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NUCLEAR BATTERY-THERMOCOUPLE TYPE SUMMARY REPORT

B. C. Blanke
J. H. Birden
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E. L. Murphy

AEC Research and Development REPORT

MONSANTO RESEARCH CORPORATION
A SUBSIDIARY OF MONSANTO CHEMICAL COMPANY

Monsanto

MOUND LABORATORY
MIAMISBURG, OHIO
OPERATED FOR
UNITED STATES ATOMIC ENERGY COMMISSION
U.S. GOVERNMENT CONTRACT NO. AT-33-1-GEN-53

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

U. S. Army Signal Corps Contract No. R-65-8-99811-SC-03-91

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ABSTRACT

The potential usefulness of approximately 1300 radioactive isotopes as a heat source for the thermoelectric generator was investigated. Only 47 are found to have the proper characteristics of high specific activity and usable half-life combined with an easily absorbable radiation. These isotopes are discussed showing possible sources of supply, the hazards involved and the expected performance. Three large polonium-210 heat sources were designed and constructed (for battery use), including one for the SNAP III generator. One small thallium-204 test heat source was made by irradiation. Eight thermoelectric generators were developed and two of these were used as thermoelectric batteries. Theoretical equations for non-semiconductor thermoelectric materials and experimental measurements to verify the Thompson and Peltier effects are given.

INTRODUCTION

The purpose of this project was to examine systems for the direct conversion of heat derived from radioactive decay into electrical energy by means of a thermopile. Emphasis was placed on study of all reported nuclides to determine those which would be feasible as nuclear heat power sources. The development of theory and the application of conventional thermoelectric materials to an atomic battery was also undertaken.

The original work on thermoelectricity at Mound Laboratory was started in 1953 to demonstrate that nuclear heat could beused for this purpose. In 1954 two patent applications were filed, and subsequently granted,[1, 2] for batteries using radioactive decay as the heat source and thermoelectricity for the generator. Results from these investigations were published in MLM-984 and MLM-1060.[3, 4]

In 1957 Mound Laboratory was approached by the US Army Signal Corps Research and Development Laboratories, Ft. Monmouth, New Jersey to begin an evaluation of radioactive material for use as a heat source in a thermoelectric generator. Half-life, shielding required, efficiency, health hazards, availability and costs were to be considered. Once the radioisotope was selected, a heat source would be designed and prototypes would be constructed. The thermoelectric generator design would use conventional materials, or material and information furnished by the Signal Corps Laboratory.

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 to insure not overlooking a likely isotope. Further eliminations were made of isotopes which (1) were gamma emitters only, (2) had radioactive transitions yielding gamma with energies greater than one million electron volts, (3) had transitions having an occurrence of gamma emission greater than ten per cent with energies over 0.1 million electron volts, (4) had decay schemes which involved daughter elements having any of the preceeding gamma characteristics, or (5) had particle energies so slight that it would require more than one per cent conversion efficiency to give a 0.01 watt output. A literature search of the nuclides was conducted to eliminate isotopes which did not have desired nuclear properties. Calculations were based primarily on values obtained from the Trillinear Chart of Nuclides by William H. Sullivan.

These nuclear property restrictions eliminated consideration of all but the following isotopes: sulfur-35, argon-42, calcium-45, nickel-63, krypton-85, strontium-89, strontium-90, yttrium-91, cadmium-113m, tin-121m, tin-123, cerium-144, promethium-146, promethium-147, samarium-151, europium-149, europium-152, europium-154, europium-155, gadolinium-148, thulium-170, thulium-171, lutetium-174, tungsten-185, tungsten-188, osmium-194, thallium-204, lead-210, polonium-208, polonium-209, polonium-210, radium-228, actinium-227, thorium-228, uranium-232, plutonium-236, plutonium-238, americium-241, americium-242, curium-242, curium-243, curium-244, californium-248, californium-250, californium-252, einsteinium-252 and einsteinium-254.

All of the above isotopes, with the exception of naturally occuring lead-210, polonium-210 and several of the fission products, must be manufactured, and almost all of them must be separated to obtain a sufficiently high specific activity. Discussions of their manufacture and separation are given in "Nuclear Battery - Thermocouple Type", first and second quarterly reports.[5, 6]

One method, which was not mentioned in those reports for their manufacture, was suggested at PLOWSHARE II Conference, which considered the peaceful use of atomic explosives. When atomic explosions are confined, as in underground shots, there is a chance that multiple neutron additions can be made to the plutonium or uranium in the critical mass, which fissions less than two per cent of its total weight. In the intense neutron flux of the explosion, which is many orders of magnitude greater than that of any reactor, these metals are confined and many isotopes which could never be formed in any other way might be manufactured with the help of some very unstable short-lived intermediate. If the shots were fired in a salt bed, as planned in project GNOME, this material could be mined after the heat of the explosion had been utilized, and these unusual isotopes separated from the mined salt. Rough separation problems would not be too difficult because of the chemical differences between salt and actinides, and the separation of the actinides is routine at many of the AEC sites.

In the following discussion, tables and graphs are used to characterize each of the possible usable nuclides. Wattage values and temperatures obtainable with these isotopes, as well as for probable chemical compounds used, were also given. Values were also listed for power to be expected if a mole of a naturally occuring element were irradiated for given times and reactor flux.

Shielding requirements have only been sketchily discussed because of the extreme variability in use and in characteristics of the power sources. It can be stated that in every case some shielding will be required for the separation, preparation, and handling of the source, and in many cases heavy shielding will be required. Over total body area 25 REM is generally considered a maximum permissible limit for man and 450 REM is considered a 50 per cent lethal dose (LD50). Plastics and electronics materials as transisters are affected by roughly 10^5 REM, ordinary construction materials such as aluminum show changes at 10^7 REM.

Even an isotope emitting almost pure alpha particles such as polonium-210, exhibits over seven REM per hour at the surface of a source giving off 26.1 watts of heat. All other alpha sources considered have more intense radiations associated with them, and all the beta sources have, in addition to their associated gammas, bremsstrahlung from self absorption of the beta radiation. The nuclide containers can be used as portable shielding for many of the sources, and careful exposure, timing, and the use of handling tools such as tongs to provide distance, will permit usage under very carefully controlled conditions. Toxicity of any radioactive material is extremely high compared with the toxicity of normal poisons. The biological half-life of each compound of each nuclide differs greatly and is also dependent on mode of entry such as ingestion, inhalation or injection, as well as its physiological chemistry. Packaging and use of the heat sources must preclude source container rupture in case of any accident, even an accident of large magnitude, to avoid contamination or interdiction of areas. Certain isotopes, considerably more difficult to contain such as argon-42 and krypton-85, however, would escape into the atomosphere leaving little residual activity (to worry about).

ISOTOPIC DISCUSSION

The nuclear properties of the isotopes are summarized in Table 1.

Sulfur-35, which can be prepared by irradiating sulfur-34 in a reactor, has a specific heat activity of 16.9 watts per gram.

$$S^{34} + {}_0n^1 \rightarrow S^{35} \rightarrow Cl^{35} + \beta^- \text{ (0.167 Mev)}$$

Sulfur-34 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 sulfur-36 (0.017% of natural sulfur) which forms a very short-lived radioisotope and would not be present a short time after removal from a reactor. Since sulfur forms a number of stable gasses (SO_2, H_2S), it may be possible to separate sulfur isotopes more easily than most others. Techniques of using thermal diffusion columns, diffusion barriers or other methods for such separations are well known.

Sulfur has a melting point of 112.8°C and a boiling point of 444.6°C and would be a low pressure gas at temperatures required for efficient operations.

Argon-42 would be the second order product of neutron irradiation of argon-40 (99.% of natural argon).

$$A^{40} + {}_0n^1 \rightarrow A^{41} + {}_0n^1 \rightarrow A^{42} \rightarrow K^{42} + \beta^- \rightarrow Ca^{42} + \beta^- \text{ (3.54 Mev)}$$

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. 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 by thermal diffusion or other gaseous isotope separation techniques.

Calcium-45 with a specific heat activity of 10.6 watts per gram can be prepared by irradiation of calcium-44 (2.0% of natural calcium).

$$Ca^{44} + {}_0n^1 \rightarrow Ca^{45} \rightarrow Sc^{45} + \beta^- \text{ (0.25 Mev)}$$

There is no feasible means of isotopic separation for large amounts of calcium. This means that the calcium-44 would be contaminated with calcium-41 from calcium-40 (96.96% of natural calcium). Since calcium-41 has a long (2×10^5 years) half-life and emits a weak gamma, this diluent would not cause difficulties except to lower the specific activity. Since the melting point of calcium is 842-8°C, and the boiling point is 1240°C, the oxide or fluoride form would be less demanding in their fabrication into heat sources.

Nickel-63 with a specific activity of 0.0106 watts per gram can be prepared by irradiating nickel-62 (3.66% of natural nickel).

$$Ni^{62} + {}_0n^1 \rightarrow Ni^{63} \rightarrow Cu^{63} + \beta^- \text{ (0.063 Mev)}$$

As with calcium, there is no feasible method for separation of large quantities of nickel isotopes. Although nickel carbonyl is a gas, it is thermally unstable and would decompose on attempts to separate by thermal diffusion and also tend to deposit in a barrier used for separation. There will be some contamination by formation of nickel-59 from nickel-58 (67.8% of natural nickel). Nickel-59 has an 80,000 year half-life and is a positron emitter. The long half-life should keep the gamma emmission at a sufficiently low level. Nickel has a melting point of 1455°C and a boiling point of 2900°C; hence containment would offer no problem.

Krypton-85 has a specific activity of 0.631 watts per gram and is a fission product, that is, a waste product of uranium fuel. Mass 85 is formed to the extent of 1.3 per cent of the mass of uranium fissioned. Krypton-85 occurs to 45.7 per cent 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 physical methods of separation. Krypton-85 can also be made by neutron irradiation of krypton-84 (56.9% of natural krypton).

$$Kr^{84} + {}_0n^1 \rightarrow Kr^{85} \rightarrow Rb^{85} + \beta^- \text{ (0.67 Mev)}$$

There is a slight contamination with krypton-81 (half-life 2.1×10^5 years) formed from krypton-80 (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 would be encountered in condensing a gas above its critical temperature. The boiling point of krypton is -152.9°C. Confinement under the heat and pressures required for efficiency might present a problem.

Strontium-89 with a specific activity of 99.7 watts per gram can be prepared by irradiating naturally occurring strontium-88 (82.6% of natural strontium).

$$Sr^{88} + {}_0n^1 \rightarrow Sr^{89} + \beta^- \text{ (1.48 Mev)}$$

There will be a small amount of contamination with strontium-85 (half-life of 65 days, a weak gamma emitter) formed from strontium-84 (0.55% of natural strontium).

Strontium-89 also occurs as a fission product to the extent of 4.8 per cent (maximum) of the mass of fissionable uranium reacted.

Strontium-90 with a specific activity of 45.6 watts per gram is a fission product whose mass occurs to 5.6 per cent of the mass of fissionable material reacted. A mixture of strontium-89 and strontium-90, can be chemically separated from fission products or pure strontium-90 can be obtained by allowing the fission products to age before chemical separation.

The watt values given for strontium-90 include those of its short-lived daughter, yttrium-90. Since strontium is a very toxic bone-seeker, only the higher melting and most stable compounds, such as the oxides and borides, have been considered as heat sources. Its primary advantage is the availability as a fission product in relatively pure forms.

Yttrium-91 has a specific activity of 90.9 watts per gram and is a fission product whose mass occurs to 5.8 per cent of the fissionable material reacted. There is no first or second order method of producing yttrium-91 in the reactor.

Cadmium-113m with a specific activity of 0.853 watts per gram may be prepared from cadmium-112 (24.07% of natural cadmium) by neutron irradiation.

$$Cd^{112} + {}_0n^1 \rightarrow Cd^{113} \rightarrow In^{113} + \beta^- \text{ (0.58 Mev)}$$

There is some contamination with cadmium-109 (half-life 1.3 years) formed from cadmium-108 (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. It has a melting point of 320.9°C and a boiling point of 767°C, so that a compound such as the oxide might be considered to avoid confinement difficulties.

