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METHOD OF MAKING MEANS FOR VENTING HELIUM FROM A RADIOISOTOPE CONTAINER

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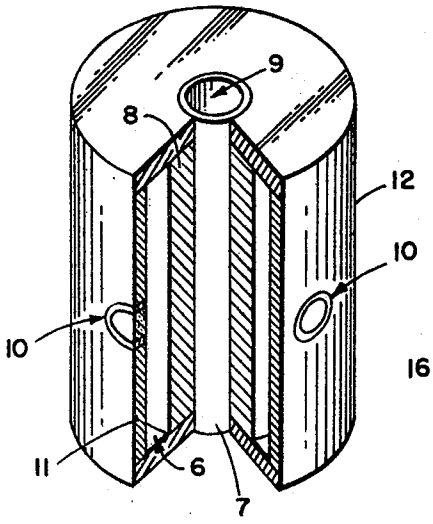


Fig. 1

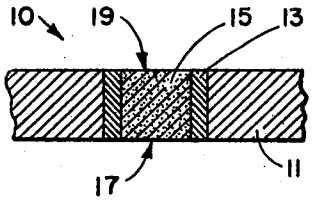


Fig. 1a

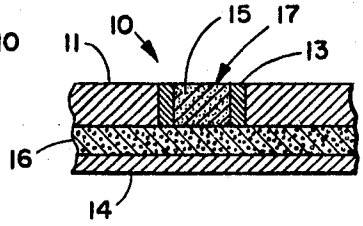


Fig. 1b

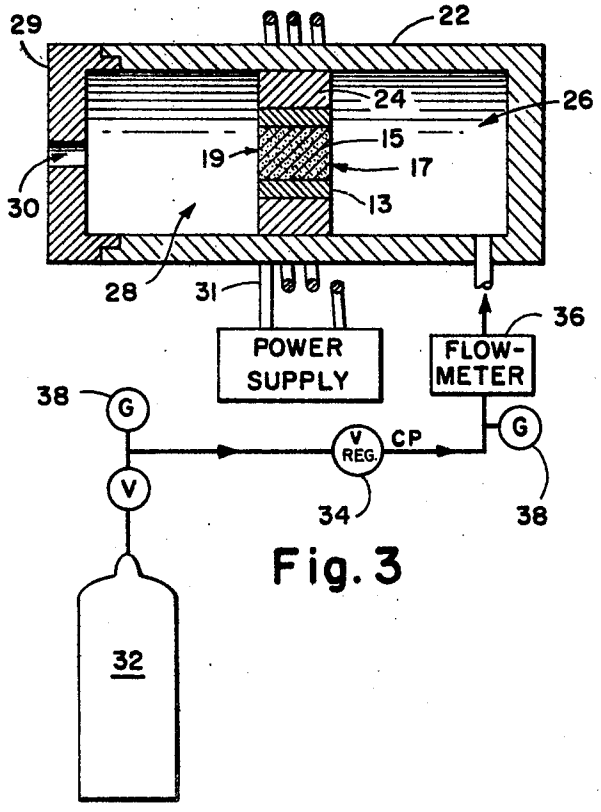


Fig. 3

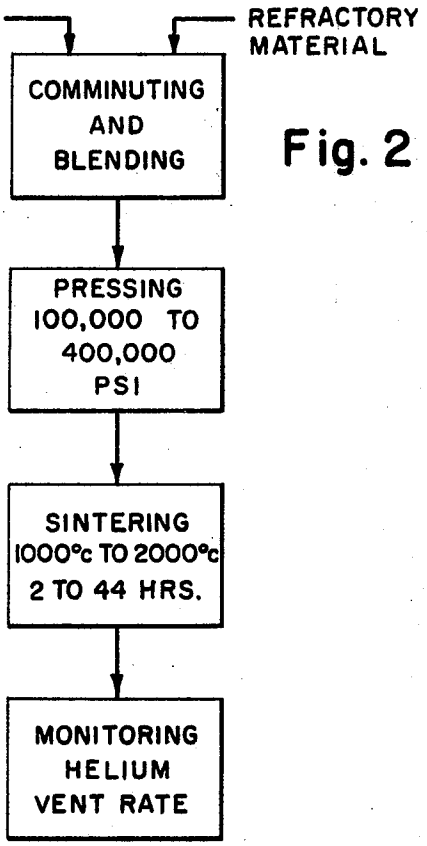


Fig. 2

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METHOD OF MAKING MEANS FOR VENTING HELIUM FROM A RADIOISOTOPE CONTAINER

