Jan. 7, 1969 J. J. FITZGERALD ET AL 3,421,001

RADIOISOTOPIC HEAT SOURCE AND METHOD OF PRODUCTION

Filed March 16, 1964 Sheet 1 of 4

FIG. 1

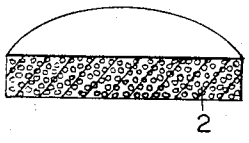
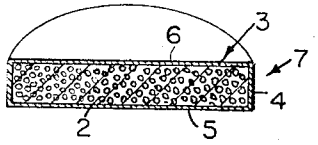


FIG. 2



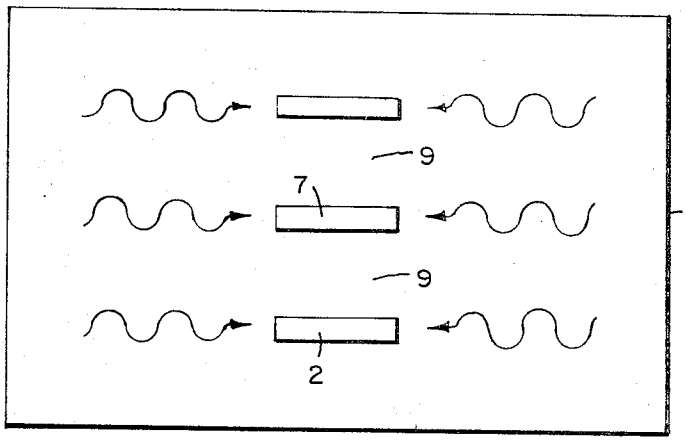


FIG. 3

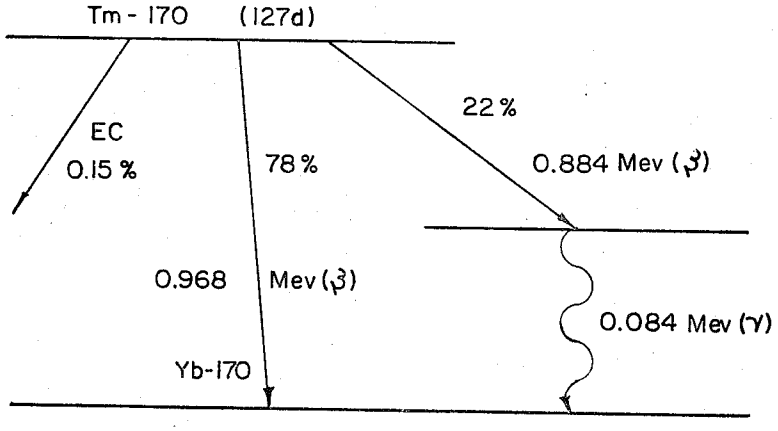


FIG. 4

Joseph J. Fitzgerald, Gordon L. Brownell INVENTORS
BY
Ezekiel Wolff, Wolff & Greenfield
ATTORNEYS

