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[54] **α-RAY HEAT SOURCE, SUITABLE FOR USE IN A CARDIAC PACEMAKER**
8 Claims, 1 Drawing Fig.

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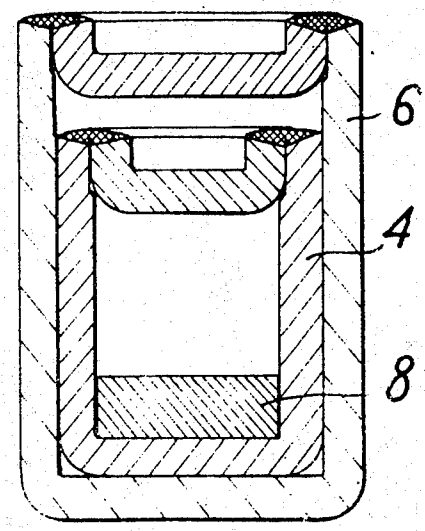
ABSTRACT: The heat source comprises a leak-tight container and a body of α-ray emitting material in the container. The material is an alloy of Pu 238 and of Sc, Ce, In, Ga or Am in such proportions that the alloy is in δ phase. The container consists of an inner sheath of tantalum or tungsten and of an outer sheath of platinum.



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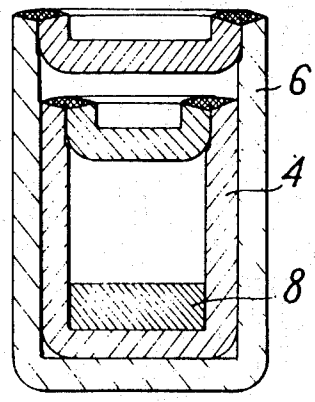

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α-RAY HEAT SOURCE, SUITABLE FOR USE IN A CARDIAC PACEMAKER

The invention is directed to a radioactive heat source which is primarily intended to supply energy to a cardiac pacemaker.

It has already been proposed to make use of radioactive α-ray sources for supplying the energy which is necessary for the operation of a cardiac Pacemaker. Among the α-emitters proposed for this purpose can be mentioned the plutonium isotope 238 which does not call for the presence of substantial shielding material since there is no emission of penetrating radiation. The plutonium metal is enclosed in an inner sheath of tantalum-base alloy which is in turn enclosed in an outer sheath of titanium.

In principle, radioactive sources have advantages over electric batteries in that their power falls off at a very low rate and that they can be constructed in very small sizes. On the other hand, the use of an α-emitter makes it necessary to satisfy a certain number of criteria and it is essential to ensure that the α-emitter (plutonium-238) is free of contaminants such as light elements and the isotope 236 which would produce penetrating radiations.

Unfortunately plutonium in the unalloyed state has a number of unfavorable properties which have virtually precluded its use for reasons of safety or ease of fabrication: the crystal structure of plutonium changes a number of times between 20° C. and the melting temperature with a correlative variation of volume and of expansion coefficient. Plutonium oxides very readily in air in the presence of moisture and gives rise to a powdery oxide which is liable to form aerosols having a very high degree of toxicity since the total amount which is permissible in the human body is less than 0.6 μg.

The aim of the invention is to provide a radioactive source which contains plutonium-238 and which is not attended by the disadvantages referred-to above or which is subject to such disadvantages only to a very limited extent. To this end, the invention proposes a radioactive source in which the α-ray emitting material consists of an alloy of plutonium-238 which contains less than 1 p.p.m. of Pu^{236}, said alloy being such as to contain at least one addition element from the group consisting of indium, scandium, gallium, cerium and americium in proportions such that the alloy remains in delta phase and retains a face-centered-cubic crystal structure between normal atmospheric temperature and a temperature of at least 450° C.

It is necessary to ensure that the proportion of Pu^{236} does not exceed the value of 1 p.p.m. mentioned above in order that the gamma radiation produced by its daughter elements and received by human tissues when the source is employed in a cardiac Pacemaker of customary dimensions should not exceed the permissible threshold. In regard to the proportion of light elements, the atomic number of which is smaller than 14, this proportion must also remain sufficiently small to ensure that the neutron flux resulting from the (α, n) reaction remains lower than the permissible threshold.

The first four additives mentioned above serve to stabilize the alloy in delta phase even in respect of low proportions which do not reduce the power density of the alloy to any marked extent with respect to that of pure plutonium-238. Since americium is also an alpha emitter, it is evident that the use of a high proportion of this additive does not carry any disadvantage. Moreover, none of the five additives undergoes the (α, n) reaction and this holds true in the case of scandium which could give some cause for apprehension by reason of its low atomic number.

A better understanding of the invention will be gained from the following description of a radioactive source which constitutes one nonlimitative embodiment of the invention and of a method for the fabrication of said source. Reference is made in the description to the single FIGURE of the accompanying drawings in which the source under consideration is shown in cross section along a vertical midplane.

