

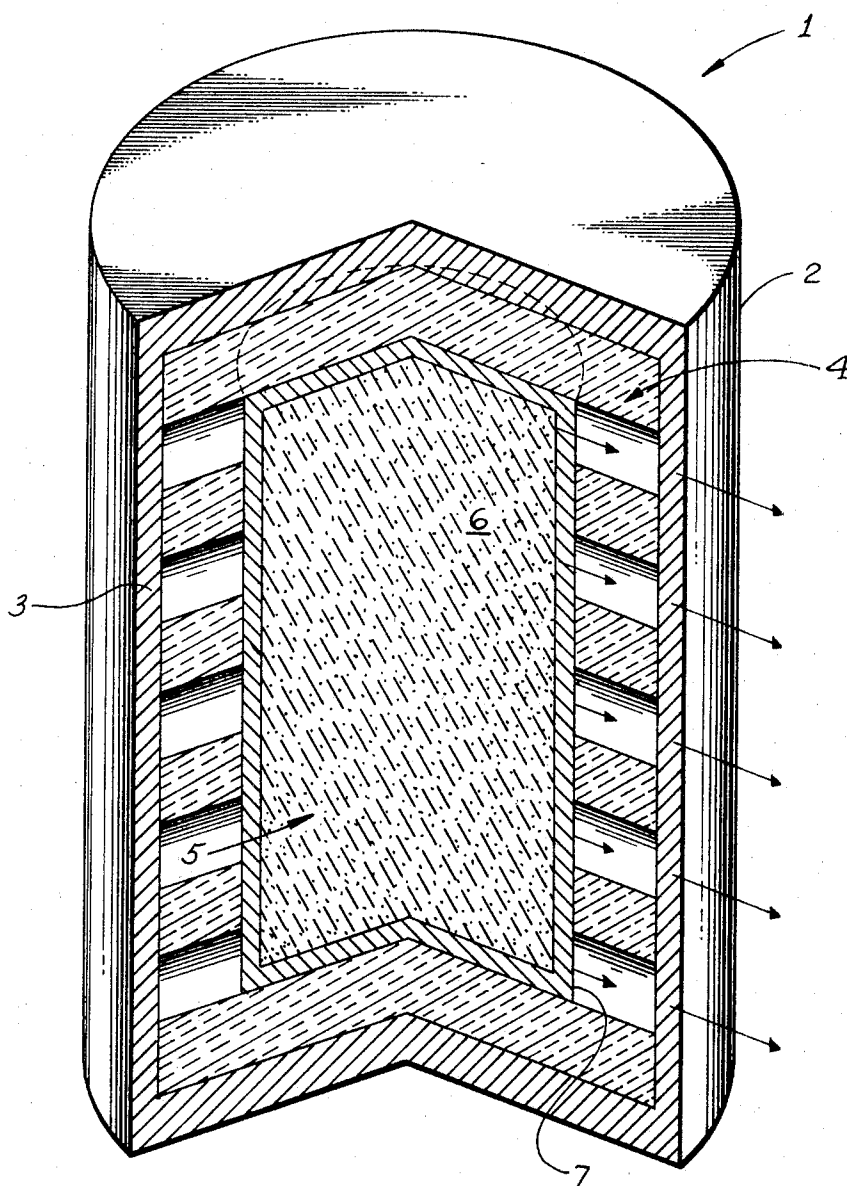
June 2, 1970

V. KESHISHIAN

3,515,875

ALPHA-PARTICLE-EMITTING RADIOISOTOPE GENERATOR

Filed June 17, 1965



INVENTOR
VAHE KESHISHIAN

By *Harry Kolin*
ATTORNEY

1

3,515,875

ALPHA-PARTICLE-EMITTING RADIOISOTOPE GENERATOR

Vahe Keshishian, Sherman Oaks, Calif., assignor to North American Rockwell Corporation, a corporation of Delaware

Filed June 17, 1965, Ser. No. 464,702

Int. Cl. G21h 5/00

U.S. Cl. 250—106

5 Claims

This invention relates to a radioisotope generator. More particularly it relates to a radioisotope generator wherein neutron shielding requirements are substantially reduced compared with similar generators.