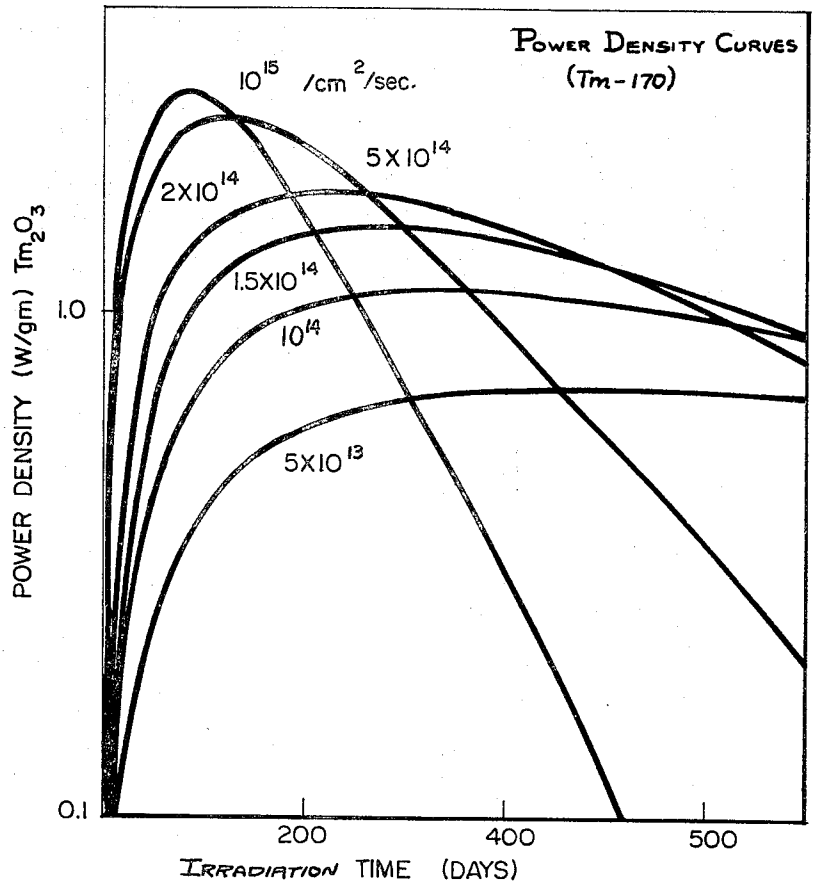


FIG. 5

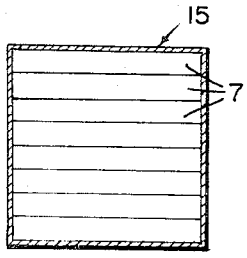


FIG. 6

Joseph J. Fitzgerald, Jordan L. Brownell INVENTORS
BY
Ezekiel Wolf, Wolf & Greenfield
ATTORNEYS

