

NUCLEAR BATTERY - THERMOCOUPLE TYPE

FIRST QUARTERLY REPORT

January 1, 1957 - March 31, 1957

The object of this contract is to conduct research investigating radioactive materials and thermocouples suitable for the direct conversion of heat to electrical energy.

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U. S. Army Signal Engineering Laboratory

Project No. 162B

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Prepared By: B. C. Blanke

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TABLE OF CONTENTS

	Page
ABSTRACT	4
CONFERENCES	4
FACTUAL DATA	4
CONCLUSIONS	10
PROGRAM FOR NEXT INTERVAL	11
KEY TECHNICAL PERSONNEL	11
APPENDICES	14

PURPOSE

A thermal energy conversion system for the direct conversion of the heat energy derived from radioactive decay into electrical energy is being developed.

Initially, a survey of the literature is being made to determine the suitability with respect to half-life, shielding requirements, efficiency, health hazards, cost, and availability of radioactive isotopes. When isotopes having characteristics which indicate they may be suitable sources of heat are found, the heat source will be designed, and a prototype battery will be constructed.

ABSTRACT

The potential usefulness as a heat source for the thermal battery of 1300 radioactive isotopes was investigated. All but thirty-two have been eliminated because of too long or too short a half-life and/or excessive gamma radiation of the isotope or element. Further eliminations are being made on the basis of relative availability and costs.

CONFERENCES

A conference was held between Mound Laboratory personnel and Power Engineering Branch personnel of U. S. Army Signal Engineering Laboratories at Ft. Monmouth on January 18, 1957. The subject discussed was the battery program to be followed by Mound Laboratory. It was decided to begin with a literature search and an evaluation of radioactive material on the basis of half-life, shielding, efficiency, health hazards, availability and cost. Once the radioactive isotopes are selected, a heat source will be designed and a prototype manufactured. Problems involving thermocouple composition and design are to be carried out mainly by the Army Signal Engineering Laboratory, but are to be closely coordinated with Mound Laboratory.

FACTUAL DATA

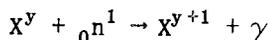
A search of the following literature was conducted in chronological order. Occasional pieces of information were derived from other sources.

1. Way, K., *Nuclear Data*, National Bureau of Standards Circular No. 499, (Sept. 1, 1950)
 Suppl. No. 1, (April 21, 1951)
 Suppl. No. 2, (Nov. 26, 1951)
 Suppl. No. 3, (June 9, 1952)
2. Wachtel, I. S., *Nuclear Data Index*, Tech. Info. Service, Oak Ridge, (1952) (Card system)
3. Hollander, Perlman, and Seaborg, "Table of Isotopes", *Rev. Modern Physics*, 25, 469-651 (1953)
4. *Chart of Nuclides*, General Electric Company, April, 1956.
5. Way, K., *Nuclear Data Cards*, Nat. Research Council (1957)
6. Sullivan, W. H., *Trilinear Chart of Nuclides*, 2nd Ed., Tech. Info. Service Oak Ridge (January, 1957)

Initial elimination of isotopes as heat sources was made on the basis of half-life. Any isotope which had a half-life of less than 100 days or greater than 100 years was discarded. A few exceptions were made at the shorter period to insure not overlooking a likely isotope. Further eliminations were made of isotopes which were gamma emitters only, which had radioactive transitions yielding gammas with energies greater than 1.0 million electron volts, which had transitions having an occurrence of gamma emission greater than ten per cent with energies over 0.1 million electron volts, which had decay schemes which involved daughter elements having any of the preceding gamma characteristics, or which had particle energies so slight that it would require more than one gram atomic weight of the isotope to construct a battery with a one per cent conversion efficiency to give a 0.01 watt output.