Radioisotope powered generators are known. Such units are of particular interest for space missions for supplying the power needed by the instruments of the space vehicle. These generators are also of utility in situations where there is need for a remote, unattended, long-lived small power source that is relatively impervious to conditions and hazards of its environment. Such uses include earth-based ones such as navigational aids in remote areas, communication relay stations, forest warning equipment, ocean cable boosters, and the like.

For space missions, particularly manned ones, shielding requirements against radiation contribute significantly to the overall weight of the space vehicle. Astronauts present in the vehicle would require shielding not only from external radiation but also from the radiation emitted by the isotopic power unit itself.

In general, isotopes which are alpha-particle emitters are preferred for fuel use in manned mission space flights because they are relatively easy to shield against, alpha radiation being the least penetrating of all. Exemplary of such a suitable isotopic fuel is plutonium-238. However, the alpha-particle-emitting isotopes are not ordinarily usable as fuel in elemental forms, but are present in the form of their compounds, alloys and mixtures so as to provide an isotopic fuel with suitable properties with respect to melting point, hardness, ease of fabrication and handling, and other related physical and metallurgical characteristics.

While alpha-particle emission per se requires but minimal shielding, secondary radiation resulting from interaction of the primary alpha particle with material in the immediate vicinity of the isotope emitter accounts for a significant increase in shielding requirements. The most important secondary source of radiation requiring shielding arises from the alpha-neutron reaction in which an element is transmuted by absorption of an alpha particle, a neutron leaving the excited nucleus.

Accordingly, it is an object of this invention to provide a radioisotope generator wherein neutrons shielding requirements are substantially reduced.

It is another object to provide a primary alpha-particle-emitting radioactive source wherein secondary neutron generation as a result of an alpha-neutron reaction is substantially reduced.

It is still another object to provide a method for preparing an alpha-particle emitter for a radioisotope generator whereby there results minimal secondary neutron generation.

In accordance with this invention a radioisotope gen-

2

erator is provided which includes a fuel capsule and a radiation shield in cooperative relation therewith wherein the radioisotope fuel material is an alpha-particle-emitting radioactive isotope combined with components which essentially have a threshold for the alpha-neutron reaction greater than the maximum energy of the emitted alpha particles. Thereby secondary neutron generation resulting from an alpha-neutron reaction is substantially reduced, with a corresponding reduction in neutron-shielding requirements. Preferred as fuel materials for use in the isotope fuel generator of this invention are alpha-emitting radioisotopes of the actinide series combined in molecular form with particular low atomic number electronegative elements, or with selected isotopes of these elements, which have a threshold energy for the alpha-neutron reaction greater than the maximum energy of the alpha particles emitted by the radioisotope. Particularly preferred as fuel material is radioactive plutonium oxide, wherein the plutonium consists essentially of the plutonium-238 isotope and the oxygen consists essentially of the oxygen-16 isotope substantially free of, or with only trace amounts of, the oxygen-17 and oxygen-18 isotopes. Ordinarily, plutonium-238 oxide will emit a primary neutron by spontaneous fission and about 15 times as many secondary neutrons by an alpha-neutron reaction on natural oxygen. Thus complete elimination of secondary neutrons will reduce overall neutron emission by a factor of 16.

For a more complete understanding of the invention, reference is made to the sole figure of the drawing showing a perspective view, partly in section, of an embodiment of a radioisotope generator suitable for use in practice of this invention.

Referring to the drawing, which is intended as illustrative and not restrictive of the present invention, a simplified view of a radioisotope generator 1 is shown from which its principal components may be seen. An outer shell 2, usually in the form of a thin cylindrical can of metal, protects the internal components from contamination and may serve as a heat radiator where required. A radiation shield 3 is required to provide safety requirements in handling the generator during launch and following impact and also to protect astronauts present in the space vehicle during manned space flights. These shields are of high density and prevent the emission of primary and secondary radiation. Exemplary of such shields are lead, depleted uranium, and cast iron. Low density neutron shields such as lithium hydride are also required. These shields all contribute considerably to the weight of the radioisotope generator. This increase in shielding weight is a major disadvantage for space missions. Where the minimizing of shielding weight is a primary consideration, it is preferred to use radioisotopes which are alpha-radiation emitters, since alpha radiation is the least penetrating of all and requires minimal shielding provided no secondary radiation of a penetrating nature occurs.