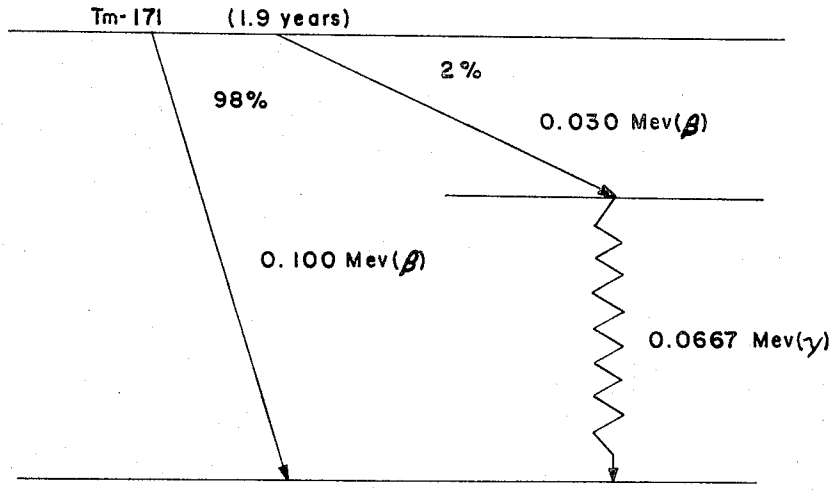


FIG. 7

INVENTORS
Joseph J. Fitzgerald,
BY Gordon L. Brownell
Wolff, Greenfield & Hieken

RADIOISOTOPIC HEAT SOURCE AND METHOD OF PRODUCTION

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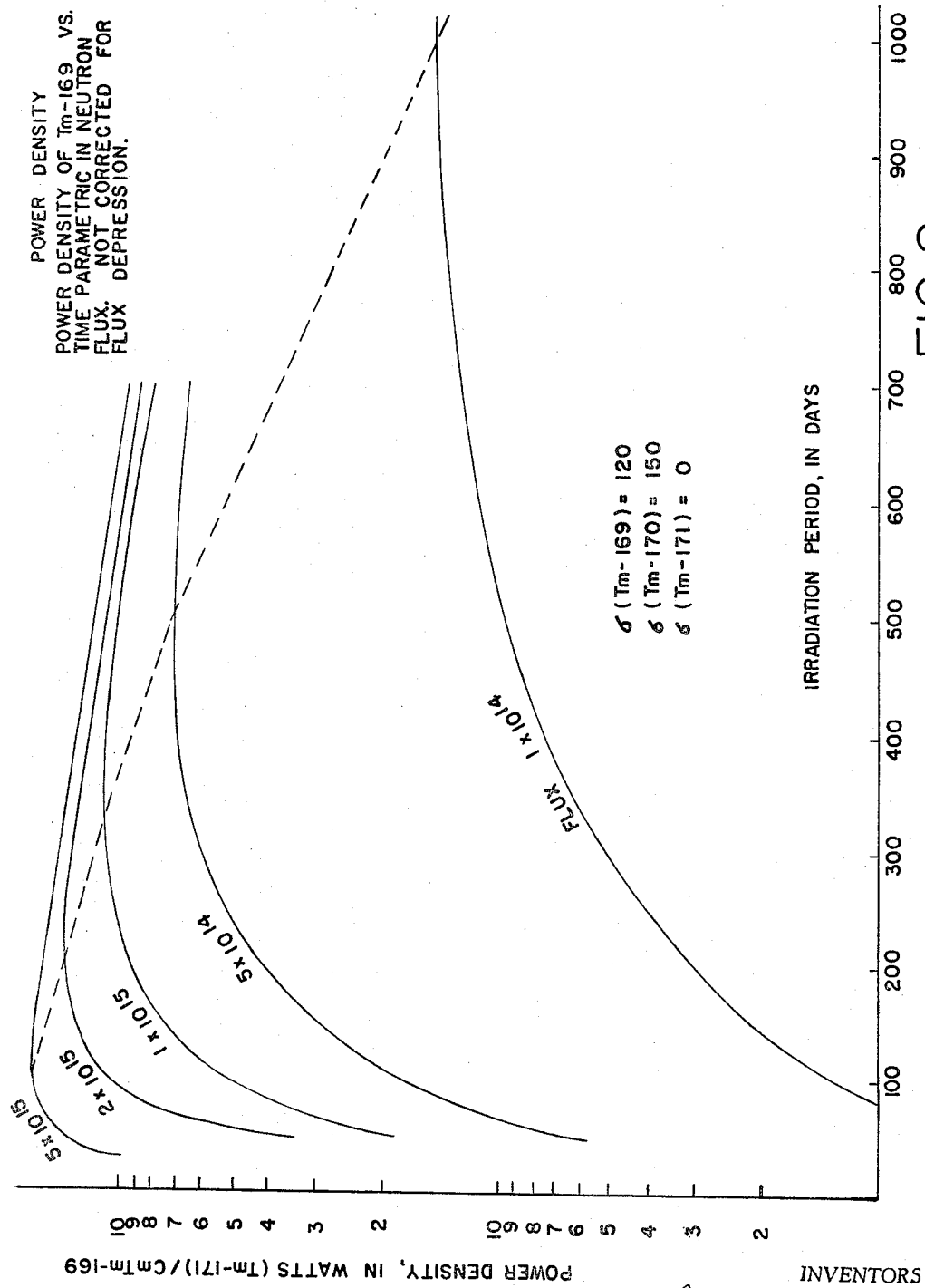

FIG. 8

INVENTORS

BY Joseph J. Fitzgerald,
Gordon L. Brownell
Wolf, Greenfield & Hieken.

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3,421,001
RADIOISOTOPIC HEAT SOURCE AND METHOD OF PRODUCTION

Joseph J. Fitzgerald, Winchester, and Gordon L. Brownell, Weston, Mass., assignors to Iso/Serve, Inc., Cambridge, Mass., a corporation of Massachusetts

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U.S. Cl. 250—106 18 Claims
Int. Cl. G21h 5/00

The present invention relates to a means and method of providing heat and ionizing radiation sources for energy generation and material irradiations and more particularly relates to a means and method of providing "cold encapsulated" radioactive heat sources which may be used as energy generating components; as product and food sterilizers; and as plastic and detergent irradiators.

