United States Patent

Selle et al.

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[54] RADIOISOTOPIC FUEL CAPSULE

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[73] Assignee: **The United States of America as represented by the United States Atomic Energy Commission**

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[51] **Int. Cl.** **G21h 5/00**

[58] **Field of Search** .....250/106 S, 106 R, 84; 23/344, 23/355; 252/301.1; 264/0.5

[56] **References Cited**

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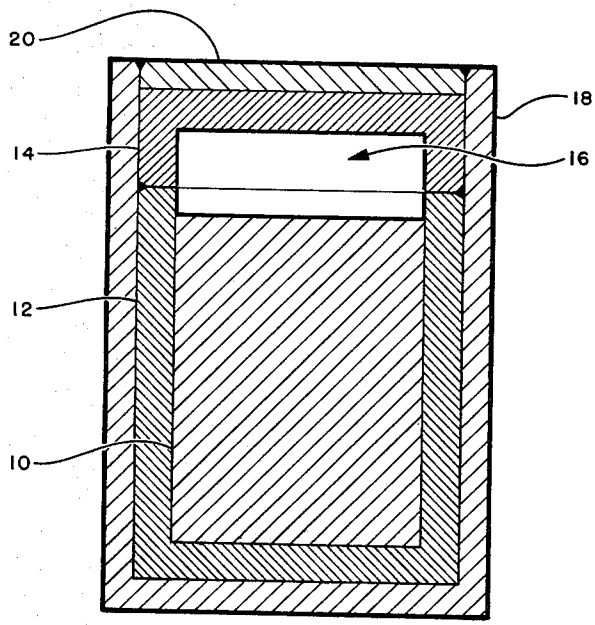
[57] **ABSTRACT**

A radioisotopic fuel capsule or cell and making thereof for use in such as a thermoelectric or thermionic generators which utilizes substoichiometric plutonium dioxide enclosed within a refractory container.

1 Claims, 1 Drawing Figure



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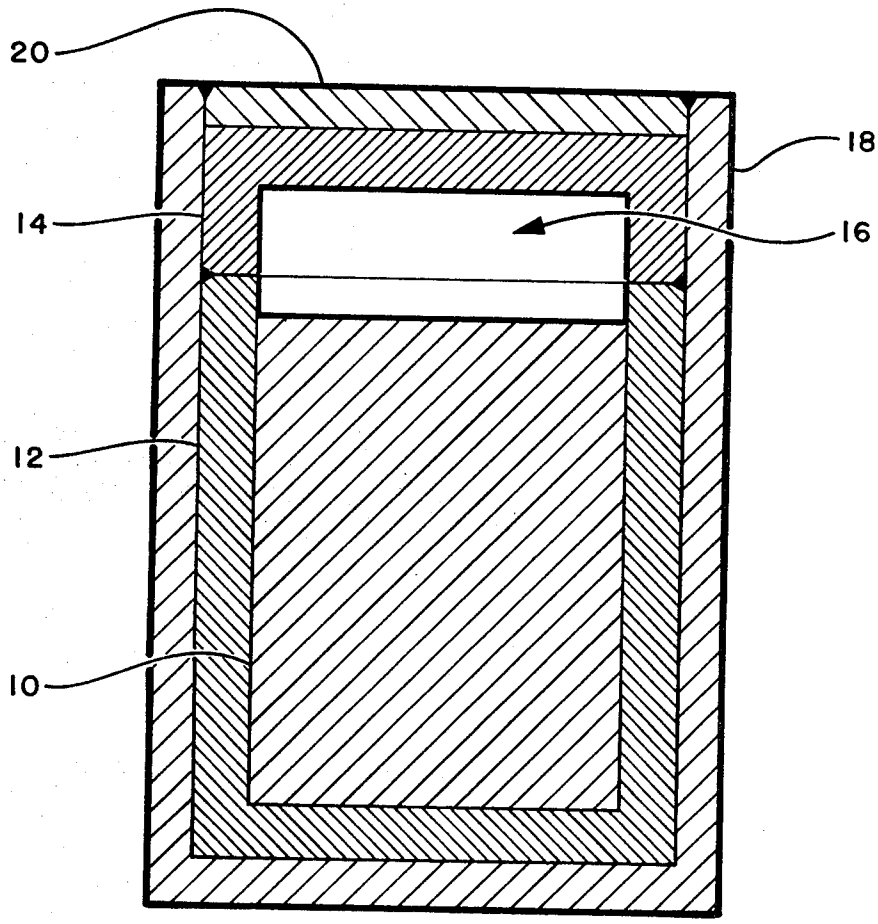