An energy converter section 4 is used to transform part of the isotope decay heat into electricity. This may consist of an array of thermoelectric elements or thermionic converters. At the heart of the generator is the energy source, shown as a fuel capsule 5 in which a radioisotopic fuel material 6 is enclosed by a capsule wall 7.

The radioisotope generator may be of any desired shape, cylindrical shapes or spherical shapes being more common. In one type of assembly, the energy converters are placed around a space reserved for the fuel capsule. The shield is then wrapped around the converters. The outer shell, except for an end left open for fuel insertion, is soldered or welded around the shield. The fuel capsule is then usually inserted by remote control for safety reasons, and the last piece of the outer shell is then sealed in place. Almost all of the nuclear particles emitted by the decaying radioisotopic fuel are absorbed inside the fuel capsule. During the absorption process, the fast nuclear particles collide with the atoms in the fuel capsule, causing them to move more violently and thus raise the capsule temperature. The kinetic energy of the particles is thereby converted to heat. Generally about 5 to 10 percent of the total heat flow, shown by directional arrows, is converted into electricity. The remaining heat energy produced by the fuel flows into the outer shell from where it is radiated or conducted to a surrounding environment.

Because of the physical and metallurgical requirements for the fuel material, an alpha-particle-emitting isotope cannot ordinarily be used in the pure elemental form. For example, Pu^{238} has a melting point ranging from 120°C . to 640°C ., depending upon the particular crystalline structure. Whereas, in the form of PuC its melting point is 1654°C .; PuO_2 melts at 2282°C ., PuN 2450°C ., PuB 2040°C . Thus the isotopic materials will ordinarily be used in the form of their molecular compounds, alloys, or nonstoichiometric mixtures.

It is an essential feature of this invention that secondary neutron generation resulting from an alpha-neutron reaction is substantially reduced by selecting the component that is combined with the alpha-particle-emitting radioactive isotope to have a threshold for the alpha-neutron reaction that is greater than the maximum energy of the emitted alpha particles. Generally a threshold above about 5.6 mev. is required, this threshold value varying somewhat depending upon the particular alpha-particle-emitting source.

The selection of the particular alpha-particle-emitting radioactive isotope and its combining component will be determined by many factors. As mentioned, metallurgical and physical properties play a primary role. Also of importance is compatibility of the fuel material with cladding materials, as well as its chemical stability, availability and cost. Where a space mission of brief duration is contemplated, an isotope such as polonium-210 which has a half-life of 138 days may be suitable; whereas for a Mars emission, which would require approximately 2 years, a longer-lived isotope, such as plutonium-228, which has a half-life of 90 years, would be required. Also the choice of isotope would be governed in part by the relative freedom of the alpha-particle-emitting isotope from other primary radiation emission such as gamma rays and neutrons.

The alpha-particle-emitting radioactive isotopes of the actinide series are preferred in the practice of this invention. Exemplary of suitable alpha-particle-emitting isotopes of this class are: Pu^{238} , Po^{210} , Th^{228} , U^{232} , Cm^{242} , Cm^{244} , and Am^{241} . Pu^{238} is advantageous for use as an alpha-particle-emitting radioactive source because of its long half-life and the relatively small amounts of gamma rays and neutrons which are also primarily emitted. Thus shielding requirements are considerably minimized. As examples of electronegative components with which the electropositive alpha-particle-emitting isotopes may be combined, either as molecular compounds, alloys, or nonstoichiometric mixtures are the oxides, carbides, nitrides, silicides, and oxychlorides.

In Table I is shown the threshold energy for the alpha-neutron reaction for the relatively light weight natural isotopes. The relative abundance of these natural isotopes is also shown.