The source which is illustrated in the FIGURE is designed to withstand a temperature of 1000° C. for a period of 30 minutes without exhibiting crack formation; this corresponds to the case of an accident followed by a fire. In view of the fact that it is impossible to provide a single sheath which, at this temperature, would afford resistance both to the plutonium in liquid phase (the melting temperature of plutonium-base alloys being 640° to 700° C.) as well as to atmospheric oxidation, provision is made for a container which is made up of two sheaths fitted one inside the other. The inner sheath 4 is formed of tantalum, of tungsten or of tungsten-base alloy containing a low percentage of copper whilst the outer sheath 6 is formed either of platinum, rhodium-platinum or iridium-platinum.

It is essential to provide within the interior of the inner sheath 4 between the end-cap and the pellet 8 of α-emitting material an expansion chamber which is intended to collect the helium given off by the plutonium: this requirement is readily apparent when it is recalled that one half-gram of plutonium-238 gives off in 10 years (which is the minimum life of the source) 3.6 $cm.^3$ of helium at normal temperature and pressure; once the helium has accumulated, the sheaths must also be capable of withstanding a temperature of 1000° C. without bursting.

The α-emitting radioactive material consists of an alloy of plutonium-238 which is free of additives or contaminants liable to give rise to the emission of penetrating radiation, especially the majority of penetrating radiation, especially the majority of light elements and the isotope-236 of plutonium.

This material must meet a certain number of conditions:

- its resistance to atmospheric oxidation must be greater than that of unalloyed plutonium;
- it must be free from allotropic transformation between normal atmospheric temperature and a temperature which is distinctly higher, which must in any case be above 100° C. and which is preferably at least 450° C.,
- the melting point must be sufficiently high: it may be postulated that the melting point must be substantially higher than 450° C;
- the material must be capable of undergoing a shaping operation by means of a mechanical cold-working process, especially rolling (which excludes, for example, the Pu–Zr alloys).

In practice, the foregoing conditions call for an alloy in delta phase having a face-centered-cubic crystal structure.

Furthermore the alloy should preferably have good thermal conductivity, must be compatible with the material constituting the inner sheath and must have a coefficient of thermal expansion which does not differ too greatly from that of the inner sheath.

The conditions outlined above are satisfied by binary alloys comprising plutonium and at least one of the elements: In, Sc, Ga, Ce and Am when the proportion of addition element is comprised within the following ranges:

- scandium: 7 to 14 atom percent, namely 1.4 to 2.28 percent by weight (minimum melting point: about 700° C.)
- indium: 1 to 4 atom percent, namely 0.48 to 1.97 percent by weight (minimum melting point of an alloy which corresponds to this composition: about 800° C.)
- gallium: 4 to 10 atom percent, namely 1.20 to 3.16 percent by weight (minimum melting point: about 660° c.)
- cerium: 3.5 to 17 atom percent, namely 2.09 to 10.7 percent by weight (minimum melting point: about 620° C.)
- americium: 7 to 30 atom percent, namely 7.05 to 30.2 percent by weight (minimum melting point: higher than 550° C.).

It is apparent that these addition elements must also be free of any impurities which would be liable to give rise to (α, n) reactions.

Among the alloys which have been mentioned above, the most advantageous appears to be the Pu–Sc alloys which, in respect of the different proportions of Sc, have the following properties:

Proportion of Sc (atom %)	8	10	14
(weight %)	1.61	2.05	2.98
Density at 25° C. (g./cm.³)	14.78	14.56	13.90
Mean expansion coefficient between 20 and 400° C.	4.4×10^{-6}	6.0×10^{-6}	8.3×10^{-6}
Electrical resistivity between 20° C. and 160° C. (microhm cm.²/cm.	120±15	125±15	120±15

It is apparent that the coefficient of expansion varies rapidly with the proportion of Sc, thereby permitting the possibility of adjusting the coefficient to a value which is compatible with the coefficient of the sheaths simply by making a suitable choice of percentage content.

Resistance of the alloy to atmospheric oxidation is considerably improved with respect to that of Pu in the pure state. Thus, whereas a reference sample of Pu is highly oxidized and collapses after being maintained at 100° C. for a period of 15 hours, then at 150° C. in air for a period of 1 hour, a sample of identical shape and containing 10 atom percent of Sc withstood the following treatment prior to incipient oxidation: maintaining at 100° C. for 15 hours, then at 150° C. for 170 hours, then at 200° C. for 48 hours, then at 300° C. for 23 hours and at 400° C. for 2 hours.

The Pu-Sc alloy can be very readily cold-rolled: at 25° C., an alloy containing 10 atoms per cent of Sc undergoes a thickness reduction of 500 percent prior to the appearance of cracks.