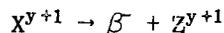
These nuclear property restrictions eliminated consideration of all but the following isotopes: Sulfur³⁵, Argon⁴², Calcium⁴⁵, Nickel⁶³, Krypton⁸⁵, Strontium⁸⁹, Strontium⁹⁰, Yttrium⁹¹, Cadmium^{113m}, Tin^{121m}, Tin¹²³, Cerium¹⁴⁴, Promethium¹⁴⁶, Promethium¹⁴⁷, Samarium¹⁵¹, Europium¹⁴⁹, Europium¹⁵², Europium¹⁵⁴, Europium¹⁵⁵, Gadolinium¹⁴⁸, Thulium¹⁷⁰, Thulium¹⁷¹, Lutetium¹⁷⁴, Tungsten¹⁸⁵, Tungsten¹⁸⁸, Osmium¹⁹⁴, Thallium²⁰⁴, Lead²¹⁰, Polonium²⁰⁸, Polonium²⁰⁹, Polonium²¹⁰, Curium²⁴².

Each of these elements was considered with regard to availability, or ease of manufacture and separation. Many of these elements may be manufactured in nuclear reactors in a reaction of the following type:



where X is the element of y atomic weight and γ is the residual excited energy. ${}_0n^1$ is a neutron.

Occasionally X^{y+1} has a very short half-life and decays into another isotope Z^{y+1} , which may be the desired isotope, thus:



For these reactions the following expression was derived (see Appendix A) to determine the amount of the desired isotope formed in a reactor:

$$N_2 = \frac{N_1^0 \sigma_1 nv}{\lambda_2 + nv(\sigma_2 - \sigma_1)} \left[e^{-nv\sigma_1 t} - e^{-[\lambda_2 + nv\sigma_2]t} \right]$$

where N_2 = number of moles of isotope X^{y+1} or Z^{y+1}

N_1 = number of moles of isotope X^y originally present in reactor.

[If N_1^0 equals Avogadro's number of molecules per mole, then the result will be in moles.]

σ_1 = neutron cross section of X^y in barns ($\text{cm}^2 \times 10^{-24}$)

σ_2 = neutron cross section of X^{y+1} or Z^{y+1} in barns ($\text{cm}^2 \times 10^{-24}$)

λ_2 = $\log_e 2$ / half-life of X^{y+1} or Z^{y+1} (in sec^{-1})

nv = flux of neutrons in reactor (neutrons/ cm^2/sec)

t = time (sec)

If a second order reaction of the type $X^y + {}_0n^1 \rightarrow X^{y+1} + {}_0n^1 \rightarrow X^{y+2}$ is required, the following expression for amount of isotope applies: (Appendix B)

$$N_3 = \frac{N_1^0 (nv)^2 \sigma_1 \sigma_2}{\lambda_2 + nv(\sigma_2 - \sigma_1)} \left[\frac{e^{-nv\sigma_1 t} - e^{-[\lambda_3 + nv\sigma_3] t}}{\lambda_3 + nv(\sigma_3 - \sigma_1)} + \frac{e^{-[\lambda_3 + nv\sigma_3] t} - e^{-[\lambda_2 + nv\sigma_2] t}}{\lambda_3 - \lambda_2 + nv(\sigma_3 - \sigma_2 + \sigma_1)} \right]$$

where N_3 = number of moles of isotope X^{y+2}

λ_3 = $\log_e 2$ /half-life of X^{y+2} (in sec^{-1})

σ_3 = neutron cross section of X^{y+2} in barns

To make our calculations, we have assumed reactor flux (nv) values of 10^{13} , 10^{14} , and 10^{15} n/cm²/sec which are nominal values, and irradiation times of 30 days, 90 days, 180 days, and 360 days. We have also assumed one mole (gram atomic weight) of starting isotope because this figure is easily converted into molecules, specific activity, or activity per unit volume.

Some of the desired isotopes are fission products, and their abundance can be calculated from previously calculated fission product mass ratios, and rates of uranium burn-up.

Power obtainable from any of these isotopes as heat may be calculated by determining the number and energy of the particles emitted per unit time, and then expressing this in terms of watts (see Appendix C).

$$\text{Power (watts/mole)} = \lambda N E \cdot 1.602 \times 10^{-19}$$

where E is the effective energy of the particle expressed in electron volts

For estimation purposes E is considered 100 per cent of α energy, 40 per cent of β energy and zero per cent of the γ energy.