There has been an increasing demand for radioactive heat sources for energy purposes which are included in such devices as thermionic and thermoelectric generators. Heretofore, attempts have been made to provide such heat sources by the fabrication of radioisotope capsules or wafers that are incorporated into such devices as energy sources. Great difficulty, however, has been encountered in the suitable manufacture of such components in view of their preexisting radioactive nature. Prior to the development of the present concept of cold encapsulation, sources of Sr-90, Ce-144, and Pu-238 that are radioactive were "radioactively hot encapsulated." Because of the inherent dangers in the handling and encapsulation of these radioactive components, elaborate facilities are necessary at substantial cost to provide adequate radiation protection. These elaborate facilities greatly reduce the flexibility in design and fabrication of such heat sources and, consequently, result in greater production and handling costs.

There has also been an increasing demand for radiation sources to reduce the bacteria content in food and some nonedible products and to change the properties of certain materials such as plastics and detergents. Heretofore, costly high energy isotope sources that require considerable shielding have been used for such purposes. In some instances, accelerators have also been used for the same purposes. But both high energy isotopes and accelerators require considerable space and shielding. However, the use of cold encapsulated relatively low energy sources permit the relatively inexpensive preparation of a source that needs a relatively small amount of shielding. Space requirements and shielding weight is so low that the unit can be readily moved from place to place.

It is, therefore, an object of the present invention to provide a means and method of fabricating wafers or discs that can be made radioactive for use as heat sources for energy generation or for other applications requiring ionizing radiation sources.

It is also an object of the present invention to provide a means and method for fabricating heat and ionizing radiation sources that reduce hazards normally associated with preparing radioisotope sources. The present invention also eliminates the necessity for elaborate nuclear or radiation facilities with a consequent reduction in cost for the preparation of radioactive sources.

A further object of the present invention is to provide a method of encapsulating a stable source or non-radioactive material in an encapsulating material that is also stable or nonradioactive. Another object of this invention is to provide for the cold encapsulation of a stable material having a relatively high thermal neutron cross section in a stable encapsulating material having a relatively low thermal neutron cross section with a relatively short half life. With the cold encapsulation method, the radioactively inert or stable material may be subjected thereafter to a neutron flux of a specific level to induce sufficient activity into the material for its use as a heat source or a materials irradiator.

The present invention also provides a means and method by which stable material suitable for exposure to neutron flux for use as a radiation source may be fabricated and stored prior to exposure to neutron flux for indefinite periods of time.

In the present invention there is provided a means and method in which a stable material which has a relatively high thermal neutron cross section and a relatively long half life may be encapsulated prior to neutron exposure in such a manner that the encapsulation containing the stable material is essentially inactive and safe for handling. This stable material may, after fabrication and encapsulation, be subjected to a neutron flux. As this means and method does not contain radioactively "hot" material but rather inert or "cold" material, it is called a "cold encapsulation" process.

These and other objects of the present invention will be more clearly understood when considered in conjunction with the accompanying drawings in which:

FIG. 1 is a cross section of a stable compressed and sintered compound prior to irradiation;

FIG. 2 is a cross section of an encapsulation formed in accordance with the present invention;

FIG. 3 is a schematic illustration of a plurality of encapsulations during irradiation;

FIG. 4 is a graphical illustration of a decay scheme for thulium-170;

FIG. 5 is a graphic representation of power density of thulium oxide versus time, parametric in neutron flux;

FIG. 6 schematically illustrates a group of packaged capsules;

FIG. 7 is a graphical illustration of a decay scheme for Tm-171; and

FIG. 8 is a graphic representation of power density of thulium oxide versus time, parametric in neutron flux.

This invention is directed primarily toward an encapsulation of a compound or isotope which is stable, compressed and sintered and which is made adaptable for irradiation into an active heat source in its encapsulated form. As the source is fabricated and encapsulated before the isotope is subject to irradiation it may be referred to as a "cold encapsulation" process.