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ABSTRACT OF THE DISCLOSURE

Filter means and method of making the same for venting helium gas through a radioisotope container wall and preventing or minimizing escape of radioisotopes and radioactivity, comprising a porous tablet forming a part of the wall including a pressed and sintered particulate blend of a refractory metal and a heat resistive material to provide porosity and strength at elevated temperatures.

BACKGROUND OF INVENTION

Many radioactive heat sources employ as fuel radioisotopes such as plutonium-238 and polonium-210 which emit alpha particles. The alpha particles may acquire electrons to become complete helium atoms. Helium may buildup undesirably high pressures, e.g., exceeding 1500 p.s.i., within the heat source housing if not vented. In many cases a radioactive heat source may operate at elevated temperatures such as about 2000° C., which may weaken the housing or container walls as well as further increase helium pressure within the heat source container. Consequently, materials which have high strength at high temperatures must be used for enclosing such helium producing radioisotopes. Frequently double walls of thick, high strength material are necessary to minimize or prevent rupture and escape of the radioactive radioisotopes. The additional weight resulting from the use of such high strength heat source housing materials may present a difficult problem in rocket and space vehicle applications as well as other uses where excess weight is undesirable.

Venting or releasing the helium pressure accumulated within the heat source container may be difficult to accomplish. Helium should be allowed to escape while radioactive elements must be contained within the housing. Possible high operating temperatures and pressures limit the choice of materials which may be used in constructing a suitable helium venting device. Material choice is further complicated by the chemical reactiveness of radioisotope and heat source materials which may be employed.

Other problems arise in producing helium venting devices with predictable or ascertainable vent rates. The vent rate must be adequate to offset helium production by the radioisotope. However, if helium is freely vented to the atmosphere, radioisotope escape may occur. Therefore selectivity or ability to pass helium gas while occluding other materials or particles must be included as criteria for design of a satisfactory helium vent.

SUMMARY OF INVENTION

In view of the problem discussed above it is therefore an object of this invention to provide a means for venting helium from a container while preventing escape of other elements.

It is a further object to provide a filter having high structural strength at elevated temperatures.

It is also an object to provide a composition for such a filter which is chemically compatible with common radioisotopes and heat source materials at elevated temperatures.

It is a further object to provide a method for preparing a helium vent means which will accomplish the above objects and discharge a predictable rate of helium free from radioactive contamination.

Various other objects and advantages will become apparent from the following description of embodiments of the present invention.

The invention comprises a filter for venting helium, while retaining other materials such as heavier atomic weight elements, including a supporting sleeve or annular member encompassing a porous cermet frit tablet which contains a sintered and pressed particulate mixture of a refractory metal and a heat resistant material.

DESCRIPTION OF DRAWINGS

The following drawings are illustrative of the present invention wherein:

FIG. 1 is a partially cutaway view of a radioactive heat source having the filter vents of the present invention;

FIG. 1*a* is a fragmentary cross-sectional view of one filter vent of FIG. 1;

FIG. 1*b* is a fragmentary cross-sectional view of an alternate embodiment of a casing for the heat source of FIG. 1;

FIG. 2 is a flow diagram of the method of producing the filter of FIG. 1 and FIG. 1*a*; and

FIG. 3 is a schematic showing of a system for sintering the filter of FIG. 1 and FIG. 1*a*.