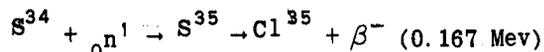
The total energy, E_t in watt-hours, given off by one mole of isotope over a one year period is:

$$E_t = (N_1^0 - N_1^{1yr}) \cdot E \cdot 4.45 \times 10^{23} \text{ (Appendix C)}$$

One factor of importance in determining the availability and power output of these isotopes is isotopic purity. In most cases, it would be economically unfeasible to separate the wanted isotope from other isotopes of the same element present or formed in the reactor. Calculations have been made to indicate the amount of dilution resulting for the desired isotope, and the decrease in power per mole in such cases.

Each isotope listed above is now considered in more detail with reference to Table I.

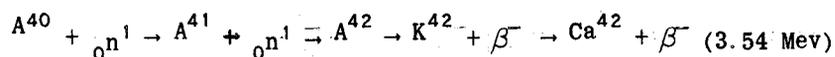
Sulfur³⁵ can be prepared by irradiating sulfur³⁴ in a reactor. Writing the complete reaction (in the reactor and later decay in the heat source ignoring very short-lived metastable states formed in the reactor).



Sulfur³⁴ occurs to the extent of 4.215 per cent in natural sulfur. The other isotopes of sulfur on acceptance of a neutron become other stable isotopes of sulfur except sulfur³⁶ (0.017 % of natural sulfur) which forms a very short-lived radioisotope and would not be present after a short time.

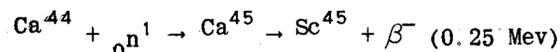
Since sulfur forms a number of stable gases (SO₂, H₂S) it may be possible to separate sulfur isotopes more easily than most others. Techniques of using thermal columns or diffusion barriers for such separations are well known.

Argon⁴² would be the second order product of neutron irradiation of argon⁴⁰ (99.4% of natural argon).



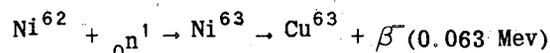
Since this is a second order reaction, it would give a very low yield. The gas would be somewhat difficult to handle in the pile as it is above its critical temperature (cannot be liquified). This would also be true of argon as a heat source. However, since it is a gas, it can be separated from other argon isotopes.

Calcium⁴⁵ can be prepared by irradiation of calcium⁴⁴ (2.07% of natural calcium)



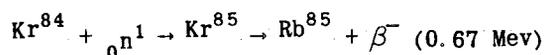
There is no feasible means of isotopic separation for large amounts of calcium. This means that the calcium would be contaminated with Ca⁴² from Ca⁴⁰ (96.96% of natural calcium). Since calcium⁴² has a long (2 X 10⁵ years) half-life and emits a weak gamma, this diluent would not cause difficulties.

Nickel⁶³ can be prepared by irradiating nickel⁶² (3.66% of natural nickel).



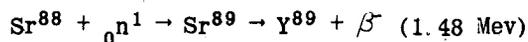
There is no feasible method for separation of large quantities of nickel isotopes. There will be some contamination by formation of nickel⁵⁹ from nickel⁵⁸ (67.8% of natural nickel). Nickel⁵⁹ has an 80,000 year half-life and is a positron emitter. The long half-life should keep the gamma emission at a sufficiently low level.

Krypton⁸⁵ is a fission product, that is, a waste product of uranium fuel burn-up in a reactor. Mass 85 is formed to the extent 1.3% of the mass of uranium fissioned. Krypton⁸⁵ occurs to 45.7% of krypton gases formed as fission products. It can be easily separated from most other fission products as it is an unreactive gas. Separation would be required from xenon, but this could be managed by fractional distillation. Krypton⁸⁵ can also be made by neutron irradiation of krypton⁸⁴ (56.9% of natural krypton).