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RADIOISOTOPIC FUEL CAPSULE

BACKGROUND OF INVENTION

Plutonium dioxide, in the form of the alpha emitting radioactive plutonium–238, may be utilized in fuel elements or capsules as heat sources for thermoelectric, thermionic, and other power conversion units or simply as heat sources for space or the like applications. A quantity of plutonium dioxide may be enclosed within a sealed container or capsule and the capsule then used as a heat source for whatever application desired. In these applications, the radioactive fuel element or capsule may operate at elevated temperatures, such as at about 500° C. and higher. At these temperatures, some corrosion of the container or capsule material may occur and cause a weakening of the container structure or even failure thereof.

Utilization of radioisotopes as high temperature heat sources for electrical power generation or space propulsion or the like requires positive containment under all potential conditions. While plutonium–238 is usable at moderate temperatures in space and the like applications, it would be desirable to use the same at higher temperatures to achieve more efficient utilization of the thermal energy available with minimum or no corrosion or degradation of the container material.

SUMMARY OF INVENTION

It is an object of this invention to provide an improved radioisotopic fuel capsule using plutonium oxide fuel.

It is a further object of this invention to provide a plutonium oxide fuel exhibiting reduced corrosive attack of radioisotopic fuel capsule container materials.

Various other objects and advantages will appear from the following description of the invention, and the most novel features will be particularly pointed out hereinafter in connection with the appended claims.

It will be understood that various changes in the details, materials and arrangements of the parts, which have been herein described and illustrated in order to explain the nature of the invention, may be made by those skilled in the art.

This invention comprises a radioactive fuel capsule using substoichiometry plutonium dioxide fuel within a sealed container and a method of making same.

DESCRIPTION OF DRAWING

Aspects of the present invention are shown in the accompanying drawing of a sectional view representing a radioisotopic fuel capsule incorporating the fuel of this invention.

DETAILED DESCRIPTION

A plutonium oxide fuel **10**, described in more detail below, may be disposed within a suitable container or cell **12**, which may be of cylindrical or other configuration, and the container sealed by welding a lid or cover **14** thereon in an inert, oxygen free atmosphere. A void **16** may be left within container **12** wherein it may accumulate gaseous material or products emitted from the plutonium oxide fuel, or appropriate gas pressure relieving means (not shown) may be positioned in a wall or cover of the container to facilitate escape of gas from the container and minimize pressure buildup therein. An outer protective housing **18** or housings with sealed cover **20** may be disposed about container **12**, if desired.

Container **12** and cover **14** may be made of refractory material, such as certain refractory metals or alloys thereof. For the plutonium oxide fuel of this invention, such refractory metals may include tantalum, niobium, rhenium, tungsten, hafnium, vanadium, zirconium, nickel and titanium and alloys thereof or alloys with other refractory metals. Of these materials, tantalum and tantalum-10% tungsten may be preferred because of their particularly high corrosion resistance and ease of fabrication. Other refractory metals and alloys may react to a greater or lesser extent depending on the materials and environmental conditions.