The tin isotopes, tin-121m with a specific activity of 0.588 watts per gram, and tin-123 with a specific activity 0.272 watts per gram, should be considered jointly as irradiation products. There is no inexpensive method at present for separating these isotopes. Mass 121 occurs as 0.015 per cent and mass 123 as 0.019 per cent of the fissionable uranium reacted:

$$\left.\begin{array}{l} Sn^{120} \text{ (32.9\% natural Sn)} \\ Sn^{122} \text{ (4.7\% natural Sn)} \end{array}\right\} + {}_0n^1 \left\{\begin{array}{l} Sn^{121m} - Sb^{121} + \beta^- \text{ (0.42 Mev)} \\ Sn^{123} \quad - Sb^{123} + \beta^- \text{ (1.42 Mev)} \end{array}\right.$$

There would be some contamination in both cases with tin-113 (112-day half-life), which is formed by irradiation of the tin-112 (0.95% of natural tin) and which emits a characteristic x-ray on electron capture

Cerium-144 is a fission product whose mass occurs to the extent of 5.7 per cent of the fissioned uranium. Since it has a very short-lived daughter, praseodymium-144, their energies have been combined to give the specific power activity of 23.6 watts per gram.

$$Ce^{144} \rightarrow Pr^{144} + \beta^- \rightarrow Nd^{144} + \beta^-$$

Since it is a fission product, large quantities can be separated from burned out reactor fuel.

Promethium-146 with a specific activity of four watts per gram is not a fission product, nor does there appear to be a good production method for this isotope in large quantity.

Promethium-147 with a specific activity of 0.5 watt per gram is a fission product and its mass occurs to 2.4 per cent of the fissioned uranium. It can also be prepared by irradiating neodymium-146 (17.26% of natural neodymium).

$$Nd^{146} + {}_0n^1 \rightarrow Nd^{147} \xrightarrow{11d} Pm^{147} + \beta^- \rightarrow Sm^{147} + \beta^- \text{ (0.22 Mev)}$$

Samarium-151, specific activity of 4.21×10^{-3} watt per gram, occurs to 1.1 per cent of the fissioned uranium. However, there are many other samarium fission products and samarium-151 occurs as 11.2 per cent of these. Samarium-151 may also be prepared by reactor irradiation of samarium-150 (4.4% of natural isotopes).

$$Sm^{150} + {}_0n^1 \rightarrow Sm^{151} + \gamma \rightarrow Nd^{151} + \beta^- \text{ (0.076 Mev)}$$

Europium-149 was eliminated from consideration as no feasible production scheme was found.

Europium-155 with a 0.54 watt per gram specific activity occurs as 0.03 per cent of the mass of fissioned uranium. There are several europium fission products and europium-155 is 4.36 per cent 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: europium-151 (47.77%), europium-153 (52.23%) and samarium-154 (22.35%).

Because of separation difficulties these will be considered as one source with a specific activity of 2.09×10^{-4} watt per gram.

$$\left.\begin{array}{l} Eu^{151} \\ Eu^{153} \end{array}\right\} + {}_0n^1 \rightarrow \left\{\begin{array}{l} Eu^{152} \rightarrow Gd^{152} + \beta^- \text{ (0.07 Mev)} \\ Eu^{154} \rightarrow Gd^{154} + \beta^- \text{ (1.5 Mev)} \end{array}\right.$$

$$Sm^{154} + {}_0n^1 \rightarrow Sm^{155} \rightarrow Eu^{155} + \beta^- \rightarrow Gd^{155} + \beta^- \text{ (0.15 Mev)}$$

Gadolinium-148, specific activity of 0.323 watt per gram, does not seem to have a feasible large production scheme.

Thulium-170 with a specific activity of 13.5 watts per gram may be produced by irradiating naturally occurring thulium-169 (100% of natural isotope). There is a large second order production of thulium-171 (specific activity 0.267 per gram)

$$\begin{tikzcd}
Tm^{169} + {}_0n^1 \arrow[r] & Tm^{170} \arrow[r] \arrow[d] & Yb^{170} + \beta^- \text{ (0.97 Mev)} \\
& {}_0n^1 \quad Tm^{171} \arrow[r] & Yb^{171} + \beta^- \text{ (0.1 Mev)}
\end{tikzcd}$$

Thulium-171 may also be produced by irradiating erbium-170 (14.9% of natural isotope).

$$Er^{170} + {}_0n^1 \rightarrow Er^{171} \rightarrow Tm^{171} + \beta^- \rightarrow Yb^{171} + \beta^- \text{ (0.1 Mev)}$$

Lutetium-174 having specific activity of 1.29 watts per gram does not seem to have a feasible large production scheme.

Tungsten-185 specific activity of 9.7 watts per gram may be prepared by irradiating tungsten-184 (30.6% of naturally occurring isotopes).

$$W^{184} + {}_0n^1 \rightarrow W^{185} \rightarrow Re^{185} + \beta^- \text{ (0.43 Mev)}$$

Tungsten-188 may be produced by second order irradiation of tungsten-186 (28.4% of natural isotopes).

$$W^{186} + {}_0n^1 \rightarrow W^{187} + {}_0n^1 \rightarrow W^{188} \rightarrow Re^{188} + \beta^- \text{ (0.43)}$$

A small amount of tungsten-181 (half-life 140 days, which has a characteristic x-ray) from tungsten-180 (0.13% of natural tungsten) will contaminate the tungstens which are considered together as one heat source.

Osmium-194 may be produced by second order irradiation of osmium-192 (41.0% of natural isotopes).

$$Os^{192} + {}_0n^1 \rightarrow Os^{193} + {}_0n^1 \rightarrow Os^{194} + Ir^{194} + \beta^- \;{}^{19h}\; Pt^{194} + \beta^- \text{ (2.24 Mev)}$$

This would be contaminated by osmium-185 (95 d half-life with a characteristic x-ray) from osmium-184 (0.018% of natural osmium). Insufficient data permits an estimate of the specific activity of these last two isotopes.

Thallium-204 with a specific activity of 1.04 watts per gram may be produced by irradiating thallium-203 (29.5% of natural isotopes) in a reactor.

$$Tl^{203} + {}_0n^1 \rightarrow Tl^{204} \rightarrow Pb^{204} + \beta^- \text{ (0.76 Mev)}$$

The melting point of thallium is 302°C and the boiling point is 1457 ± 10°C. Its specific gravity is 11.85. A discussion of experimental thallium irradiation follows this section.

Table 1

HEAT OUTPUT OF ISOTOPES

Isotope as Element or Compound	Half-life	Watts/gram	Watts/cm³	Bare Spherical Heat Source Radius, cm		λ (days⁻¹)	$\lambda \times 365{,}2422$	$e^{-\lambda \times 365.242}$	$1-e^{-\lambda \times 365.2422}$	Watt-hours/year/watt of Isotopes
				200°C	2300°C					
Promethium-146	2 Y 73.0484×10^{1}d	4.06	2.80×10^{1}	2.63×10^{-2}	2.30×10^{1}	9.48887×10^{-4}	3.46574×10^{-1}	0.70710	0.29290	7.40823×10^{3}
Pm O_3 (assumed)	-	1.50	-	-	-	-	-	-	-	-
Promethium-147	2.52 Y 92.041×10^{1}d	5.09×10^{-1}	3.51	2.10×10^{-1}	1.84×10^{1}	7.53085×10^{-4}	2.75058×10^{-1}	0.75952	0.24048	7.66383×10^{3}
Pm O_3 (assumed)	-	1.90×10^{-1}	-	-	-	-	-	-	-	-
Samarium-151	100 Y 36.5242×10^{3}d	4.21×10^{-3}	3.28×10^{-2}	2.24×10^{1}	1.96×10^{4}	1.89777×10^{-5}	6.93146×10^{-3}	0.99309	0.00691	8.73866×10^{3}
Sm_2O_3	-	1.56×10^{-3}	1.16×10^{-2}	6.35×10^{1}	5.56×10^{4}	-	-	-	-	-
Europium-149	-	-	-	-	-	-	-	-	-	-
Eu_2O_3	-	-	-	-	-	-	-	-	-	-
Europium-152	13 Y 47.4815×10^{2}d	7.69×10^{-2}	5.31×10^{-1}	1.39	1.22×10^{3}	1.45983×10^{-4}	5.33192×10^{-2}	0.94807	0.05193	8.53745×10^{3}
Eu_2O_3	-	2.87×10^{-2}	-	-	-	-	-	-	-	-
Europium-154	16 Y 58.4388×10^{2}d	4.75×10^{-1}	3.28	2.25×10^{-1}	1.97	1.18611×10^{-4}	4.33217×10^{-2}	0.95760	0.04240	8.57930×10^{3}
Eu_2O_3	-	1.78×10^{-1}	-	-	-	-	-	-	-	-
Europium-155	1.7 Y 62.0912×10^{1}d	5.38×10^{-1}	3.71	1.98×10^{-2}	1.74×10^{2}	1.11634×10^{-3}	4.07734×10^{-1}	0.66515	0.33485	7.19887×10^{3}
Eu_2O_3	-	2.02×10^{-1}	-	-	-	-	-	-	-	-
Gadolimum-148	35 Y 127.8348×10^{2}	3.23×10^{-1}	2.23	3.30×10^{-2}	2.89×10^{2}	5.42220×10^{-5}	1.98041×10^{-2}	0.98039	0.01961	8.67986×10^{3}
Gd_2O_3	-	1.20×10^{-1}	8.84×10^{-1}	8.33×10^{-1}	7.29×10^{2}	-	-	-	-	-
Thulium-170	129 d	1.35×10^{1}	9.31×10^{1}	7.91×10^{-3}	6.93	5.37323×10^{-3}	1.96253	0.14050	0.85950	3.83903×10^{3}
Tm_2O_3 (assumed)	-	5.21	-	-	-	-	-	-	-	-
Thulium-171	693 d	2.67×10^{-1}	1.84	4.00×10^{-1}	3.50×10^{2}	1.00021×10^{-3}	3.65319×10^{-1}	0.69397	0.30603	7.34319×10^{3}
Tm_2O_3 (assumed)	-	1.03×10^{-1}	-	-	-	-	-	-	-	-
Lutecium-174	165 d	1.29	8.90	8.27×10^{-2}	7.24×10^{1}	4.20089×10^{-3}	1.53434	0.21559	0.78441	4.48139×10^{3}
Lu_2O_3	-	4.99×10^{-1}	-	-	-	-	-	-	-	-
Tungsten-185	74 d	9.72	1.88×10^{2}	3.93×10^{-3}	3.44	9.36685×10^{-3}	3.42117	0.03267	0.96733	2.47852×10^{3}
WO_2	-	7.05	8.54×10^{1}	8.62×10^{-3}	7.55	-	-	-	-	-
Tungsten-188	65 d	-	-	-	-	1.06638×10^{-2}	3.89487	0.02034	0.97966	2.20482×10^{3}
WO_2	-	-	-	-	-	-	-	-	-	-
Osnium-194	V 2 Y $73.04844 \times 10^{+1}$	-	-	-	-	9.48887×10^{-4}	3.46574×10^{-1}	0.7071	0.29290	7.40823×10^{3}
Thallium-204	3 Y 109.573×10^{1}d 19.4Y	1.04	1.23×10^{1}	5.98×10^{-2}	5.23×10^{1}	6.32589×10^{-4}	2.31048×10^{-1}	0.79370	0.20630	7.82688×10^{3}