There is a slight contamination with krypton⁸¹ (half life 2.1×10^5 years) formed from krypton⁸⁰ (2.27 of natural krypton) (which emits a characteristic x-ray on electron capture). Since krypton is a gas, isotopic separation is feasible, but difficulties are encountered in handling a gas above its critical temperature.

Strontium⁸⁹ may be prepared by irradiating naturally occurring strontium⁸⁸ (82.6% of natural strontium).



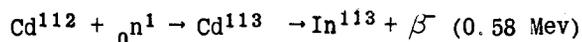
There will be a small amount of contamination with strontium⁸⁵ (half-life of 65 days, a weak gamma emitter) formed from strontium⁸⁴ (0.55% of natural strontium).

Strontium⁹⁰ also occurs as a fission product to the extent of 4.8% (maximum) of the mass of fissionable material reacted.

Strontium⁹⁰ is a fission product whose mass occurs to 5.6% of the mass of fissionable material reacted. A mixture of strontium⁹⁰ and strontium⁹⁰; or pure strontium⁹⁰ can be obtained by chemical means by allowing the fission products to age. The values for strontium⁹⁰ include those of its short-lived daughter, yttrium⁹⁰.

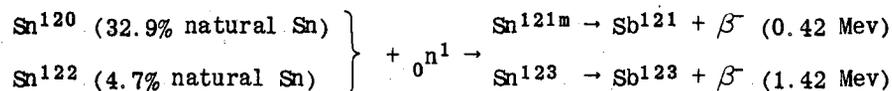
Yttrium⁹¹ is a fission product whose mass occurs to 5.8% of the fissionable material reacted. There is no first or second order method of producing yttrium⁹¹ in the reactor.

Cadmium^{113m} may be prepared from cadmium¹¹² (24.07% of natural cadmium) by neutron irradiation.



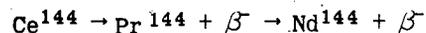
There is some contamination with cadmium¹⁰⁹ (half-life 1.3 years) formed from cadmium¹⁰⁸ (0.88% of natural cadmium) which emits a characteristic x-ray on electron capture. Because of cadmium's very large cross section, it is unlikely that any large quantity could be irradiated without causing the reactor to become inoperational.

The tin isotopes, tin^{121m} and tin¹²³ should be considered jointly as irradiation products. There is no inexpensive method at present for separating these isotopes. Mass 121 occurs as 0.015% and mass 123 as 0.019% of the fissionable material reacted.



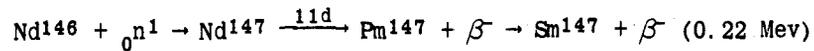
There would be some contamination in both cases with tin¹¹³ (112-day half-life), which is formed by irradiation of the tin¹¹² (0.95% of natural tin) and which emits a characteristic X-ray on electron capture.

Cerium¹⁴⁴ is a fission product whose mass occurs to the extent of 5.7% of the fissioned material. Since it has a very short-lived daughter, praseodymium¹⁴⁴, their energies have been combined in Table I to give the values for power.

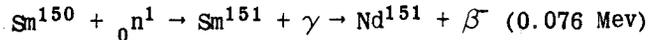


Promethium¹⁴⁶ is not a fission product, nor does there appear to be a good production method for this isotope in large quantity.

Promethium¹⁴⁷ is a fission product and its mass occurs to 2.4% of the fissionable material burned up. It can also be prepared by irradiating neodymium¹⁴⁶ (17.26% of natural neodymium).



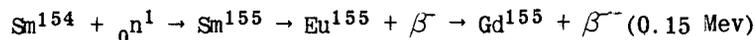
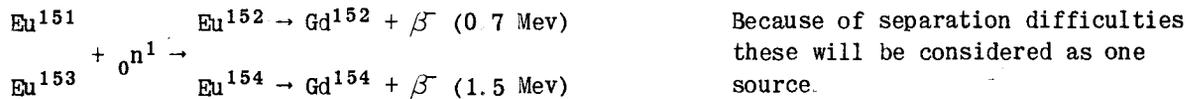
Samarium¹⁵¹ occurs to 1.1% of the fissionable material reacted. However, there are many other samarium fission products and Sm¹⁵¹ occurs as 11.2% of these. Sm¹⁵¹ may also be prepared by reactor irradiation of Sm¹⁵⁰ (7.4% of natural isotopes).