DETAILED DESCRIPTION

Referring to FIG. 1, there is shown a radioactive radioistope heat source including a tubular shaped housing or container **12** with an axial passageway **9** for providing additional heat transfer area. A radioisotope **8** which may emit alpha particles may be pressed or molded into an annular shape and disposed within container **12** around the wall **7** (or walls if passageway shapes other than cylindrical are employed) of passageway **9**. Filter vent or vents **10** may be disposed on or affixed within the container wall **11** at one or more locations for allowing helium gas produced by the radioisotope to escape. An annular space **6** may be left between the radioisotope **8** and the container wall **11** to facilitate flow of helium to the filter vents **10**. Radioisotope **8** may be any conventional radioisotopic fuel such as plutonium-238 dioxide microspheres, alloys of plutonium-238, polonium-210 or any other conventional alpha emitting radioisotope.

In FIG. 1*a*, a fragment of the container wall is shown with a retained annular member or sleeve **13**. The annular member **13** may be a continuation of wall **11** or a separate sleeve member pressed, welded or otherwise suitably secured to the wall, preferably such that its annulus or central opening traverses the wall thickness. Sleeve **13** may comprise a material having high strength at elevated temperatures such as tantalum or tungsten or alloys thereof which is compatible with the particular radioisotope fuel used. A porous cermet frit pellet or tablet **15** of suitable shape and dimension may be pressure fitted or fastened, such as by electron beam welding, into the central opening of sleeve **13** which may give lateral support and structural strength to the cermet frit material. Alternatively, tablet **15** may be pressed or formed within sleeve **13** serving as a mold. The tablet **15** may form a portion of both the internal surface **17** and the external surface **19** of container wall **11** such that a continuity of porous frit material providing a vent path extends from the inside to the outside of container or heat source housing **12**. Any helium accumulation within the container **12** may

thus vent from internal surface **17** through tablet **15** and outer surface **19** to the surrounding atmosphere.

The container **12** may also be supported by an outer container wall or cladding **14**, as shown in FIG. 1*b*, separated from wall **11** by a porous, vibration absorbing member **16**. Porous member **16** may be made of tantalum or other approprate material in a sponge-like or woven pad form having sufficient porosity to contain helium vented through tablet **15**. Outer container wall **14** may be made of a material which has sufficient strength to support the container **12** under operating conditions of the heat source or may be provided with openings (not shown) or additional venting tablets (not shown) to release helium discharged from porous member **16**.

The cermet composition of tablet **15** may include a refractory metal such as tantalum; alloys of tantalum with 0.1 weight percent yttrium, 10 weight percent tungsten or 10 weight percent tungsten-2.5 weight percent hafnium; or tungsten; or other alloys of either metal in a proportion of about 80% to 99% of the total weight and a heat resistive material such as alumina, zirconia, thoria, graphite, or a combination of such heat resistive materials. Other refractory metals may also be used such as nickel which have sintering temperatures above the operating temperature of the heat source. Similarly, other heat resistive materials such as uranium carbide, triuranium octaoxide, aluminum carbide, thorium carbide and zirconium carbide as well as other materials or combinations thereof which are not excessively chemically reactive with the heat source materials and which can sustain elevated temperatures without substantial loss of strength may also be employed.

Tablet **15** may be prepared by the process shown in FIG. 2. The refractory metal and the heat resistive material may be comminuted to a particle size between about 1 micron and 150 microns and blended in suitable size reduction equipment such as a ball or hammer mill. It may be desirable to employ separate milling operations to grind or comminute each component of the cermet composition. The materials may be unequal in frangibility such that one requires more milling than the other and it may be desirable to provide the heat resistive material at a finer mesh than that of the refractory metal. The finer mesh heat resistive material particles may fill in and act as a binder between discrete refractory metal particles in the finish cermet frit. Accordingly, the particle size of the metal and especially of the heat resistive material may affect the porosity, helium venting rate and selectivity of the complete cermet tablet **15**.