Lead-210+Bi-210 + Po-210	70.8570×10^{2}d	3.00	3.39×10^{1}	2.17×10^{-2}	1.90×10^{1}	9.78233×10^{-5}	3.57292×10^{-2}	0.96489	0.03511	8.61389×10^{3}
Polonium-208	2.93 Y	3.69×10^{1}	3.47×10^{2}	2.12×10^{-3}	1.86	6.47704×10^{-4}	2.36569×10^{-1}	0.78933	0.21067	7.80615×10^{3}
Polonium-209	100 Y	4.44×10^{-1}	4.17	1.77×10^{-1}	1.55×10^{2}	1.89777×10^{-5}	6.93146×10^{-3}	0.99309	0.00691	8.73866×10^{3}
Polonium-210	138 d	1.43×10^{2}	1.35×10^{3}	5.47×10^{-4}	4.82×10^{-1}	5.02280×10^{-3}	1.83454	0.15969	0.84031	4.01518×10^{3}
Radium-228	6.7 Y 244.712×10^{1d}	-	-	-	-	2.83250×10^{-4}	1.03455×10^{-1}	0.90170	0.09830	8.32904×10^{3}
Actinium-227	21.8 Y 79.6228×10^{2d}	1.45×10^{1}	1.45×10^{2}	5.08×10^{-3}	4.45	8.70538×10^{-5}	3.17957×10^{-2}	0.96871	0.03129	8.62637×10^{3}
Thorium-228	1.91 Y 69.7613×10^{1d}	1.65×10^{2}	1.87×10^{3}	3.94×10^{-4}	3.45×10^{-1}	9.93598×10^{-4}	3.62904×10^{-1}	0.69565	0.30435	7.35145×10^{3}
Uranium-232	74 Y 27.0279×10^{3d}	6.50×10^{-1}	1.21×10^{1}	6.09×10^{-2}	5.33×10^{1}	2.56456×10^{-5}	9.36686×10^{-3}	0.99067	0.00933	8.73132×10^{3}
Plutonium-236	2.7 Y 98.6154×10^{1d}	1.91×10^{1}	3.06×10^{2}	2.41×10^{-3}	2.11	7.02880×10^{-4}	2.56721×10^{-1}	0.77360	0.22640	7.73047×10^{3}
Plutonium-238	89.6 Y 32.7257×10^{3d}	5.45×10^{-1}	8.73	8.44×10^{-2}	7.38×10^{1}	2.11805×10^{-5}	7.73601×10^{-3}	0.99228	0.00772	8.74769×10^{3}
Americium-241	462 Y 168.741×10^{3}	1.05×10^{-1}	1.23	5.99×10^{-1}	5.24×10^{2}	4.10775×10^{-6}	1.50032×10^{-3}	0.99850	0.00150	8.76392×10^{3}
Americium-242	100 Y 36.52422×10^{3d}	5.70×10^{-1}(K)	6.67×10^{-1}(K)	1.10(K)	9.67×10^{2}(K)	1.89777×10^{-5}	6.93146×10^{-3}	0.99309	0.00691	8.73866×10^{3}
Curium-242	162.5 d	1.20×10^{2}	2.04×10^{3}	3.61×10^{-4}	3.16×10^{-1}	4.26552×10^{-3}	1.55795	0.21055	0.78945	4.44185×10^{3}
Curium-243	V 35 Y 127.835×10^{2d}	1.40	2.38×10^{1}	$3.09 \times {}^{-2}$	2.71×10^{1}	5.42220×10^{-5}	1.98042×10^{-2}	0.98039	0.01961	8.67986×10^{3}
Curium-244	18.4 Y 67.2046×10^{2d}	2.73	4.64×10^{1}	1.59×10^{-2}	1.39×10^{1}	1.03140×10^{-4}	3.76711×10^{-2}	0.96303	0.03697	8.60266×10^{3}
Californium, 248	250 d	8.08×10^{1}	1.29×10^{3}	5.71×10^{-4}	5.00×10^{-1}	2.77259×10^{-3}	1.01263	0.36327	0.63673	5.51164×10^{3}
Californium-250	10 Y 36.5242×10^{2d}	5.10	8.16×10^{1}	9.02×10^{-3}	7.90	1.89777×10^{-4}	6.93146×10^{-2}	0.93303	0.06697	8.46929×10^{3}
Californium-252	2.2 Y 80.3533×10^{1d}	2.33×10^{1}	3.73×10^{2}	1.97×10^{-3}	1.73	8.6516×10^{-4}	3.16156×10^{-1}	0.72895	0.27105	7.51511×10^{3}
Einsteinium-252	140 d	1.50×10^{2}	2.40×10^{3}	3.07×10^{-4}	2.69×10^{-1}	4.95105×10^{-3}	1.80833	0.16393	0.83607	4.05281×10^{3}
Einstenium-254	300 d	6.88×10^{1}	1.10×10^{3}	6.69×10^{-4}	5.86×10^{-1}	2.31049×10^{-3}	8.43888×10^{-1}	0.43003	0.56997	5.92051×10^{3}

$$\text{Watt} = \frac{24\ W}{T\ \text{days}}\ 1 - e^{-\lambda \times 365\ \text{watt-hours}}$$

$$\lambda = \frac{0.693147}{T\ ½}$$

where W_o = Total initial watts output of source

M = Multiplier to watt-hours per years

Lead-210 is radium D of the uranium-238 decay chain, and can be milked from radium-226. However, the amounts produced in this way are limited by the moderately long (1638 yr.) half-life and relative scarcity of radium. Lead-210 does have the advantage of having short-lived daughters, (one β emitter, and one α emitter) which are quite energetic, and the chain has a total specific activity at equilibrium of 3.00 watts per gram.

There is no feasible production scheme for lead-210. Its melting point is 327.43°C and its boiling point is 1620°C. It has a half-life of 19.4 years and decays in the following manner:

$$\mathrm{Pb}^{210} \xrightarrow[19.4y]{\beta^-} \mathrm{Bi}^{210} \xrightarrow[5\mathrm{d}]{\beta^-} \mathrm{Po}^{210} \xrightarrow[138\mathrm{d}]{\alpha} \mathrm{Pb}^{206} \text{ (Stable)}$$

Because the half-life of the daughters, compared with that of the lead, is very short, the energy output would increase for the first year and then follow the lead decay rate. Lead-210 is always found grossly contaminated with stable lead-206, the end product of the uranium family. If it were not separated, the mixture would have a correspondingly lower specific activity.

Polonium-208 is the daughter of radon-212 and astatine-208. It has been made in very small amounts in the following ways:

$$\mathrm{Pb}^{206}\ (\alpha,\ 2n)\ \mathrm{Po}^{208}$$
$$\mathrm{Pb}^{207}\ (\alpha,\ 3n)\ \mathrm{Po}^{208}$$
$$\mathrm{Bi}^{209}\ (d,\ 3n)\ \mathrm{Po}^{208}$$
$$\mathrm{Bi}^{209}\ (p,\ 2n)\ \mathrm{Po}^{208}$$

All of these methods require accelerators and are quite selective in their particle energy requirement. Competing reactions occur and are generally favored. Polonium-208 has a half-life of 2.93 years and decays in the following manner:

$$\mathrm{Po}^{208} \xrightarrow[2.93Y]{\alpha} \mathrm{Po}^{204} \text{ (Stable)}$$

Polonium-208 has a specific energy for use as a heat source of 36.9 watts per gram. It has the additional advantages of being both gamma and beta-free and of having a single, stable daughter.

Polonium-209 has been prepared by the particle bombardment of bismuth-209.

$$\mathrm{Bi}^{209}\ (d,\ 2n)\ \mathrm{Po}^{209}$$
$$\mathrm{Bi}^{209}\ (p,\ n\ )\ \mathrm{Po}^{209}$$

Polonium-209 has a specific energy for use as a heat source of 0.444 watt per gram. It decays with a half-life of nearly 100 years to lead-205 which is unstable but which has a 10^5-year half-life. A small amount of gamma radiation is associated with this polonium isotope.

Polonium-210 is the daughter of bismuth-210 and astatine-210. At present, the methods of production of polonium-210 are either to separate the isotope from radium residues, (i.e. milk from lead-210) or prepare by irradiation of bismuth-209.

$$\mathrm{Bi}^{209}\ (n,\ \gamma)\ \mathrm{Bi}^{210} \xrightarrow[5\mathrm{d}]{\beta^-} \mathrm{Po}^{210}$$

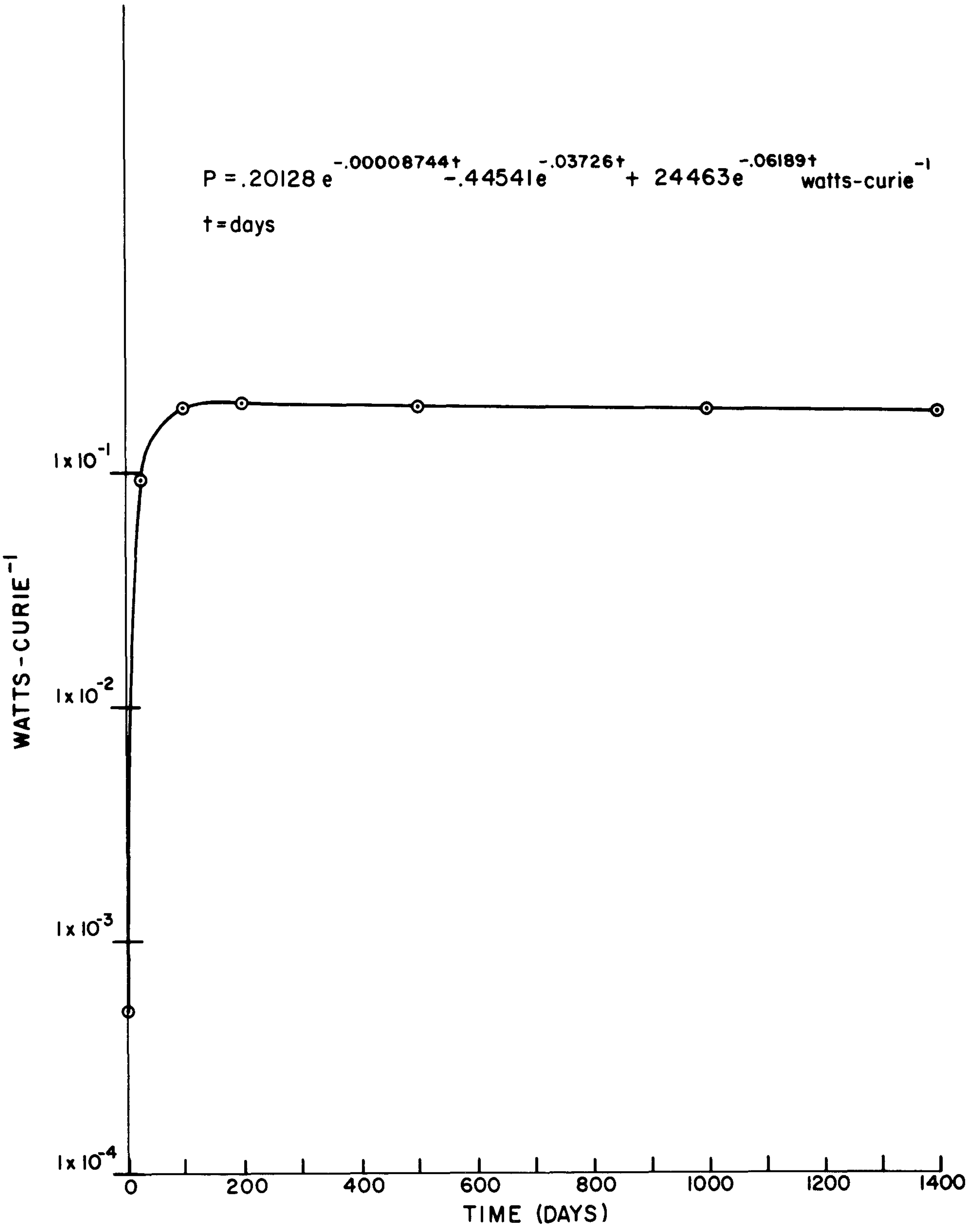

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FIGURE 1 - WATTS PER CURIE OF ACTINIUM-227

Polonium-210 has a specific energy for use as a heat source of 143 watts per gram. It is relatively gamma-free, emitting one gamma per 10^5 alphas. It decays with a half-life of 138 days to lead-206. It can be obtained in moderately large quantities at the present time by the process of bismuth irradiation. Other methods of preparation are:

$$Bi^{209}\ (d,\ n)\ Po^{210}$$
$$Bi^{209}\ (p,\ \gamma)\ Po^{210}$$

The short half-life gives it a large specific activity, but generators powered by this isotope would have a rapidly diminishing output.

Radium-228 is the natural daughter of thorium-232 and of actinium-228. It has a half-life of 6.7 years and decays in the following manner:

$$Ra^{228} \xrightarrow[6.7Y]{\beta^-} Ac^{228} \xrightarrow[6.13h]{\beta^-} Th^{228} \xrightarrow[1.91Y]{\alpha\text{'s}} \text{Thorium Series}$$

Thorium Series:

$$\begin{tikzcd}
Th^{228} \arrow[r, "\alpha\ (5.4\ \text{Mev})", "1.9\ y"'] & Ra^{224} \arrow[r, "\alpha\ (5.6\ \text{Mev})", "3.64\ d"'] & Em^{220} \arrow[r, "\alpha\ (6.3\ \text{Mev})", "5.4\ s"'] & Po^{216} \arrow[d, "\alpha\ 6.8\ \text{Mev}"', "0.158\ s"] \\
Tl^{208} \arrow[d, "\beta^-\ 1.8\ \text{Mev}"', "3.1\ m"] & Bi^{212} \arrow[l, "33\%\ \alpha\ (6.0\ \text{Mev})"', "60\ m"] \arrow[d, "\beta^-\ 2.25\ \text{Mev}\ 67\%"', "60.5\ m"] & Pb^{212} \arrow[l, "\beta^-\ (0.33\ \text{Mev})"', "10\ h"] & \\
Pb^{208} & Po^{212} \arrow[l, "\alpha\ (8.8\ \text{Mev})"', "0.3\ \mu s"] & &
\end{tikzcd}$$

No method has been used successfully to produce radium-228 artificially. This would be a highly desirable element from the standpoint of specific activity because of its moderately short half-life and its active daughters, but somewhat undesirable because of the strong gamma from thallium-208.