Europium¹⁴⁹ was eliminated from consideration as no feasible production scheme was found.

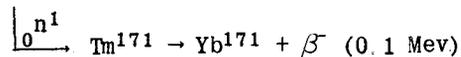
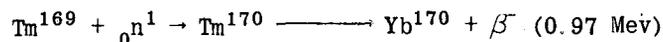
Europium¹⁵⁵ mass occurs 0.03% of the mass of fissionable material reacted. There are several europium fission products and Eu¹⁵⁵ is 4.36% of the longer-lived isotopes.

Europium^{152, 154} and ¹⁵⁵ may also be prepared by reactor irradiation. Their parents occur in natural isotopes in the following percentages: Eu¹⁵¹ (47.77%), Eu¹⁵³ (52.23%) and Sm¹⁵⁴ (22.35%).

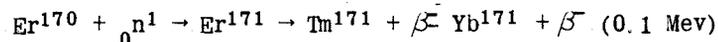


Gadolinium¹⁴⁸ does not seem to have a feasible large production scheme.

Thulium¹⁷⁰ may be produced by irradiating naturally occurring thulium¹⁶⁹ (100% of natural isotope). There is a large 2nd order production of thulium¹⁷¹. These will be considered together as one heat source.

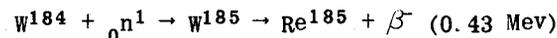


Thulium¹⁷¹ may also be produced by irradiating erbium¹⁷⁰ (14.9% of natural isotope)

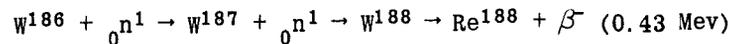


Lutetium¹⁷⁴ does not seem to have a feasible large production scheme.

Tungsten¹⁸⁵ may be prepared by irradiating tungsten¹⁸⁴ (30.6% of naturally occurring isotopes).

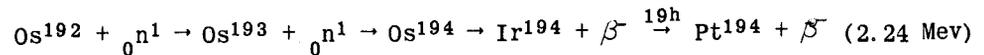


Tungsten¹⁸⁸ may be produced by second order irradiation of tungsten¹⁸⁶ (28.4% of natural isotopes).



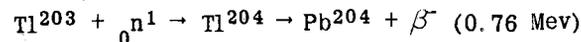
A small amount of tungsten¹⁸¹ (half-life 140 days, which has a characteristic x-ray) from tungsten¹⁸⁰ (0.13% of natural tungsten) will contaminate the tungstens which are considered together as one heat source.

Osmium¹⁹⁴ may be produced by second order irradiation of osmium¹⁹² (41.0% of natural isotopes).



This would be contaminated by osmium¹⁸⁵ (95 d half life with a characteristic x-ray) from Osmium¹⁸⁴ (0.018% of natural osmium).

Thallium²⁰⁴ may be produced by irradiating thallium²⁰³ (29.5% of natural isotopes) in a reactor.

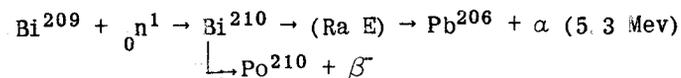


Lead²¹⁰ is radium D of the U²³⁸ decay chain, and can be milked from radium. However, the amounts produced in this way are limited by the moderately long (1638 yr) half life of radium and the relative scarcity of radium. Lead²¹⁰ does have the advantage of having short-lived daughters (one β emitter, and one α emitter) which are quite energetic.

There is no feasible production scheme for Pb²¹⁰.

Polonium²⁰⁸ and ²⁰⁹ have no feasible scheme of production.

Polonium²¹⁰ is prepared by neutron irradiation of bismuth²⁰⁹ (100% of natural isotope)



This isotope has the advantage of being produced in fair quantity as a by-product of bismuth-cooled reactors, and no new production plant need be built.