After thoroughly mechanically mixing or blending the refractory metal and heat resistive material particles, as within a ball mill, the particulate mixture is pressed to form a compact and integral tablet. Compaction may be performed with a suitable press and an appropriately shaped mold, or the annular member **13** as discussed above may serve as the mold. Pressures of about 100,000 to 400,000 p.s.i. have satisfactorily been employed but pressures somewhat lower may also produce a sufficiently compact tablet for certain applications. The higher the pressure used the more compact and less porous the tablet will become thus controlling the helium vent rate. Conversely, insufficient compaction may result in inadequate structural strength and filter selectivity, that is, increased likelihood of radioactivity escape with the helium. Accordingly, an optimum pressure may be empirically derived for a particular application. Using a homogeneous mixture, pressure and temperature, parameters may be standardized for prescribed vent rates.

After pressing, the table is sintered in a suitable furnace at a temperature of about 1000° C. to 2000° C. to produce a frit-like material. Possibly all or a portion of the necessary sintering may have been provided in a hot pressing operation instead of the above described cold compaction step. Sintering may continue for a period of about two to forty-four hours depending on the cermet components selected and the desired porosity. As sintering continues, pores and passageways within the tablet material are fused closed thus reducing venting rates while improving gas venting selectivity.

FIG. 3 shows, in more or less schematic or diagrammatic form, a system for sintering the cermet tablet while passing or venting helium therethrough at a monitored rate. A container or housing **22** composed of a suitable heat resistant, furnace construction material may have a partition or supporting wall member **24** at one boundary of a helium pressure chamber **26**. Tablet **15** and optionally sleeve **13** may be affixed in a leak tight manner within partition **24** for sintering. Alternatively the tablet **15** and sleeve **13** may be fastened within the heat source container wall **11** (FIGS. 1 and 1*a*) where it will ultimately be used and sintered therein for a heat expansion fit. A second chamber **28** may receive helium vented through tablet **15** from chamber **26** and allow such gas to escape through orifice or opening **30**. Chamber **28** may shield the tablet from excessive heat loss during sintering. An induction heater coil **31** and power supply or other suitable means, e.g., a standard tube or muffle furnace, may be wound around or disposed adjacent container **22** to provide the necessary heat for sintering table **15**, without blocking the helium entry and exit faces **17** and **19** of the tablet. A helium reservoir **32** may supply gas to chamber **26** through a pressure regulating valve **34** and a flowmeter **36** which permit controlling of the helium flow rate at a constant pressure differential, between chambers **26** and **28**. Suitable pressure gages **38** provide further checks on the system operation. The vent rate through the tablet **15** may be monitored at opening **30** with a conventional mass spectrometer or residual gas analyzer (not shown), simultaneously with sintering of the tablet. When the predetermined desired vent rate is obtained, the heat is cut off and sintering operation terminated by opening the end cap **29** of chamber **28** and removing the tablet **15** and its holder **13**. The desired helium vent rate may be determinated by the alpha radiation rate of the radioisotope to be used or other helium production rate and from empirical standards correlating helium vent rate with selectivity or the ability to block escape of larger radioactive particles.

The completed cermet frit tablet **15** may be electron beam welded into sleeve **13** if not already formed therein as mentioned above. It has been found that a stronger bond may be provided between the frit **15** and annular member or sleeve **13** through electron beam welding than by merely forming the cermet frit within sleeve **13** serving as a mold. Annular member or sleeve **13** carrying the tablet **15** may than be pressed, welded or otherwise suitably affixed within or to container wall **11**.

EXAMPLE

Eight parts by weight of tantalum powder having an average particle size of about 40 microns were mixed with one part by weight of zirconium dioxide (zirconia) having an average particle size of about 20 microns. The blended powder was then pressed at about 300,000 p.s.i. to form a tablet in the shape of a flat, right-circular cylinder having a ⅜ inch diameter and a thickness or length of ⅛ inch (other sizes and shapes may be also satisfactory). These cylinder or tablet was heated to about 1600° C. and sintered for about 16 hours, followed by sintering at about 1800° C. for about 30 minutes more. The sintered tablet was then permanently mounted into a tantalum sleeve by electron beam welding.