Actinium-227, the daughter of protactinium-231 and of radium-227 can be produced by neutron irradiation of radium-226:

$$Ra^{226}\ (n,\ \gamma)\ Ra^{227} \xrightarrow[41\ m]{\beta^-} Ac^{227} \xrightarrow[21\ y]{\beta^-} Th^{227} \xrightarrow[18.4\ d]{\alpha} Ra^{223} \xrightarrow[11.6\ d]{\alpha} Em^{219} \xrightarrow[3.92\ s]{\alpha}$$

$$Em^{219} \longrightarrow Po^{215} \xrightarrow[1.8\ ms]{\alpha} Po^{211} \xrightarrow[36.1\ m]{\beta^-(1.4\ \text{Mev})} Bi^{211} \xrightarrow[2.16\ m]{\alpha(6.61\ \text{Mev})} Tl^{207} \xrightarrow[4.78\ m]{\beta^-(1.47\ \text{Mev})} Pb^{207}$$

The actinium decays through a chain of short-lived alpha emitters, most of which emit a moderate amount of gamma radiation as well. Actinium has a half-life of 21.8 years, but has a much higher specific activity because of its decay products.

The following equation can be used to calculate the watts per curie. It includes ⅓ of the gamma energy and the value of t is expressed in days.

$$\text{Watts/curie} = 0.20156e^{-0.00008744t} - 0.44541e^{-0.03726t} + 0.024463e^{-0.06189t}$$

See Figure 1

Thorium-228 is the daughter of actinium-228, uranium-232, and also protactinium-228. Thorium-228 can also be made by the secondary irradiation of radium-226.

$$\begin{tikzcd}
Ra^{226}(n, \gamma)\ Ra^{227} \arrow[r, "\beta^-", "41\ m"'] & Ac^{227} \arrow[r] \arrow[d, "(n, \gamma)"] & \text{Chain of short-lived alphas.} \\
 & Ac^{228} \arrow[r, "\beta", "6.13\ h"'] & Th^{228} \arrow[r, "1.91\ y"'] & \text{Chain of short lived alphas which are shown under the paragraph on radium-228.}
\end{tikzcd}$$

The power output from thorium-228 can be calculated from the following equation where t is in days.

$$\text{Watts/curie} = 0.20156e^{-0.0009988t} - 0.17655e^{-0.19043t} + 0.0076e^{-1.5694t}$$

The combination of actinium-227 and thorium-228 holds some promise provided high-level gamma radiation is not objectionable. Measurements, calculations and engineering data indicate that up to 25 watts output per curie of starting radium are obtainable for an irradiation time of one year at a neutron (nv) flux of 5×10^{14}. The radium should be sealed in a container suitable for both the irradiation and the final heat source flux levels given. The main disadvantage of this system is the strong gamma radiation emitted by the daughter thallium-208. One advantage of this system is that the heat source can be reactivated by reirradiating in a reactor.

Uranium-232 is the daughter of protactinium-232, of plutonium-232, and plutonium-236. It has also been prepared by:

$$Pa^{231}\ (d, n)\ U^{232}$$
$$Pa^{231}\ (\alpha, \text{-}p2n)\ U^{232}$$
$$Th^{232}\ (\alpha, 4n)\ U^{232}$$

If thorium is used as a blanket about a fast neutron reactor, a small amount of the thorium will be converted to uranium-232 as well as to the more common uranium-233. If these uranium isotopes could be separated, an excellent source for uranium-232 would be available. Uranium-232 has a half-life of 74 years, losing an alpha particle to become thorium-228, which is also useful as a heat source, as discussed above. It would have a specific activity of 0.62 watt per gram without counting the additional energy of the thorium-228 daughter series.

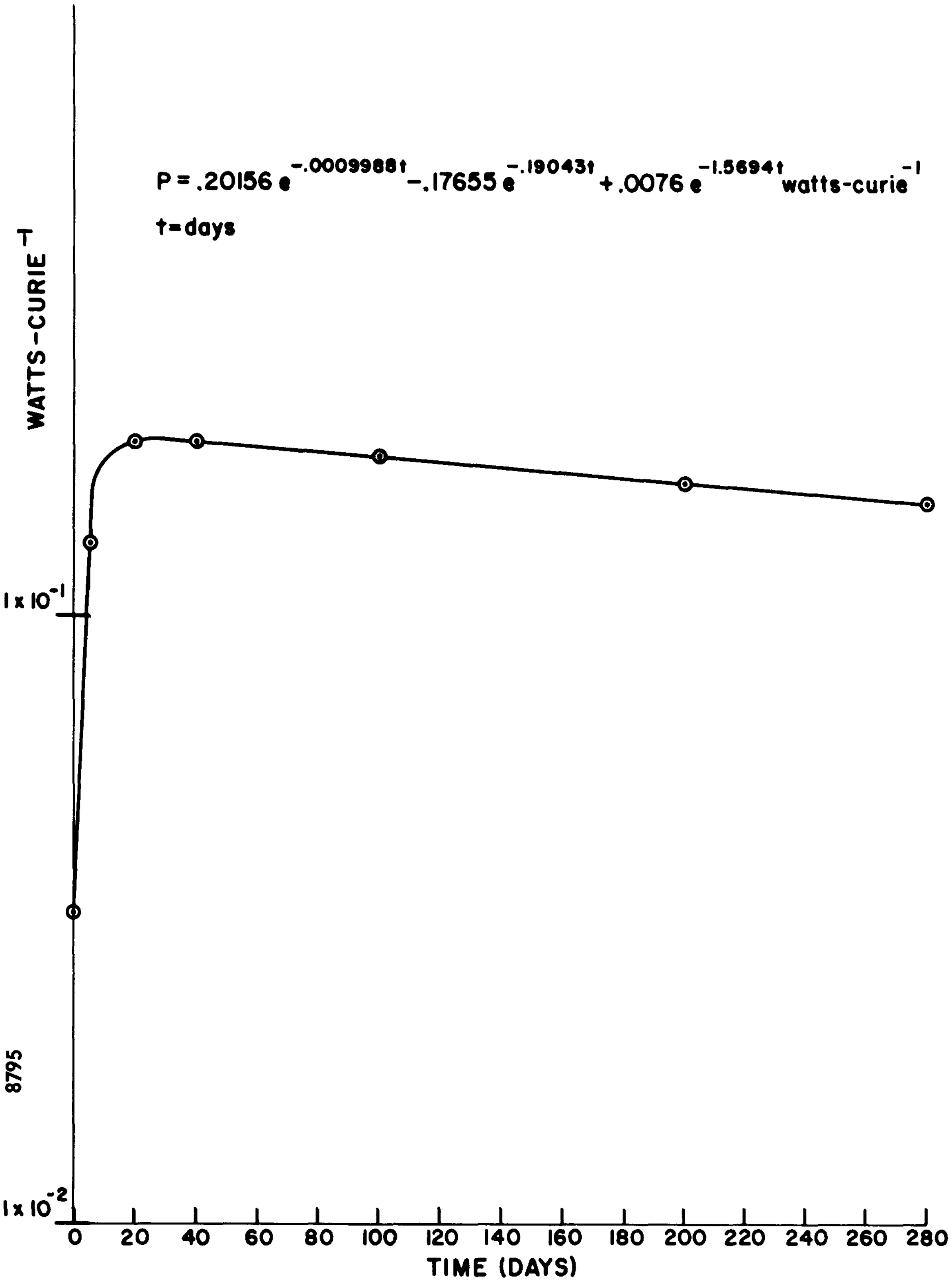


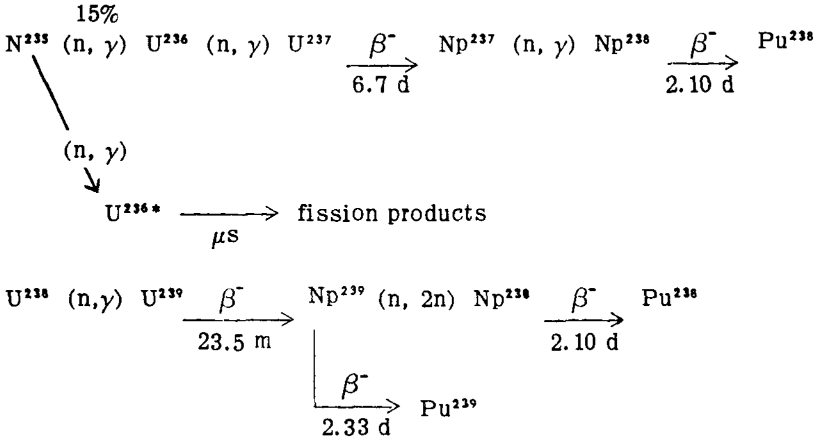
FIGURE 2 - WATTS PER CURIE OF THORIUM-228

Plutonium-236 is the daughter of curium-240 and of neptunium-236. It has also been prepared in the following manner:

U^{235} $(\alpha, 3n)$ Pu^{236}
U^{238} $(\alpha, 6n)$ Pu^{236}
Np^{237} $(d, 3n)$ Pu^{236}
U^{233} (α, n) Pu^{236}

Plutonium-236 decays by alpha emission to uranium-232 with a half-life of 2.7 years, and the heat provided by daughters down to lead-204 should be included in its discussion.

Plutonium-238 is the daughter of curium-242 and of neptunium-238. It could also be made as a reactor fuel by-product as shown below:



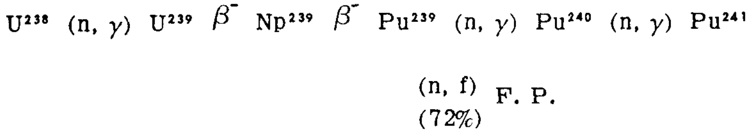
It has also been produced by:

U^{238} $(d, 2n)$ Pu^{238}
U^{238} $(\alpha, 4n)$ Pu^{238}
U^{235} (α, n) Pu^{238}

While yields for such an involved chain would be very small, the large amount of enriched uranium fuel used in reactors makes this scheme appear feasible. Separations might be made at several points. Isotopic separations of the uranium-236 from uranium-235 residues could be made by barrier or thermal diffusion. Neptunium-237 can be separated from plutonium and uranium and used as a source of material for further irradiation.

Plutonium-238 decays with a half-life of 89.6 years to uranium-234, a nearly stable isotope (${}^{t}½ = 2.5 \times 10^{5}$ years). It has a moderate amount of 17 and 44 Kev gamma rays associated with its alpha, and a specific activity of 0.545 watt per gram.

Plutonium-241 is found as a by-product of plutonium-239 production.



It is also formed as.

$$U^{238}\ (n, \gamma)\ U^{239}\ (n, \gamma)\ U^{240} \xrightarrow{\beta^-} Pu^{240}\ (n, \gamma)\ Pu^{241}$$

and

$$U^{238}\ (\alpha, n)\ Pu^{241}$$

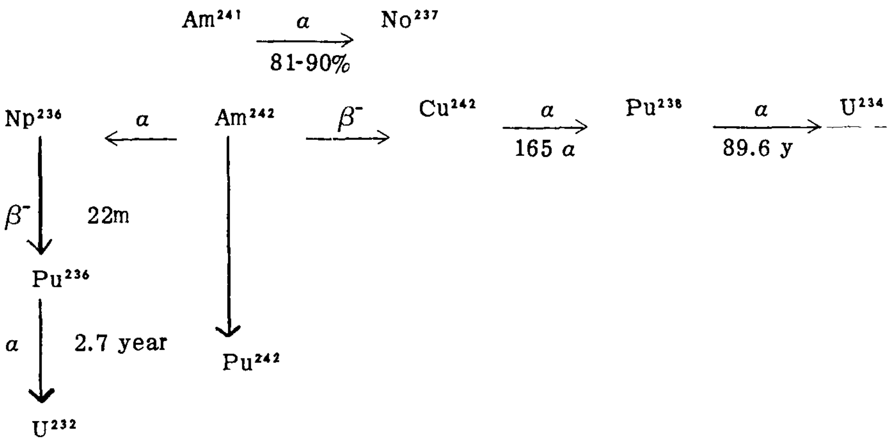
To be useful, plutonium-241 would have to be separated from isotopes of plutonium which are present in much larger amounts. Plutonium-241 decays in two ways-mainly, through the 13.2 year half-life β^- decay.

$$Np^{237} \xleftarrow[675]{\beta^-} U^{237} \xleftarrow{\overset{10^{-3}}{\alpha}} Pu^{241} \xrightarrow[13.2]{\overset{99+\%}{\beta^-}} Am^{241} \xrightarrow[470\ y]{\alpha} Np^{237}$$

it has a low power output, as the beta particle has a maximum energy of 20 Kev, and the moderately long-lived americium-241 would take time to grow in.