The Pu–Ce alloy also appears as having properties which make its use an advantage although its corrosion resistance is lower than that of the PU–Sc alloy. In respect of different proportions of Ce, the alloy exhibits the following properties:

Proportion of Ce (atom %)	10	16
(weight %)	6.1	10.04
Density at 25° C. (g./cm.³)	14.89	14.33
Mean expansion coefficient between 20° C. and 400° C. (per ° C.)	2×10^{-6}	9×10^{-6}
Electrical resistivity between 20° C. and 300° C. (microhm cm².cm.)	110±15	

Corrosion resistance is satisfactory since a sample having the same shape as in the previous instance withstands 100° C. for a period of 15 hours, then 150° C. for 146 hours, 200° C. for 48 hours and 300°

for 15 hours before it begins to oxidize in air. The rolling property of an alloy containing 10 atom percent Ce is equivalent to that of the alloy which contains 10 atom percent Sc.

The Pu-In alloy has a degree of resistance to corrosion in air at 25° C. which is considerably higher than that of unalloyed plutonium but is nevertheless subject to one defect: although the alloy is in principle in δ phase in the case of an atomic percentage of In within the range of 1 percent to 4 percent, a small quantity of another phase is observed in the as-cast alloy. Homogenization of the alloy is difficult and researches on the possibility of removal of this second phase by heat treatment at 500° C. have not yet been fully completed.

In conclusion, the alloys which are contemplated can be classified as follows, in order of diminishing interest:

Pu–Sc
Pu–Am
Pu–Ce
Pu–Ga
Pu–In

It should be noted that the location of Pu–Am in the list is due, not to metallurgical properties of special interest, but to the fact that the two constituents are α-emitters.

A number of different methods are open to selection for the fabrication of the source which is illustrated in the FIGURES and in particular the method which will now be described by way of example. Pu^{238} is obtained by in-pile irradiation of neptunium-237 followed by chemical separation. The alloy of plutonium-238 with the addition element is then formed in a furnace (an arc furnace, for example) in a controlled atmosphere and the product obtained is coldrolled in a moisture-free inert-gas atmosphere in order to obtain a plate. Alloy pellets are then cut from the plate by means of a press and surface decontamination of the pallets is carried out by removal of radioactive dust particles. Each pellet is placed within its first sheath in which it will be applied against the endwall either by means of a thermal diffusion-bonding process carried out under a vacuum, or by means of a tube formed of the same material as the inner sheath or by means of a light spring interposed between the pellet and the end-cap. The end-cap is fitted in position, then welded by the electron beam process. The assembly is then placed within the second sheath and the end-cap is also beam welded. The completed source is finally decontaminated and subjected to a leak-tightness test.

By way of example, a source has been designed for use in conjunction with a cardiac Pacemaker having an electric power consumption of 200 μ.; the dimensions of the shell which contains all the components (source, thermoelectric converter and electronic equipment) must not exceed 4×2×5 cms. and the weight must not exceed 100 g. The life expectancy is 10 years. The source is 13 mm. in height, 9 mm. in diameter and contains 33 mm.³ of plutonium alloy enclosed within a chamber of approximately 110 mm.³; this ratio of ½ between the volume of alloy and the volume intended for expansion may be regarded as suitable in the majority of cases.

It is self-evident that the invention is not limited to the application which has been illustrated and described by way of example.

We claim:

1. A heat source for a cardiac Pacemaker comprising a leak-tight container, a body of α-ray emitting material in said container, an inner sheath of tantalum, tungsten or tungsten-based alloy containing copper for said container, a platinum base outer sheath for said container, said material being an alloy of plutonium 238 containing less than 1 p.p.m. of Pu 236 and at least one of the elements of the group consisting of Sc, Ce, In, Ga and Am and being in δ phase and exhibiting a face-centered cubic crystal structure from ambient temperature to above 450° C.

2. A heat source according to claim 1, wherein said material is a binary Pu–Sc alloy having a Sc atomic content between 7 and 14 percent.

3. A heat source according to claim 1, wherein said material is a binary Pu–Ce alloy having a Ce atom content between 3.5 and 17 percent.

4. A heat source according to claim 1, wherein said material is a binary Pu–Ga alloy having a Ga atomic content between 4 and 10 percent.

5. A heat source according to claim 1, wherein said material is a binary Pu–In alloy having an In atomic content between 1 and 4 percent.

6. A heat source according to claim 1, wherein said material is a binary Pu–Am alloy having an Am atomic content between 7 and 70 percent.

Pu A radioactive heat source according to claim 1, including means for retaining said body of material in contact with said sheath selected from the group consisting of a spring and tube of the material constituting said inner sheath.

8. A radioactive heat source comprising a container having a leak tight chamber and a body of α-ray emitting material occupying part of said chamber, said material being a binary alloy of Pu 238 having a Pu 236 content lower than 1 p.p.m. and of one of the elements from the group consisting of Sc, Ce, Ga, In and Am, wherein the atom proportions of said element in the alloy is:

between 7 and 13 percent for Sc
between 3.5 and 17 percent for Ce
between 4 and 10 for Ga
between 1 and 4 percent for In
between 7 and 70 percent for Am.