The means and methods described may be used in connection with several different materials, although certain isotopes have properties which make their use particularly desirable as practical heat sources. The material should have a thermal neutron cross section which is high, preferably in excess of 5 barns and a half life preferably in excess of 100 days. The material also should have no significant gaseous daughter products formed during emission. For certain aplications, notably for use in thermionic generators, the material should have a high melting point, preferably in excess of 1700° C., although for other applications this is not a consideration. It has been found that thulium-169 prepared as thulium oxide Tm_2O_3 is the most practical and technically feasible material for use in cold encapsulation. This is due in part because it is the only stable isotope of thulium (100% abundant) and has a thermal neutron cross section of 118 barns which assures a sufficient activation to thulium-170, or with neutron fluxes in the range of 2 to 5×10^{15} n./cm.^{2}/sec., suitable and desirable quantities of thulium-171 can be induced into the thulium oxide encapsulated wafer. Thulium-169 in the form of thulium oxide particles is prepared by any suitable commercial method. Thulium oxide is used because pure thulium-169 as a solid metal may react with the material forming the casing of the encapsulation and would fuse to the encasing material.

The thulium oxide is compressed and sintered under heat and pressure conditions into a wafer or disc form as shown in FIG. 1. The specific dimensions are determined at least in part by the particular ultimate power or radiation level for which the unit is designed. Generally, flux depression during irradiation may be lessened by making the wafer relatively thinner, while still maintaining the structural integrity of the unit for maximum efficiency. A typical wafer would, for example, have a thickness of 2 to 5 mm. and a diameter of 1" to 2½". The closer the thulium oxide approaches its theoretical density, the better will be the power output efficiency since the maximum power per unit dimension is a function of density. In order to achieve maximum power density, it is desirable to compress the thulium oxide to a density of at least 80% of its theoretical maximum density, and most preferably to a range of 90% to 95% of theoretical density. The thulium oxide wafer is formed by compressing thulium oxide powder, utilizing conventional equipment at an elevated temperature of just below the melting point of thulium oxide which is in the range of approximately 2300° C. to 2600° C. Sintering the compressed wafer may be conducted in air, vacuum or in an inert atmosphere.

The thulium oxide wafer **2** in FIG. 1 is placed in and secured to a casing **3** of FIG. 2. The material of which the casing **3** is formed must have a high melting point which is at least in excess of the melting point of the material contained within it. In the case of the preferred embodiment, the melting point must be in excess of that of thulium oxide. The casing material should not be conducive to significant activation and, therefore, it should have a very low cross section for absorption of thermal neutrons and a short half life. Preferably, the casing should be of material having a thermal neutron cross section of less than .2 barn and a half life of less than 3 days. The casing material must not react with the fuel material or isotope which forms the wafer and also must be capable of being joined to form a sealed casing with the fuel material contained within it. It has been found that molybdenum is the most preferred material for the casing although other materials such as zirconium and tungsten might also be used. Molybdenum is preferred primarily because of its high melting point, relatively short half life and other suitable characteristics.

The casing **3** should have relatively thin walls, with the walls having a thickness in the range of 5 millimeters or perhaps less. By making the casing as thin as possible, without weakening its structural integrity, flux depression during irradiation may be minimized and, therefore, neutron absorption of the fuel material may be maximized and the power density increased. The sidewalls **4** and bottom **5** may be integrally formed and may be joined to the cover **6**, with all portions of the casing **3** joined to the fuel material **2** by conventional methods. Conventional means may, for example, comprise electron beam welding in a vacuum. It is most desirable to join the casing to the fuel material, in this case thulium oxide, to assure an appropriate contact between the fuel wafer and the container, so as to conduct heat effectively from the fuel material to the container for greater efficiency.

The encapsulation **7** illustrated in FIG. 2, when initially formed is not radioactive, and, therefore, may be termed a cold encapsulation. This wafer requires no shielding and may be handled as an inactive material.