Curium²⁴⁴ is the third order, second decay product of plutonium²³⁹. Normally this would be too remote for consideration, except that in this case plutonium is a major product of the reactor fuel irradiation.

CONCLUSIONS

The number of isotopes which can be considered as useful heat sources have been reduced from 1300 to 32 on the basis of nuclear and physical properties. A number of production considerations for these 32 isotopes were presented to further eliminate those isotopes whose production would be impractical at present.

FUTURE PROGRAM

The program for next interval is to complete the evaluation of the isotopes on the basis of properties and feasibility of preparation. On the basis of their activity, isotopic dilution, and other properties, volume/surface area ratios and temperature ranges available for the more promising isotopes as heat sources of various sizes and designs will be calculated.

IDENTIFICATION OF PERSONNEL

Bertram C. Blanke (300 hours), Acting Group Leader, received his BS in chemistry in 1938 and MS in bacteriology in 1940 from the University of Illinois where he was chief chemist for the University Health Service. He worked for Citrus Products Company of Chicago and Santa Ana, California as production and research chemist until drafted by the Army in 1942. He was discharged a major in December, 1945 and joined Monsanto in January, 1946 where he worked on research and development. Monsanto granted an academic leave in 1951 for two and one-half years study in physical chemistry at University of Kansas. Since his return to Mound Laboratory his work has been on physical measurements. He has been acting group leader since August, 1956.

Josef W. Heyd (200 hours) received his BSChE in 1931 and MS in 1932 from Washington University, and his PhD in 1937 from Pennsylvania State University. He joined Monsanto's Central Research Department in 1937 where his work included research on resins and on catalytic oxidation. He transferred to the unit which is now Mound Laboratory in 1944 where he was successively Leader of Electronics Group, Chief of Electronics Section, of the Electronic Research Section and of the Technical Information Section.

Kenneth C. Jordan (300 hours) graduated from the University of Maine with a BS in Engineering Physics in 1943. He served in the US Army Signal Corps two and one-half years assigned to communications (radio telephone). He was hired by Monsanto in 1947 and assigned to calorimetry where he conducted assays and half-life determinations. He has constructed a high-temperature calorimeter and a polonium thermal battery.

John H. Birden (480 hours) received his BS degree at Indiana Central College in 1942. He has been a research chemist at Monsanto since 1944. He worked in process development one year, the electronic division one year, and has worked in neutron source development since 1946. He constructed the polonium heat source for the thermal battery.

TABLE I

POWER/MOLE FOR VARIOUS ISOTOPIC HEAT SOURCES

ISOTOPE	HALF-LIFE	POWER WHEN REMOVED FROM REACTOR (WATTS)			ENERGY PER MOLE OF PURE ISOTOPE PER YEAR <i>watt-hours</i>	REMARKS
		PER MOLE OF PURE ISOTOPE	PER MOLE OF PURE PARENT IRRADIATED	PER MOLE OF NATURAL PARENT IRRADIATED		
SULFUR-35	87.1 d	5.93×10^2	1.57×10^{-1}	6.62×10^{-3}	7.69×10^6	
ARGON-42	3.5 y	-	-	-	-	INSUFFICIENT DATA
CALCIUM-45	164 d	4.79×10^2	5.21	1.08×10^{-1}	2.74×10^6	
NICKEL-63	80 y	6.67×10^{-1}	1.35×10^{-1}	4.94×10^{-3}	5.82×10^3	
KRYPTON-85	10.6 y	4.94×10^1	2.03×10^{-1}	1.16×10^{-1}	4.66×10^6	
STRONTIUM-89	51 d	8.88×10^3	(1.61×10^{-3})	(1.33×10^{-3})	(3.56×10^7)	
STRONTIUM-90	28 y	8.62×10^1	-	-	7.45×10^5	COMBINED WITH YTTRIUM-90
⁹⁰ STRONTIUM-89 + 90(FP)	-	-	-	-	-	
YTTRIUM-91	58 d	8.26×10^3	-	-	5.64×10^7	
CADMIUM-113m	5.1 y	9.63×10^1	1.37×10^{-4}	3.3×10^{-5}	7.89×10^6	
TIN-121m	5 y	7.09×10^1	4.66×10^{-2}	2.19×10^{-3}	$\left\{ \begin{array}{l} 5.60 \times 10^5 \\ 1.30 \times 10^7 \end{array} \right\}$	AS IRRADIATED PRODUCT †
TIN-123	131 d	3.35×10^2				
CERIUM-144	290 d	3.40×10^3	-	-	1.99×10^7	INCLUDES PRASEODYMIUM-144
PROMETHIUM-146	2 y	5.93×10^2	-	-	3.75×10^6	
PROMETHIUM-147	2.52 y	7.48	-	-	5.73×10^5	
SAMARIUM-151	100 y	6.35×10^{-3}	-	-	5.55×10^3	
EUROPIUM-152	13 y	1.17×10^1	-	-	1.00×10^6	