Americium-241 with a half-life of 462 days is the decay product of plutonium-241 discussed above, and is another by-product of plutonium-239 production. In any production scheme it would come mixed with the isotope americium-242. Since it can be separated from plutonium isotopes chemically, it shows some promise of availability in the next few years, particularly is plutonium fueled reactors become engineering realities.

The americiums have the following decay scheme:



Because of the moderately long half-life of the americium-241 and metastable americium-242 (100 years) the specific activity is low for consideration as a heat source. Also the large amount of varied gamma connected with the various daughters makes it difficult to handle. However, its future availability and its long half-life make it have an almost constant wattage output.

Americium-242 is also produced as a by-product of plutonium-239 manufacture.

$$Pu^{239}\ (n,\gamma)\ Pu^{240}\ (n, \gamma)\ Pu^{241}\ \underset{13.2}{\beta^-}\ Am^{241}\ (n, \gamma)\ Am^{242}$$

Its two isomers, one with a 16-hour half-life and the other with a 100-year half-life, are discussed with americium-241.

Curium-242 is the daughter of americium-242 and, therefore, is also a by-product of the plutonium-239 manufacturing chain. Because of the long half-life of americium-242, its large thermal neutron cross section (700 barns), and the fairly large amounts of plutonium made, there is a remote possibility of obtaining this isotope. It can also be prepared by:

$$\mathrm{Pu}^{239}\ (\alpha,\ \mathrm{n})\ \mathrm{Cu}^{242}$$
$$\mathrm{Am}^{241}\ (\mathrm{d},\ \mathrm{n})\ \mathrm{Cu}^{242}$$

It is also the decay product of californium-246.

Curium-242 has a half-life of 162.5 days and decays by alpha emission to plutonium-238. Twenty-six per cent of the alphas are associated with 44 Kev gammas, and there are other gammas present. Because of its short half-life and energetic alpha, it has a high specific activity of 120 watts per gram.

Curium-243 with a half-life of 35 years is the irradiation product of curium-242.

$$\mathrm{Cm}^{242}\ (\mathrm{n},\ \gamma)\ \mathrm{Cm}^{243}$$

It will normally be a contaminant in curium-242. It is also the daughter of berkelium-243.

$$\mathrm{Bk}^{243}\ \xrightarrow[4.5\ \mathrm{h}]{\beta^+}\ \mathrm{Cm}^{243}$$

The decay of curium-243 is½

$$\mathrm{Cm}^{243}\ \xrightarrow[93\%\ \gamma > 0.27\ \mathrm{Kev}]{\alpha}\ \mathrm{Pu}^{239}$$

Curium-244 is the irradiation product of curium-243. Although the thermal neutron cross section of curium-243 has not been measured, it has been estimated as being large with the capture cross section one-half that of the fission cross section. This isotope will also be a contaminant in curium-242.

$$\mathrm{Cm}^{243}\ (\mathrm{n},\ \gamma)\ \mathrm{Cm}^{244}$$

It is also the daughter of americium-244.

$$\mathrm{Am}^{244}\ \xrightarrow[26\ \eta]{\beta^-}\ \mathrm{Cm}^{244}$$

The presence of these higher isotopes of curium considerably increase the gamma of the curium-242.

The californium isotopes 248, 250, and 252 as well as the *einsteinium isotopes 252 and 254* have promising nuclear properties. However, at present the only production method is bombardment at exceedingly high energy with heavy particles such as carbon or nitrogen. Less than microgram amounts of these materials have been prepared to date.

Curves for the formation of some of these isotopes in the reactor are shown in Figure 3 through 19. Derivation of formulas used to calculate the growth curves is given in MLM CF 57-4-34 appendix A and B.[5] Curves showing the power growth of daughters after removal from the reactor is shown in Figures 1 and 2 for actinium-227 and thorium-228. Decay of power for the others with no or a single short-lived daughter generally follows that shown in Figure 20 with a suitable change in time scale to suit the dominant half-life.

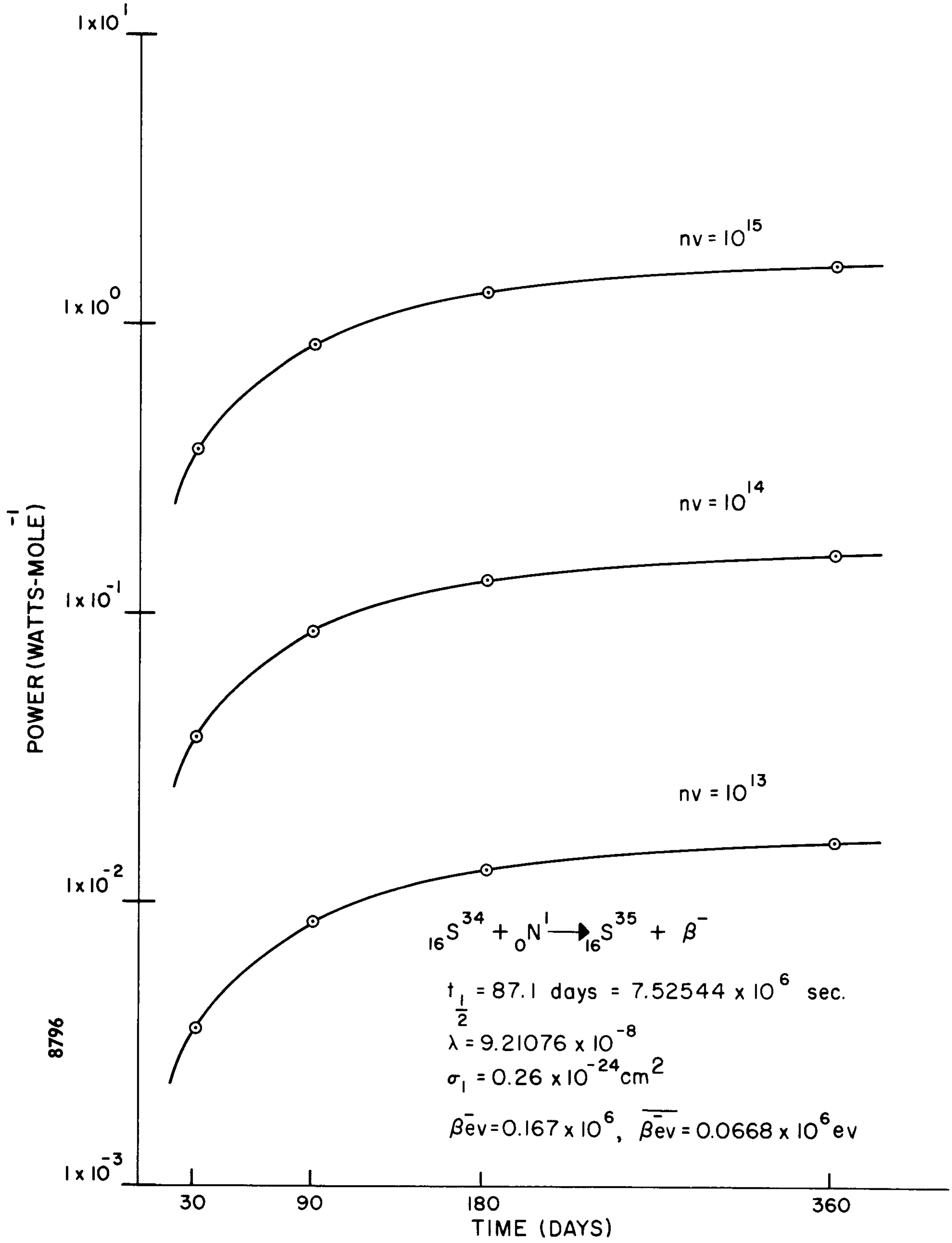


FIGURE 3 - FORMATION OF SULFUR-35

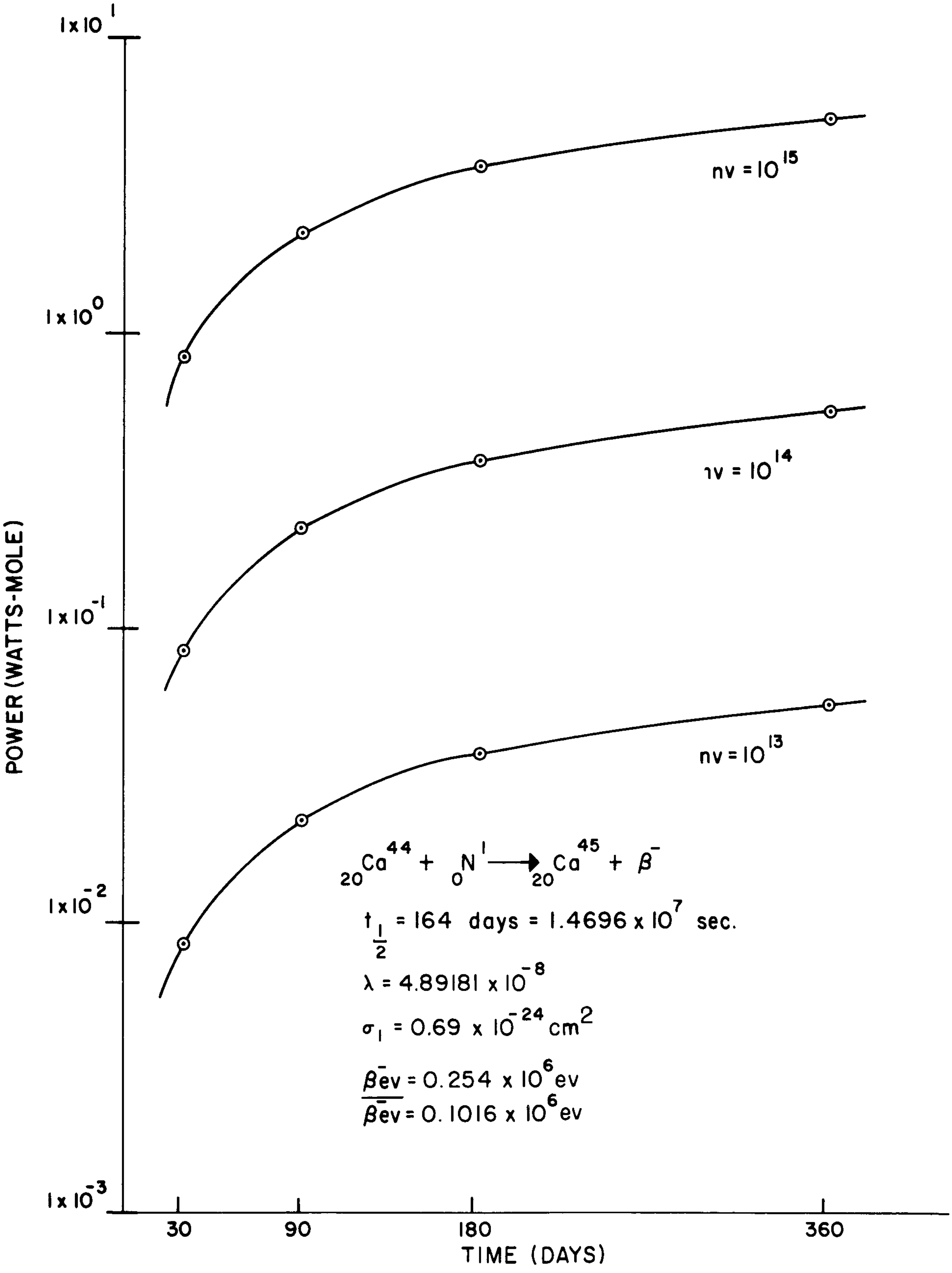


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FIGURE 4 - FORMATION OF CALCIUM-45