It has been found, that a plutonium oxide fuel **10** having an oxide to plutonium ratio in the range of about 1.60 to about 1.98, with the optimum composition being from about 1.75 to 1.85, exhibits substantially lower corrosive and other forms of attack against the container material. Such plutonium oxide fuel may be referred to as substoichiometric plutonium dioxide and may be used in the form of particles, powder or microspheres with particle sizes ranging from about 25 to 400 microns. If desired, the substoichiometric plutonium dioxide particles may be pressed into a pellet and the pellet sintered into a solid mass.

The substoichiometric plutonium dioxide fuel may be prepared by reducing stoichiometric plutonium dioxide with hydrogen or with plutonium metal, or by other convenient means.

The substoichiometric plutonium dioxide may be prepared within a fuel capsule by mixing ordinary or stoichiometric plutonium dioxide with a material which is more reactive to the oxygen than the container material. In other words, the reactive metal oxide should have a free energy of formation more negative than the free energy of formation of the container material oxide so that the reactive metal reacts preferentially with any oxides within the container. Such reactive materials may include yttrium, tantalum, cerium, titanium, uranium, thorium, and niobium and some of the rare earths as well as plutonium. By heating the stoichiometric plutonium dioxide with these reactive materials to elevated temperatures the oxygen may be preferentially "gettered" by the reactive material. When this happens, the plutonium dioxide fuel becomes substoichiometric in composition. The desired oxygen to plutonium ratio, such as a preferred ratio of about 1.8, may be achieved by selecting an appropriate reactive material and amount thereof and reacting the same for some prescribed period of time and temperature. The reactive material may be added to the stoichiometric plutonium dioxide in any of several finely divided or dispersed forms, such as by chemical vapor deposition of the reactive material onto the surface of the oxide particles.

Stoichiometric plutonium dioxide particles were placed in contact with tungsten, tungsten-25% rhenium and rhenium at 2,000° C for 1,000 hours. Test results show that gross reaction occurred between the tungsten and stoichiometric plutonium dioxide with a 250 micrometers intergranular penetration and a certain amount of general solution attack. The stoichiometric plutonium dioxide exhibited three forms of attack with tungsten — 25 atom percent rhenium (intergranular penetration, general solution and oxide precipitation) with penetration to 200 micrometers. Rhenium showed not only about 50 micrometers attack in contact with the stoichiometric plutonium dioxide but also a massive amount of vapor-phase transport from the fuel material to the capsule wall. Tantalum-10% tungsten alloy exhibited intergranular penetration to greater than 530 micrometers when contacted with stoichiometric plutonium dioxide at 1,200° C. for about 1,400 hours.

Substoichiometric plutonium dioxide particles having an oxygen to plutonium ratio of about 1.8 were placed in contact with the same materials under the same conditions. The tungsten exhibited about 25 micrometer general solution attack and less than 50 micrometer intergranular penetration while the tungsten-25% rhenium and the rhenium exhibited a total attack of less than about 10 and 25 micrometers, respectively. Further, no transport was observed in the rhenium capsule containing $PuO_{1.8}$. Substoichiometric plutonium dioxide samples exhibited 0 general solution attack and 0 and 12.5 micrometer intergranular penetration of tantalum-10% tungsten at 1,200° C. after about 1,400 hours. These latter tests included a 4.4 percent yttrium and 5.4 percent tantalum getter addition to achieve the substoichiometry.

What is claimed is:

1. A radioisotopic fuel capsule comprising a sealed container made of a material selected from the group consisting of tantalum, niobium, rhenium, tungsten, hafnium, vanadium, zirconium, nickel, titanium and alloys thereof, and in said con-

tainer oxygen-deficient fuel comprising substoichiometric plutonium dioxide particles with ratio of oxygen to plutonium in the range of about 1.7 to 1.85 and an oxygen-gettering material interspersed with said particles having a free energy of oxide formation more negative than said container material and selected from the group consisting of yttrium, tantalum, cerium, titanium, uranium, thorium, niobium, plutonium and the rare earths.

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