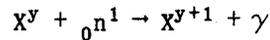
TABLE 1 (CONT'D.)

POWER/MOLE FOR VARIOUS ISOTOPIC HEAT SOURCES

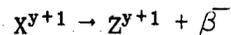
ISOTOPE	HALF-LIFE		POWER WHEN REMOVED FROM REACTOR (WATTS)			ENERGY PER MOLE	REMARKS
			PER MOLE OF PURE ISOTOPE	PER MOLE OF PURE PARENT IRRADIATED	PER MOLE OF NATURAL PARENT IRRADIATED	OF PURE ISOTOPE PER YEAR <i>watt-hours</i>	
EUROPIUM-154	16	y	1.32×10^{-1}	-	-	6.25×10^5	
EUROPIUM-155	1.7	y	8.36×10^{-1}	3.17×10^{-2}	7.14×10^{-3}	6.29×10^5	
GADOLINIUM-148	35	y	4.78×10^3	-	-	4.15×10^5	
THULIUM-170	129	d	2.31×10^3	3.18×10^2	3.18×10^2	8.76×10^6	
THULIUM-171	693	d	4.59×10^1	1.13×10^2	1.68×10^1	3.37×10^5	
LUTETIUM-174	165	d	2.25×10^2	-	-	1.01×10^6	
TUNGSTEN-185	74	d	1.80×10^3	2.93×10^1	8.97	4.46×10^6	
TUNGSTEN-188	65	d	-	-	-	-	INSUFFICIENT DATA
OSMIUM-194	-	-	-	-	-	-	INSUFFICIENT DATA
THALLIUM-204	3	y	2.10×10^2	5.4×10^1	1.59×10^1	1.64×10^6	
LEAD-210	19.4	y	6.30×10^2	-	-	5.43×10^6	INCLUDES Po210, Bi210
POLONIUM-208	2.93	y	3.69×10^3	-	-	2.88×10^7	
POLONIUM-209	100	y	9.28×10^1	-	-	8.11×10^5	
POLONIUM-210	138	d	2.97×10^4	8.12	8.12	1.19×10^8	
CURIUM-244	162.5	d	2.90×10^4	-	-	1.29×10^8	

APPENDIX A

An isotope, X^{y+1} , can be prepared in a nuclear reactor from isotope X^y by the following reaction:



where y is the atomic number. If the desired isotope, X^{y+1} decays to isotope, Z^{y+1} , by emission of a β^- particle, thus:



then an expression may be derived as follows for the number of moles of desired isotope formed in a reactor having a flux of neutrons, nv .

