al.	261—.5 XR
3,330,889	7/1967	Samos et al.	264—.5
3,431,328	3/1969	Case et al.	264—.5

CARL D. QUARFORTH, Primary Examiner

S. R. HELLMAN, Assistant Examiner

U.S. Cl. X.R.

264—0.5