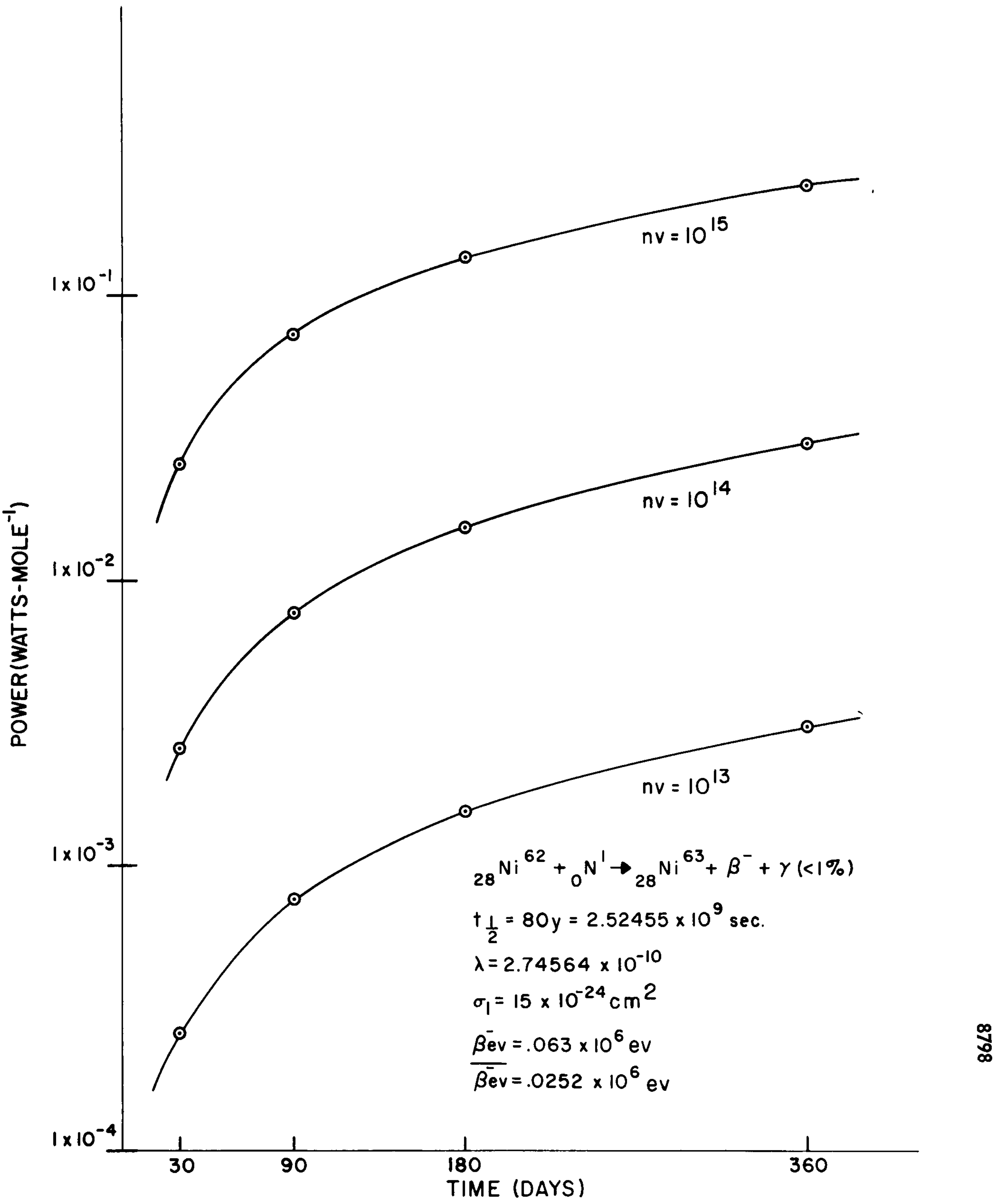

FIGURE 5 - FORMATION OF NICKEL-63

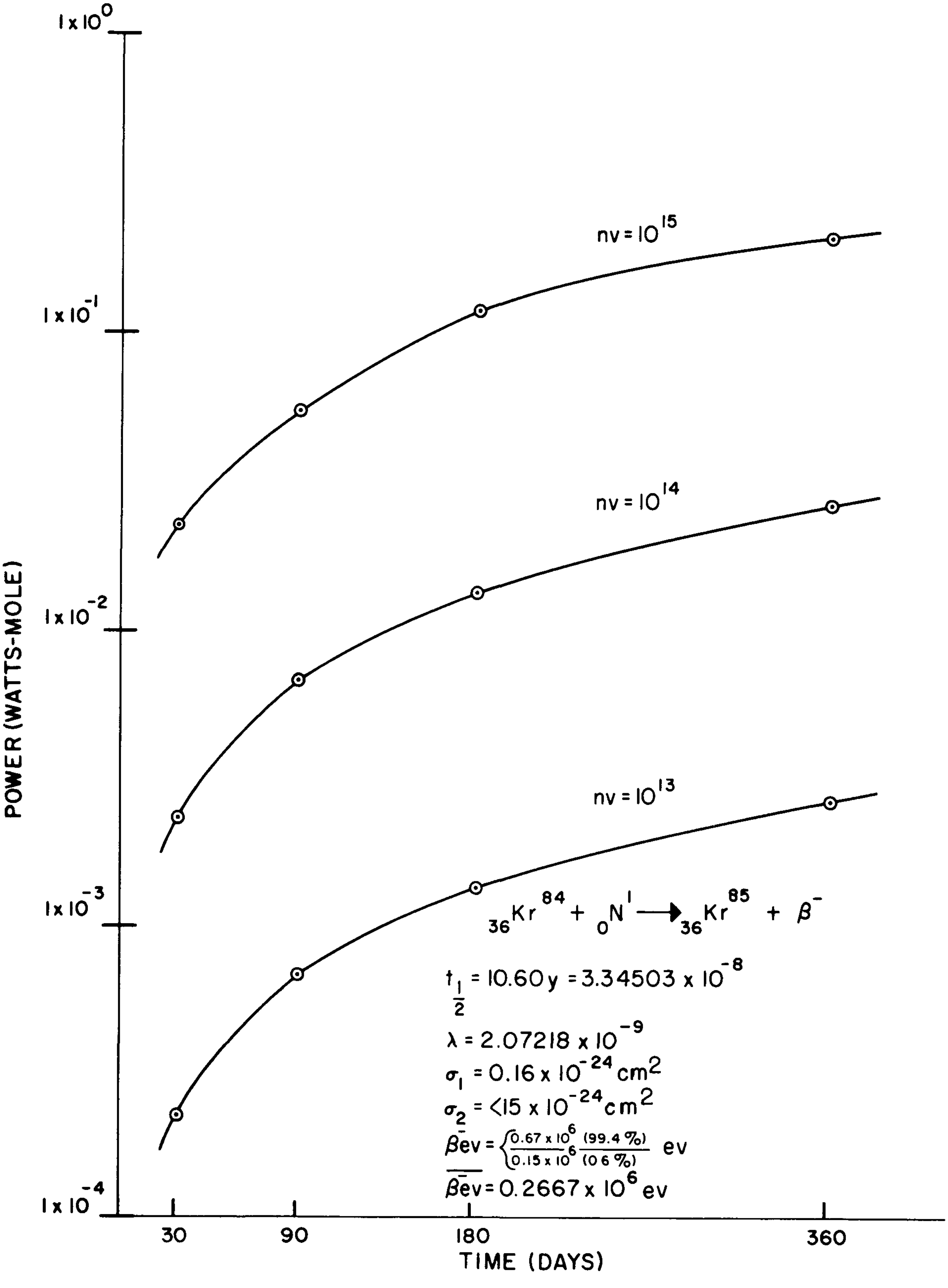

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FIGURE 6 - FORMATION OF KRYPTON-85

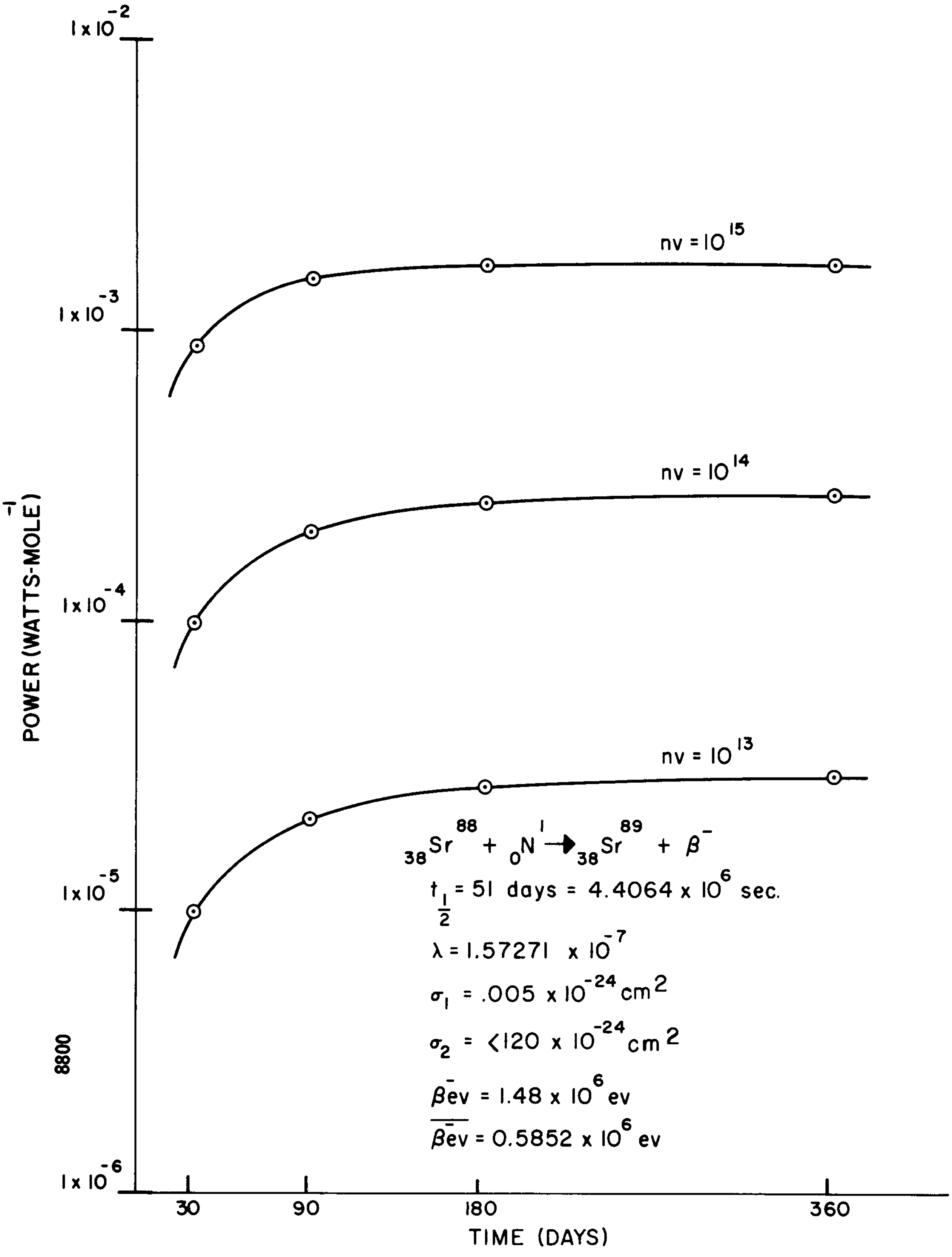
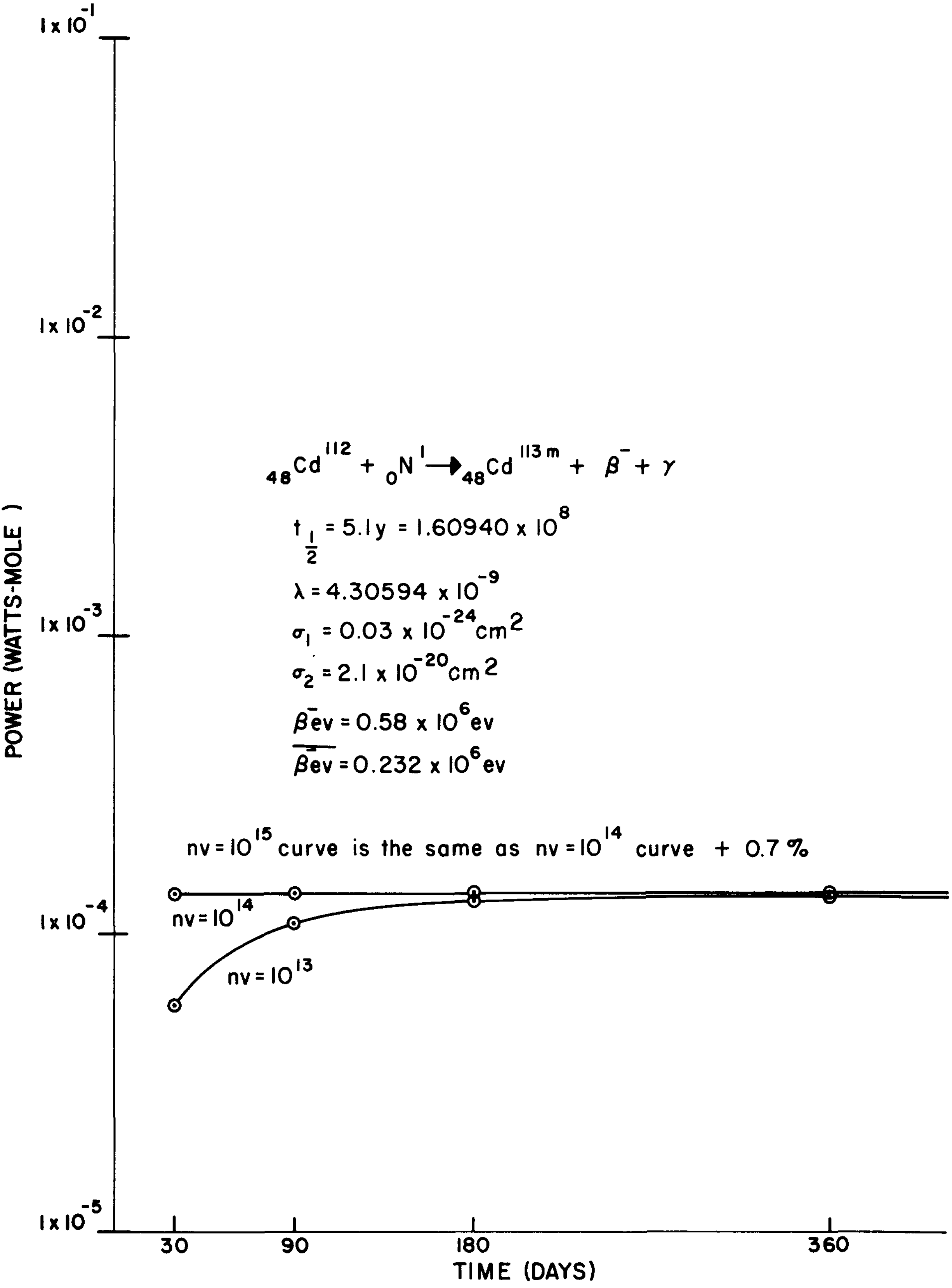