Inactivated capsules **7** may be placed in a reactor **8**, as schematically illustrated in FIG. 3, for activation and held in the reactor until just prior to use. For most applications, the capsules are placed in a reactor for a period of not less than 35 days and not more than 150 days and are exposed to a flux equal to or greater than 10^{14} n./cm.^{2}/sec. for production of thulium-170. For production of thulium-171 the capsules are exposed to a flux in the order of 10^{15} n./cm.^{2}/sec. for a period of 30 to 90 days. In order to minimize flux depression a spacing of in the order of at least 5 to 1 between capsules

in the reactor is utilized. Thus, in the preferred embodiment, the capsules **7**, having a thickness of approximately 2 millimeters are spaced a distance apart of approximately eight millimeters. The eight millimeters space **9** between wafers should be occupied by a material which is capable of moderating the neutrons and cooling the areas, so that the neutrons will be slowed down to a speed at which they will more effectively be absorbed by the fuel material. Any suitable cage may be used to separate and support the capsules in the reactor, including for example, a cage of molybdenum or aluminum. The moderating material between the wafers should be a hydrogenous material, such as water. In the preferred embodiment, the flux depression is expected to be in the order of 50%. In actually determining the flux depression, not only must the spacing of the wafers within the reactor be considered, but also the thickness of the wafer is a controlling function.

FIG. 5 illustrated the power density of thulium oxide versus time, parametric in neutron flux that may be attained. This graph is based upon the assumption that the flux exists at the target material. As is evident from an examination of FIG. 5, thulium oxide has a reasonably high specific activity that can be achieved at reactor fluxes which are generally available today with the radiation times within reason.

When exposed to the neutron flux, the thulium-169 in the form of thulium oxide absorbs neutrons which convert the stable thulium oxide to active thulium-170. The reaction that takes place by irradiating thulium-169 in a neutron flux to produce thulium-170 is:

$$_{69}Tm^{169} + {}_{0}n^{1} \longrightarrow {}_{69}Tm^{170} + {}_{-1}e^{0} \longrightarrow {}_{70}Yb^{170}$$

The thulium-170 decays to a stable daughter ytterbium-170 as illustrated in FIG. 4. However, thulium-170 has a cross section of 150 barns which indicates some of the thulium-169 is converted into thulium-171, according to the equation:

$$_{69}Tm^{170} + {}_{0}n^{1} \longrightarrow {}_{69}Tm^{171} + {}_{-1}e^{0} \longrightarrow {}_{70}Yb^{171}$$

The Tm-170 and Tm-171 which are formed by subjecting the wafer to a neutron flux appear to be the most practical and technically feasible material for use as a heat source, particularly in connection with thermionic generators. This is due at least in part to the attainable power densities which may be achieved with thulium-170 fuel wafers, and the longer half life of Tm-171 and the radiation safety aspects.

The activated encapsulated wafers **7** may be stacked and contained within an outer casing as illustrated in FIG. 6. The encapsulated wafers **2** of the preferred embodiment which may have a width of 2 millimeters are stacked to various heights.

The outer casing **15** within which the wafers **2** are contained should preferably be formed of the same material as the casing material **3**. In addition, the thickness of this outer casing should be substantially the same thickness as the thickness of the casing **3** and may be sealed in a similar way to casing **3**.

While thulium-169, the only stable isotope of thulium, is a preferred stable material for use in the present invention, other materials have also been considered. Thulium-171 can also be produced by the use of stable erbium or enriched Er-170 in the following manner:

$$_{68}Er^{170} + {}_{0}n^{1} \longrightarrow {}_{68}Er^{171} \longrightarrow {}_{-1}e^{0} + {}_{69}Tm^{171}$$

The above process requires a chemical separation of Tm-171 from the erbium isotopes or Er-170. Consequently, the encapsulation is ultimately accomplished with radioactively "hot" material, Tm-171. Since the energy emitted per disintegration from Tm-171 is very low, the radiation safety problems are very small.