TABLE I

[Threshold energies for alpha-neutron reactions in natural isotopes from Li to Ni]

	Isotope	Percent abundance	Threshold energy for (α , n) reaction (mev.)
5	Li^6	7.4	4.85
	Li^7	92.6	5.25
	Be^9	100	1.0
	B^{10}	18.8	1.0
	B^{11}	81.2	0.22
10	C^{12}	98.9	11.25
	C^{13}	1.1	1.0
	N^{14}	99.62	6.1
	N^{15}	0.38	8.15
	O^{16}	99.76	15.2
	O^{17}	0.04	1.0
	O^{18}	0.20	0.86
15	F^{19}	100	2.35
	Ne^{20}	90.52	6.7
	Ne^{21}	0.27	1.0
	Ne^{22}	9.21	8.52
	Na^{23}	100	3.47
	Mg^{24}	78.6	8.37
	Mg^{25}	10.1	1.0
	Mg^{26}	11.3	1.0
20	Al^{27}	100	3.05
	Si^{28}	92.3	8.25
	Si^{29}	4.7	1.72
	Si^{30}	3.0	3.95
	P^{31}	100	6.58
	S^{32}	95.1	9.77
	S^{33}	0.74	0.75
	S^{34}	4.2	5.06
25	S^{36}	0.0136	3.40
	Cl^{35}	75.4	6.52
	Cl^{37}	24.6	4.28
	Ar^{40}	99.632	4.28
	K^{39}	93.1	7.77
	K^{41}	6.9	3.72
30	Ca^{40}	96.97	No data
	Ca^{42}	0.64	6.58
	Ca^{43}	0.145	1.0
	Ca^{44}	0.0033	0.25
	Ca^{45}	0.185	0.15
	Sc^{45}	100	2.45
	Ti^{46}	7.94	4.75
	Ti^{47}	7.75	0.38
	Ti^{48}	73.45	2.92
35	Ti^{49}	5.52	1.0
	Ti^{50}	5.34	1.92
	V^{50}	0.23	0.20
	V^{51}	100	2.45
	Cr^{50}	4.49	5.30
	Cr^{52}	83.78	3.85
	Mn^{55}	100	3.80
40	Fe^{54}	5.81	5.95
	Fe^{56}	91.64	5.43
	Fe^{57}	2.21	1.4
	Fe^{58}	0.34	3.84
	Co^{59}	100	5.42
	Ni^{58}	67.7	10.36
	Ni^{60}	26.2	8.1
	Ni^{61}	1.2	4.17
45	Ni^{62}	3.7	6.8
	Ni^{64}	1.2	4.9

¹ Q pos.

In accordance with the teaching of the invention, if the alpha-emitting isotope is used, for example, in the form of its carbide, the C^{13} isotope would be eliminated by chemical or physical treatment so that the carbide would be substantially free of the C^{13} isotope and could consist almost exclusively of the C^{12} isotope. Thereby, since the threshold energy of the C^{12} isotope is 11.25 mev. (Table I), no alpha-neutron reaction would occur and secondary neutron emission would be substantially reduced or eliminated. Similarly, if plutonium oxide (PuO_2) were used, the radioactive fuel would consist essentially of $\text{Pu}^{238}\text{D}_2^{16}$ since the threshold energy of O^{16} required for the alpha-neutron reaction is 15.2 mev.; whereas the threshold energy of the O^{17} and O^{18} isotopes is considerably less. This radioisotope fuel, $\text{Pu}^{238}\text{O}_2^{16}$, is particularly preferred therefore in the practice of this invention because of its elimination of secondary neutron generation as well as its desirable chemical and physical properties.

Maximum benefits in reducing shielding requirements are obtained where there is present as combining component only the stable isotope of the electronegative element which has a threshold for the alpha-neutron reaction greater than the maximum energy of the alpha particles emitted by the radioactive isotope. However, substantial improvement in shielding requirements is obtained even where lesser or trace amounts are present of

5

the undesired isotopes which have low threshold values for the alpha-neutron reaction.