During sintering the flow of helium through the tablet is preferably continually monitored and heating terminated and the tablet removed from the sintering furnace upon achievement of the desired leak rate known to facilitate helium venting and blocking of radioactive particles.

Other examples and typical process parameters are illustrated in the following table.

TABLE

Sample	Refractory metals: Parts	Material	Particle size (microns)	Heat-resistant materials: Parts	Material	Particle size (microns)	Pressing pressure (p.s.i.)	Sintering condition: Temp. (° C.)	Time (hours)	He flow rate through tablet (cc./sec.)
1	5	Ta	150	1	Al_2O_3	150	180,000	1,000	12–20	1×10^{-4}
2	10	Ta	40	1	Al_2O_3	1.0	160,000	1,000	12–20	1×10^{-5}
3	20	Ta	40	1	Al_2O_3	1.0	175,000	1,000	12–20	1×10^{-6}
4	8	Ta	40	1	Al_2O_3	0.1	350,000	1,320	22	1.6×10^{-4}
5	8	Ta	40	1	Al_2O_3	0.1	400,000	1,200	14	1.3×10^{-4}
6	6	Ta	40	1	ZrO_2	1.0	300,000	1,600	44	1×10^{-7}
7	10	W	5–10	1	ThO_2	1–3	200,000	1,500	2	8.8×10^{-6}
8	40	W	5–10	1	Graphite		100,000	1,500	2	4×10^{-3}
9	80	W	5–10	1	do		100,000	1,500	2	1.4×10^{-3}

The invention provides a helium vent and filter capable of performing at the elevated temperatures found in a radiisotope heat source. A method is set forth for preparing such a filter with the desired porosity or helium vent capacity and selectivity as well as structurally reinforcing the filter within a container wall. Suitable compositions are presented which have strength at elevated temperatures and may be manufactured into the filter of the present invention.

It will be understood that various changes may be made in the details of the components, parts, or steps, described herein to illustrate the invention, by one skilled in the art within the scope of the claims.

What is claimed is:

1. The method of making means for venting helium from a radioisotope container and retaining in the container radioactive particles comprising comminuating a refractory metal selected from the group consisting of tantalum, tungsten, and alloys thereof to a particle size of about 1 to about 150 microns; comminuating a heat resistive material selected from the group consisting of alumina, aluminum carbide, thoria, thorium carbide, zirconia, zirconium carbide, uranium carbide, triuranium octaoxide. graphite, and mixtures thereof to a particle size no greater than that of said refractory metal particles; intermixing said metal particles and material particles; pressing said intermixed particles together at about 100,000 to 400,000 pounds per square inch to form a coherent tablet; sintering said tablet at about 1000° C. to 2000° C. by applying heat about the periphery of the tablet. simultaneously with said sintering subjecting a face of the tablet to helium at a pressure above the pressure at an oppositely disposed face of the tablet to effect passage of helium through the tablet; monitoring said oppositely disposed face for passage of helium through the tablet concurrent with said sintering; and terminating said sintering upon achievement of predetermined helium flow rate through the tablet.

2. The method as claimed in claim 1, wherein said intermixed particles are peripherally confined by an annular mounting member during said pressing.

3. The method as claimed in claim 1, wherein said sintering is conducted for a period of about 2 to 44 hours.

4. The method of claim 1 wherein said sintering comprises heating said tablet to a first temperature, maintaining said tablet at said first temperature for a period of time, heating said tablet to a second temperature higher than said first temperature, and maintaining said tablet at said second temperature a second period of time.

References Cited

UNITED STATES PATENTS

2,852,366 9/1958 Pinner ..... 75—201

CARL D. QUARFORTH, Primary Examiner

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U.S. Cl. X.R.

29—182.5, 182.7, 182.8; 75—201, 203, 213, 214, 221, 222, 224