What is claimed is:

1. A method of preparing a radioactive heat source encapsulation comprising:

preparing a wafer of stable thulium oxide for encapsulation by compressing and sintering particles of thulium oxide under the influence of heat,

placing said compressed and sintered wafer in a capsule of molybdenum, joining said wafer to the walls of said capsule,

sealing said capsule, and, thereafter subjecting said nonradioactive thulium oxide to a neutron flux of at least substantially 10^{14} n./cm.^{2}/sec. for a period of between approximately 35 and 150 days.

2. A method as set forth in claim **1** wherein said nonradioactive thulium oxide is subjected to a neutron flux of at least 10^{15} n./cm.^{2}/sec. for a period of between 30 and 90 days for production of thulium-171.

3. A method of preparing a radioactive heat source encapsulation comprising:

preparing a wafer of stable thulium oxide for encapsulation by compressing and sintering particles of said thulium oxide into an integral wafer,

placing said wafer in a capsule formed of material which has a melting point in excess of 2300° C., is not chemically reactive with said thulium oxide, is capable of being joined to said thulium oxide, has a cross section of less than .2 barn to thermal neutrons and half life of less than 3 days, sealing said capsule, and thereafter subjecting said nonradioactive thulium oxide to a neutron flux of at least substantially 10^{14} n./cm.^{2}/sec. for a period of between approximately 35 and 150 days.

4. A method as set forth in claim **3** wherein the specific activity of said thulium oxide after radiation is in the order of 5 to 15 watts/cc. from Tm-170.

5. A method as set forth in claim **3** wherein said thulium oxide is subjected to a neutron flux of at least 10^{15}n./cm.^{2}/sec. for a period of at least 30 days for production of thulium-171.

6. A method as set forth in claim **5** wherein said thulium oxide is subjected to said neutron flux until the specific activity of said thulium oxide after radiation is in the order of .5 to 1.5 watts/cc. from thulium-171.

7. A method as set forth in claim **4** wherein said capsule is formed of material selected from a group consisting of molybdeum, zirconium and tungsten.

8. A method as set forth in claim **7** wherein said thulium oxide is compressed to a density in excess of 80% of theoretical maximum density.

9. A method of preparing a radioactive heat source encapsulation comprising:

preparing a wafer of stable material having a cross section of greater than 5 barns to thermal neutrons,

placing said wafer in a capsule formed of material which is nonreactive with said wafer material, is capable of being sealed around said wafer material,

has a cross section of less than .2 barn to thermal neutrons and a half life of less than 3 days,

sealing said capsule, and

thereafter subjecting said nonradioactive material to a sufficient neutron flux for a period whereby the specific activity of said material after radiation is in excess of 0.5 watt per cc. of said material.

10. A nonradioactive encapsulation heat source adapted to be converted by exposure to neutron flux to an isotope fueled heat source, comprising:

a wafer of a stable compressed sintered isotope having a cross section of greater than 5 barns to thermal neutrons and a half life in excess of 100 days,

said wafer entirely contained and enclosed in a sealed capsule of material which has a melting point in excess of 2300° C., is nonreactive with said compound, is capable of being joined to said isotope, has a cross section of less than .2 barn to thermal neutrons, and a half life of less than 3 days.

11. An encapsulation as set forth in claim **10** wherein said wafer is joined to the inner walls of said capsule.

12. An encapsulation as set forth in claim **11** wherein said wafer is formed of thulium oxide.

13. An encapsulation as set forth in claim **12** wherein said wafer is formed of thulium oxide and said capsule material is selected from a group consisting of molybdenum, zirconium and tungsten.

14. An encapsulation as set forth in claim **13** wherein a plurality of wafers are contained within said capsule.

15. An encapsulation as set forth in claim **10** wherein said wafer consists essentially of said sintered isotope.

16. A method in accordance with the method of claim **9** wherein said stable material consists essentially of at least one isotope.

17. A method in accordance with the method of claim **3** wherein said wafer consists essentially of thulium oxide.

18. A method in accordance with the method of claim **1** wherein said wafer consists essentially of thulium oxide.

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RALPH G. NILSON, *Primary Examiner.*

A. B. CROFT, *Assistant Examiner.*

U.S. Cl. X.R.

250—84; 252—301.1