For purposes of illustration, without in any sense being limited thereby, a specific method of practicing this invention, and the advantages obtaining thereby, will be illustrated with reference to the radioactive isotopic compounds $\text{Pu}^{238}\text{O}_2^{16}$ and $\text{Po}^{210}\text{O}_2^{16}$.

EXAMPLE 1

Preparation of enriched oxygen-16

Enriched oxygen-16 is conveniently prepared by electrolysis of heavy water, D_2O , which is enriched in the heavier oxygen isotopes. This heavy water process is followed by hydrogen sulfide exchange and then followed by distillation to yield a product containing 90% D_2O . The 90% D_2O is then electrolyzed to produce 99.75% D_2O . During electrolysis, the lighter O^{16} isotope comes off initially as a gas, the heavier oxygen isotopes concentrating during this production of D_2O by electrolysis. Using such a process H_2O is obtainable with a depleted content of 0.007% O^{17} and 0.007% O^{18} , with the balance O^{16} . Oxygen-16 is then conveniently obtained in gaseous form by electrolysis of this O^{16} -enriched water.

EXAMPLE 2

Preparation of $\text{Pu}^{238}\text{O}_2^{16}$

(a) By reaction with plutonium metal.—Plutonium metal is produced on a continuous basis by electrolysis. The plutonium metal is then heated in O^{16} gas at a temperature of 400°C . to form PuO_2^{16} .

(b) From plutonium oxide by hydrofluorination.— PuO_2 containing natural oxygen and prepared by low firing at a temperature below 480°C . is converted to PuF_4 by hydrofluorination at 450°C . The PuF_4 is reduced to Pu metal by reaction with calcium and iodine. The Pu metal is transferred to a closed system and converted to PuO_2^{16} as above described.

(c) From plutonium oxide by treatment with phosgene.—Plutonium oxide containing natural oxygen is heated at 400°C . with phosgene to convert it to plutonium trichloride. The chloride is then reduced with calcium and iodine to form plutonium metal which is then converted to PuO_2^{16} by reaction with O^{16} gas as above described.

(d) From Plutonium Trichloride.—The plutonium trichloride is hydrolyzed in the vapor phase with O^{16} -enriched water to form PuO_2^{16} . The reaction that occurs is as follows:



EXAMPLE 3

Decreased neutron yield from Alpha-neutron reaction

Two samples of radioactive polonium-210 are compared. The samples are obtained as PoCl_4 deposited on glass. One sample is dissolved in distilled water containing oxygen of natural abundance (99.759% O^{16} , 0.037% O^{17} , 0.204% O^{18}) and the other in water enriched in O^{16} (0.007% O^{17} , 0.007% O^{18} , balance O^{16}). A small amount of HCl is added to both solutions to prevent depositions of Po on the walls of the container. The two solutions are then made to the same volume in the same size containers so that the ratio of the count rates observed with a neutron in a fixed geometry is the desired ratio of the neutron production rates. The neutrons from the source solution are thermalized with paraffin so as to permit use of a boron trifluoride neutron detector. This detector is efficient to about 10% for neutrons entering the sensitive volume. The cross-sectional area for the detector is selected to be about 20 cm^2 . For a desired count rate of 1,000 counts/min. from the solution of normal water, and a 3-cm. thickness of paraffin, about 10^5 neutrons/min. is required of the radioisotope source. One curie of

6

Po^{210} will produce 1.33×10^5 n/min. Using a curie of Po^{210} for each of the samples, approximately 1,000 counts/min. is obtained for the solution of normal water and approximately 1,000/30 or about 30 counts/min. for the O^{16} -enriched water (i.e., O^{18} depleted water by a factor of 30). Polonium-210 emits alpha particles with an energy of 5.3 mev., compared with 5.5 mev. for Pu^{238} .

According to the data of Serdiukova et al., "Investigation of the (α ,n) Reaction on Oxygen" Bull. Acad. Sci. U.S.S.R. (Phys. ser.) vol. 21, p. 1018 (1957), the neutron source from an alpha-neutron reaction on oxygen is proportional principally to the O^{18} concentration. In the present process, by depleting both the O^{17} and O^{18} isotopes, a significant improvement in reducing the emission of secondary neutrons is obtained by using an O^{16} -enriched radioactive alpha-emitting oxide.