Let N_1^0 be the number of atoms of X^y before irradiation,

N_1^c be the number of atoms of X^y at time t , and

N_2 be the number of atoms of X^{y+1} ,

then:

$nv =$ flux

$$\frac{dN_2}{dt} = -N_2\lambda_2 + N_1\sigma_1nv - N_2\sigma_2nv$$

$$\lambda_2 = \frac{0.693}{t_{1/2}}$$

$\sigma_1 =$ neutron cross section of X^y

$\sigma_2 =$ neutron cross section of X^{y+1}

$$N_1 = N_1^0 e^{-\sigma_1 nvt}$$

$$\frac{dN_2}{dt} = -N_2\lambda_2 + N_1^0 \sigma_1 nv e^{-\sigma_1 nvt} - N_2\sigma_2 nv$$

$$\frac{dN_2}{dt} + N_2 [\lambda_2 + \sigma_2 nv] = N_1^0 \sigma_1 nv e^{-\sigma_1 nvt}$$

$$N_2 = e^{-(\lambda_2 + \sigma_2 nv)t} \int_0^t N_1^0 \sigma_1 nv e^{-\sigma_1 nvt + \lambda_2 t + \sigma_2 nvt} dt =$$

$$e^{-(\lambda_2 + \sigma_2 nv)t} N_1^0 \int_0^t \sigma_1 nv e^{[\lambda_2 + nv(\sigma_2 - \sigma_1)]t} dt$$

$$= N_1^0 e^{-(\lambda_2 + \sigma_2 nv)t} \left[\frac{e^{-\sigma_1 nv}}{\lambda_2 + nv(\sigma_2 - \sigma_1)} \right] \left[e^{\lambda_2 t + nv(\sigma_2 - \sigma_1)t} - 1 \right]$$

$$= \frac{N_1^0 \sigma_1 nv}{\lambda_2 + nv(\sigma_2 - \sigma_1)} \left[e^{-nv\sigma_1 t} - e^{-[\lambda_2 + nv\sigma_2]t} \right]$$

(1)

The power output, in watts per mole, may be calculated from the following derived equation.

$$\begin{aligned}
 \text{Watts} &= \lambda N_2 \bar{\beta} \cdot 1.602 \times 10^{-19} & \bar{\beta} &= \beta_{ev} \times 0.4 \\
 &= \frac{1.602 \times 10^{-19} \lambda \bar{\beta} N_1^0 \sigma_1 nv}{\lambda + nv(\sigma_2 - \sigma_1)} \left[e^{-nv\sigma_1 t} - e^{-[\lambda + nv\sigma_2] t} \right] \\
 \text{Watts/mole} &= \frac{9.644 \times 10^4 \lambda \bar{\beta} \sigma_1 nv}{\lambda + nv(\sigma_2 - \sigma_1)} \left[e^{-nv\sigma_1 t} - e^{-[\lambda + nv\sigma_2] t} \right] & (2)
 \end{aligned}$$

$$N_3 = \frac{N_1^0 (nv)^2 \sigma_1 \sigma_2}{\lambda_2 + nv(\sigma_2 - \sigma_1)} \left[\frac{e^{-nv\sigma_1 t} - e^{-[\lambda_3 + nv\sigma_3]t}}{\lambda_3 + nv(\sigma_3 - \sigma_1)} + \frac{e^{-[\lambda_3 + nv\sigma_3]t} - e^{-[\lambda_2 + nv\sigma_2]t}}{\lambda_3 - \lambda_2 + nv(\sigma_3 - \sigma_2 + \sigma_1)} \right] \quad (3)$$

APPENDIX C

An expression giving the energy obtainable per year from a radioisotope heat source may be derived thus:

$$\frac{dN}{dt} = -\lambda N$$

$$\ln N \left[\begin{array}{l} N_2 \\ N_1 \end{array} = -\lambda t \right] \begin{array}{l} t = 365 \text{ d} = 3.16 \times 10^7 \text{ sec} \\ t = 0 \end{array}$$

$$\ln \frac{N_1}{N_2} = 3.16 \times 10^7 \lambda \qquad \log \frac{N_1}{N_2} = 1.372 \times 10^7 \lambda$$

$$\text{Energy} = (N_1 - N_2) E \cdot 1.602 \times 10^{-19} / 3600$$

$$= \Delta N E \cdot 4.45 \times 10^{-23} \text{ Watt-hours/1 year}$$