Where pure $\text{Pu}^{238}\text{O}_2^{16}$ is used, with the O^{17} and O^{18} isotopes of oxygen completely eliminated, the overall neutron source can be reduced by a factor of 16. But even with available O^{16} -enriched water, where the water is depleted to a content of 0.007% O^{17} and 0.007% O^{18} , there is a reduction of total neutron emission by a factor of about 10, or a reduction in the secondary alpha-neutron source by a factor of about 27. Thus in one design where an 11-inch thick lithium hydride neutron shield for a Pu^{238} oxide isotope source is required using natural oxygen, a reduction in shield thickness to 5 inches is obtained by using the above available O^{16} -enriched source. Thus for a space vehicle using as isotope source $\text{Pu}^{238}\text{O}_2$ containing normal oxygen, the 11-inch thick lithium hydride shield would weigh about 1000 pounds. With $\text{Pu}^{238}\text{O}_2$ containing the enriched O^{16} , 5-inch thick shield weighs about 400 pounds, resulting in a saving in weight of 600 pounds. This reduction in weight is of course highly significant in a space mission.

While the principles of the invention have been described above in connection with specific materials and processes, it is to be clearly understood that this description is made only by way of example and not as a limitation to the scope of the invention as set forth in the objects thereof and in the accompanying claims.

I claim:

1. A radioisotope generator including a fuel capsule and a radiation shield in cooperative relation therewith wherein the neutron shielding requirement for said shield is substantially reduced by preselecting as radioisotope fuel material in said capsule an alpha-particle-emitting radioactive isotope combined with components which essentially have a threshold for the alpha-neutron reaction greater than the maximum energy of the alpha particles emitted by said radioactive isotope whereby secondary neutron generation by an alpha-neutron reaction is substantially reduced.

2. A radioisotope generator according to claim 1 wherein said preselected fuel material is at least one compound selected from the class consisting of the oxides, carbides, nitrides, silicides, and oxychlorides of the radioactive actinide metals.

3. A radioisotope generator according to claim 2 wherein said radioisotope fuel material consists essentially of $\text{Pu}^{238}\text{O}_2^{16}$.

4. A radioisotope generator including a fuel capsule containing a primary alpha-particle-emitting radioactive fuel material having a substantially reduced secondary neutron emission wherein the radioisotope fuel material in said capsule comprises an alpha-particle-emitting radioactive isotope combined with components which essentially have a threshold for the alpha-neutron reaction greater than the maximum energy of the alpha particles emitted by said radioactive isotope whereby secondary neutron generation by an alpha-neutron reaction is substantially reduced.

5. A radioisotope generator according to claim 4

wherein said primary alpha-particle-emitting radioactive fuel material consists essentially of $\text{Pu}^{238}\text{O}_2^{16}$.

References Cited**UNITED STATES PATENTS**

3,145,181 8/1964 Courtois et al.
3,230,374 1/1966 Jones et al.

R. G. NILSON, Primary Examiner
M. J. FROME, Assistant Examiner

U.S. Cl. X.R.

⁵ 23—344, 345, 347, 349, 354, 355; 74—122.5, 122.7;
252—301.1.

PO-1050
(5/69)

UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,515,875 Dated June 2, 1970

Inventor(s) Vahe Keshishian

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

The title should read --ALPHA-PARTICLE-EMITTING-RADIOISOTOPE-FUELED GENERATOR--. Column 3, line 51, "plutonium-228" should read --plutonium-238--. Column 4, line 60, " $\text{Pu}_{82}^{238} 16$ " should read -- $\text{Pu}_{82}^{238} 16$ --. Column 6, line 48, "duel" should read --fuel--.

Signed and sealed this 6th day of March 1973.

(SEAL)
Attest:

EDWARD M. FLETCHER, JR.
Attesting Officer

ROBERT GOTTSCHALK
Commissioner of Patents