FIGURE 7 - FORMATION OF STRONTIUM-89



$_{48}Cd^{112} + _{0}N^{1} \rightarrow _{48}Cd^{113m} + \beta^{-} + \gamma$

$t_{\frac{1}{2}} = 5.1y = 1.60940 \times 10^{8}$

$\lambda = 4.30594 \times 10^{-9}$

$\sigma_1 = 0.03 \times 10^{-24} cm^2$

$\sigma_2 = 2.1 \times 10^{-20} cm^2$

$\beta^{-}ev = 0.58 \times 10^{6} ev$

$\overline{\beta ev} = 0.232 \times 10^{6} ev$

nv = 10^{15} curve is the same as nv = 10^{14} curve + 0.7 %

FIGURE 8 - FORMATION OF CADMIUM-113M

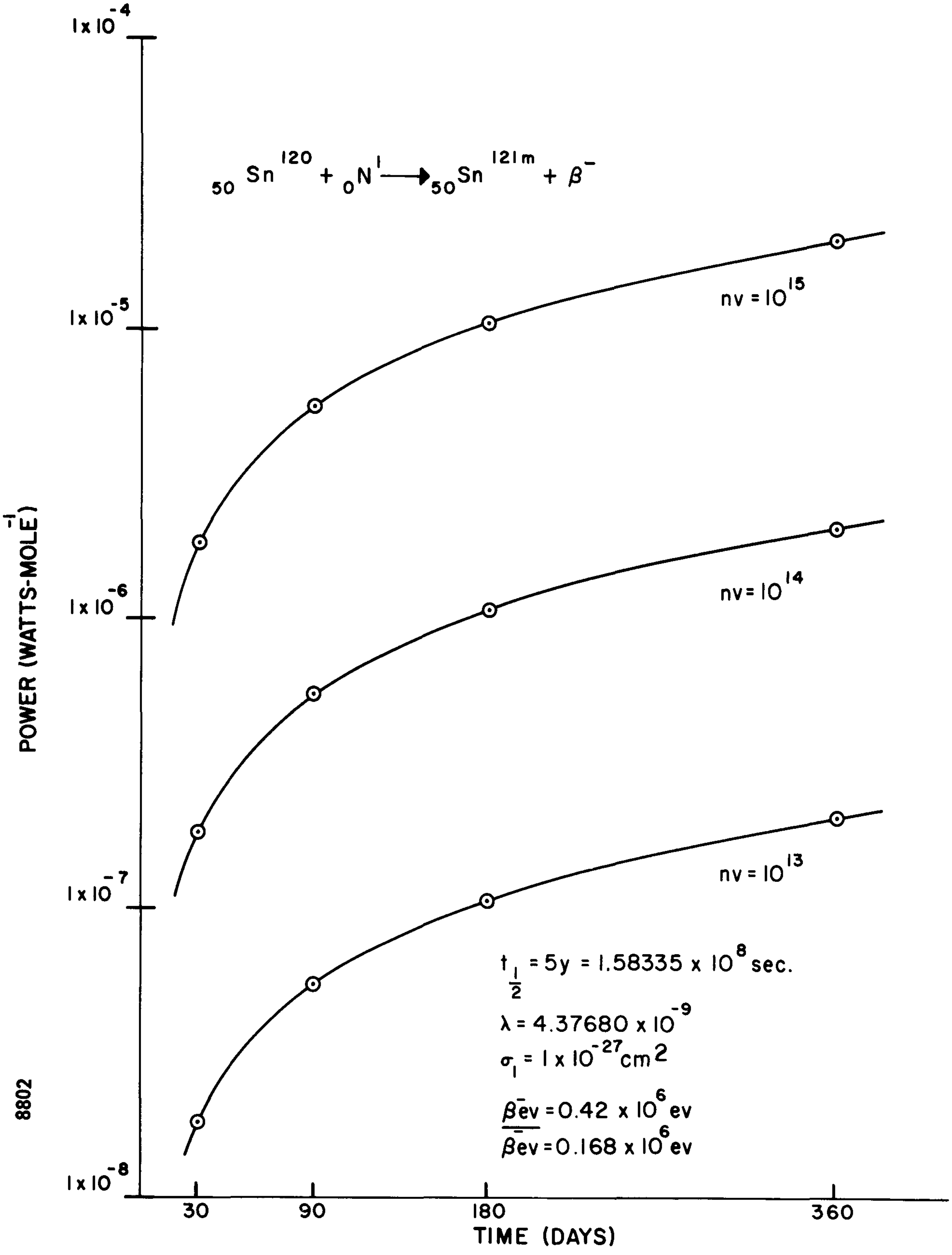


FIGURE 9 - FORMATION OF TIN-121M

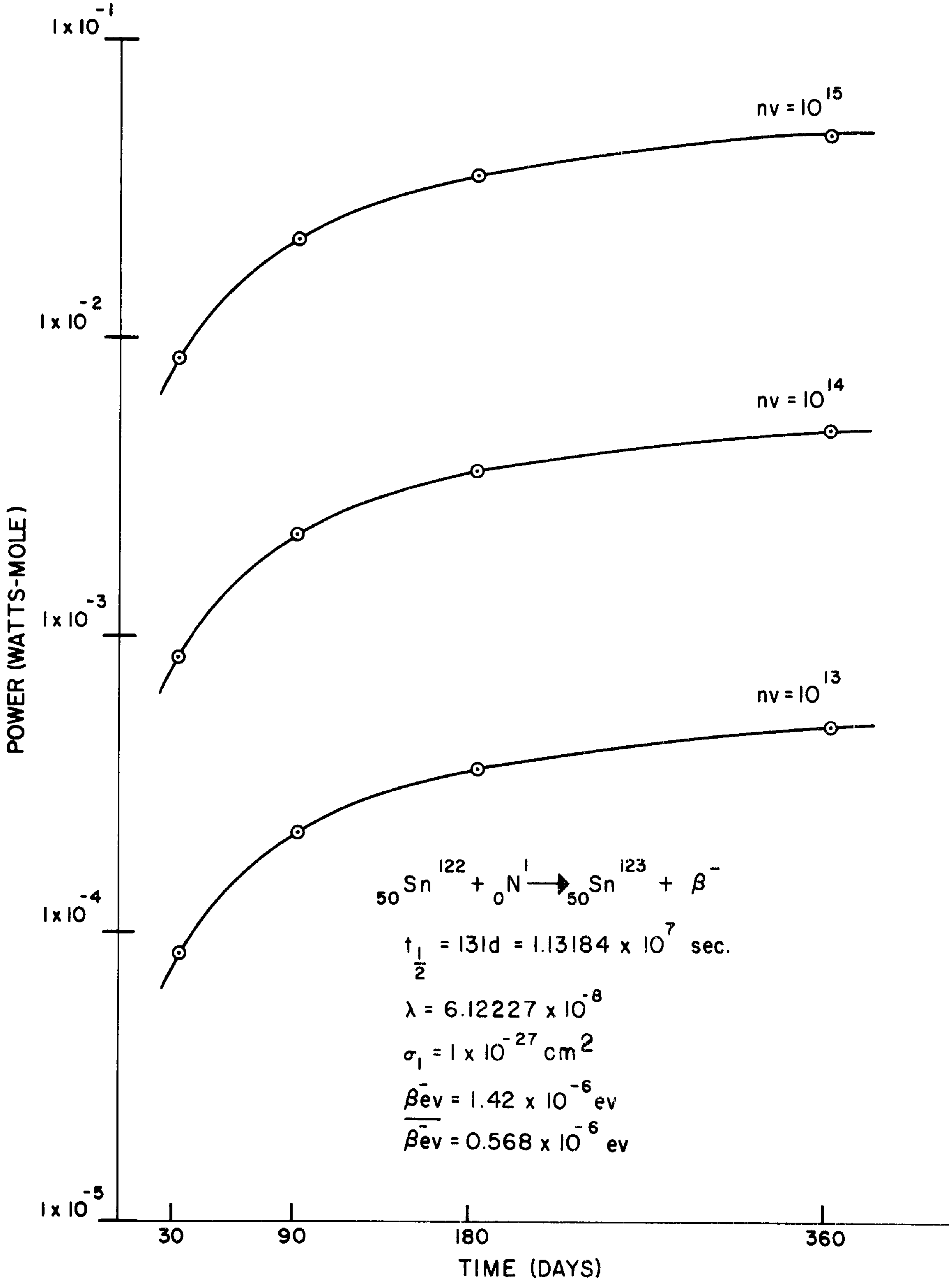


FIGURE 10 - FORMATION OF TIN-123

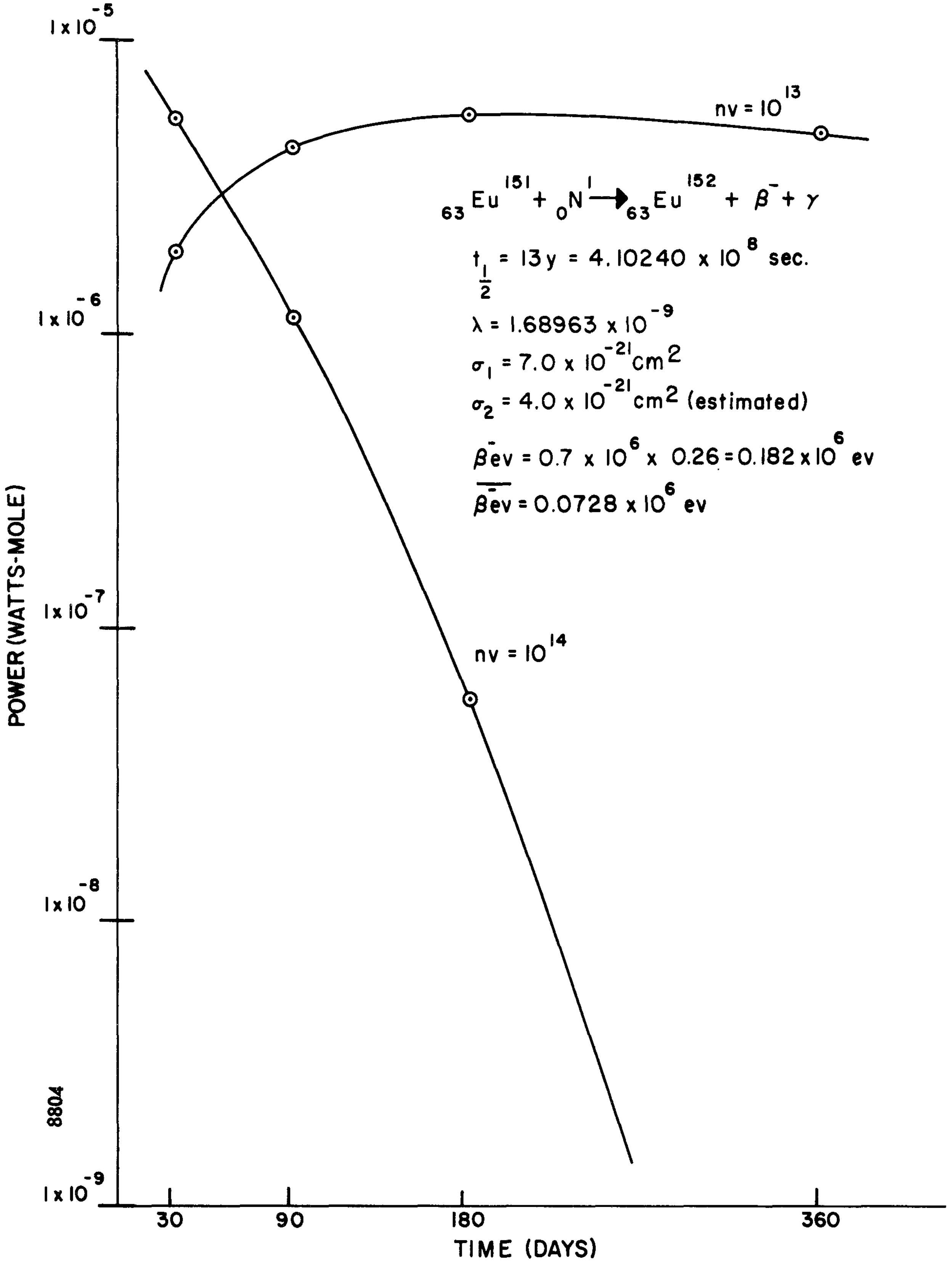


FIGURE 11 - FORMATION OF EUROPIUM-152

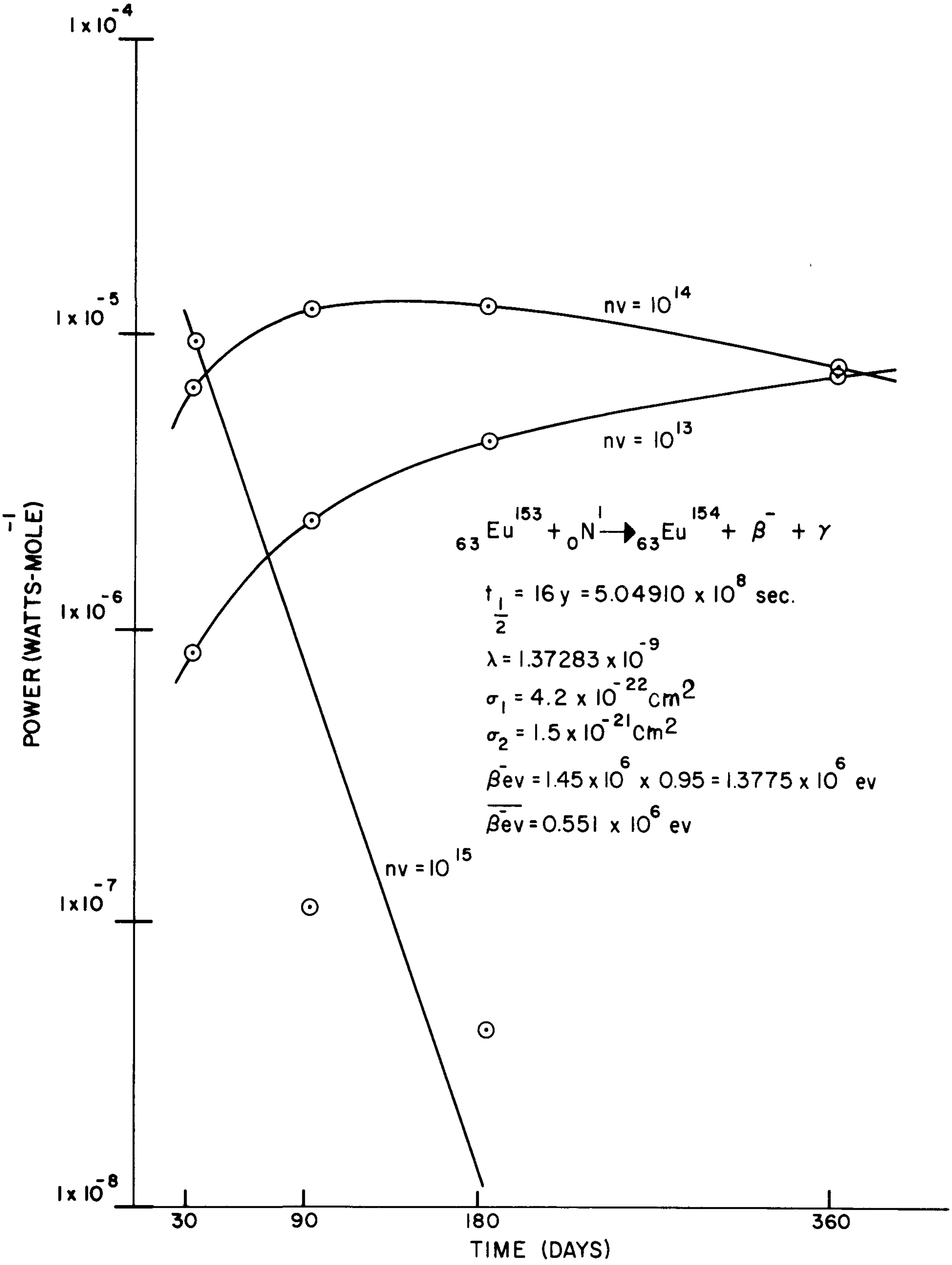
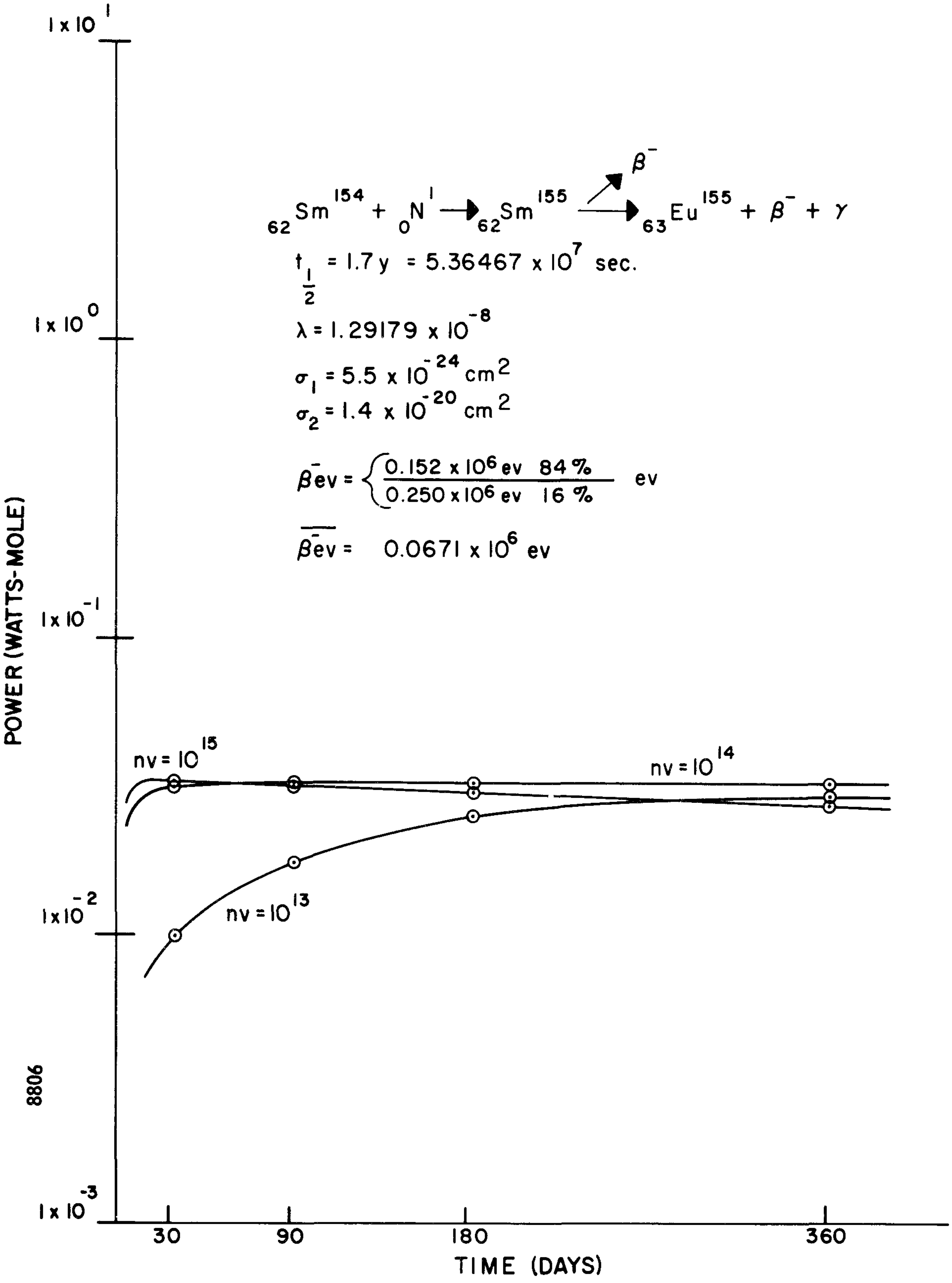


FIGURE 12 - FORMATION OF EUROPIUM-154



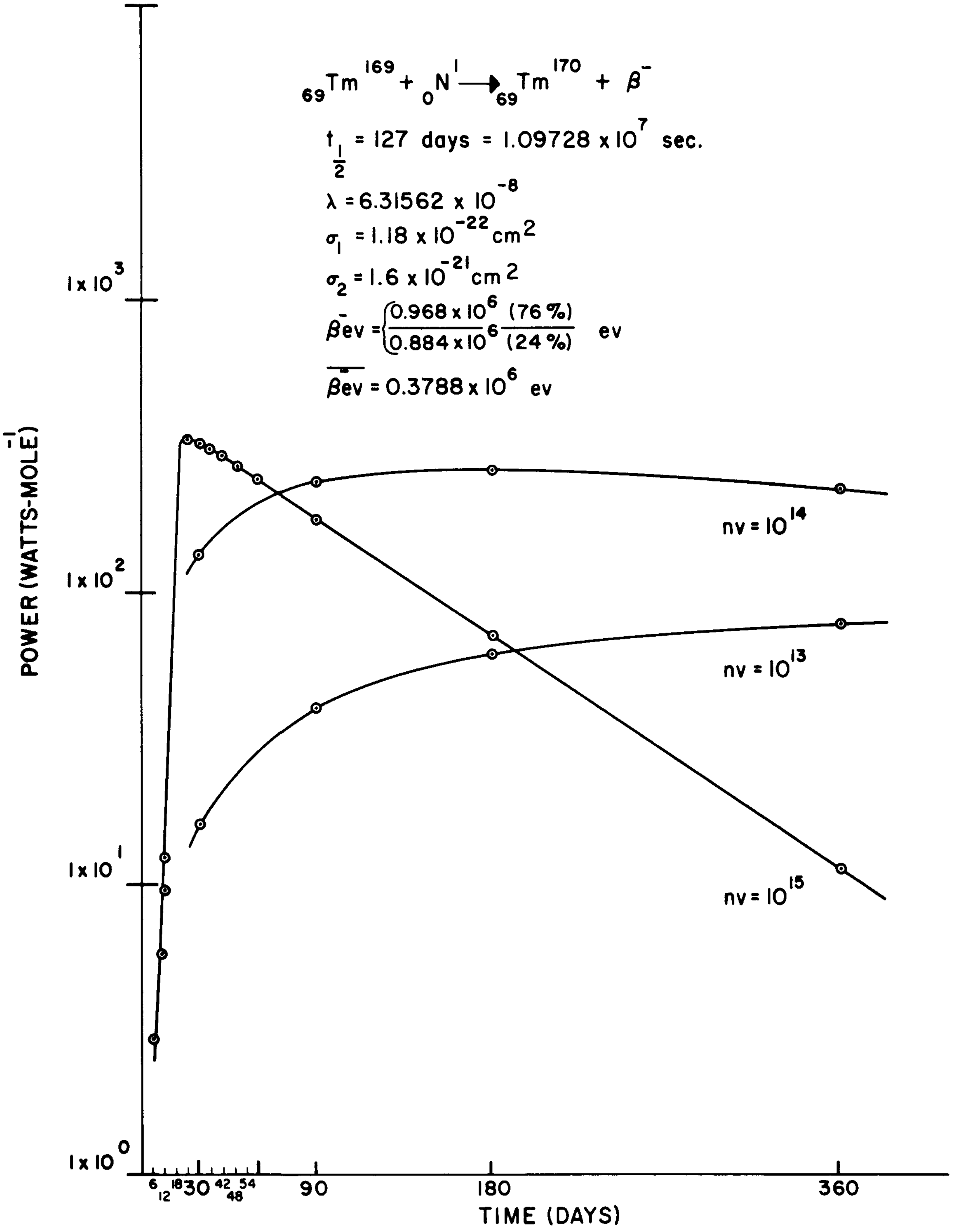


FIGURE 14 - FORMATION OF THULIUM-170

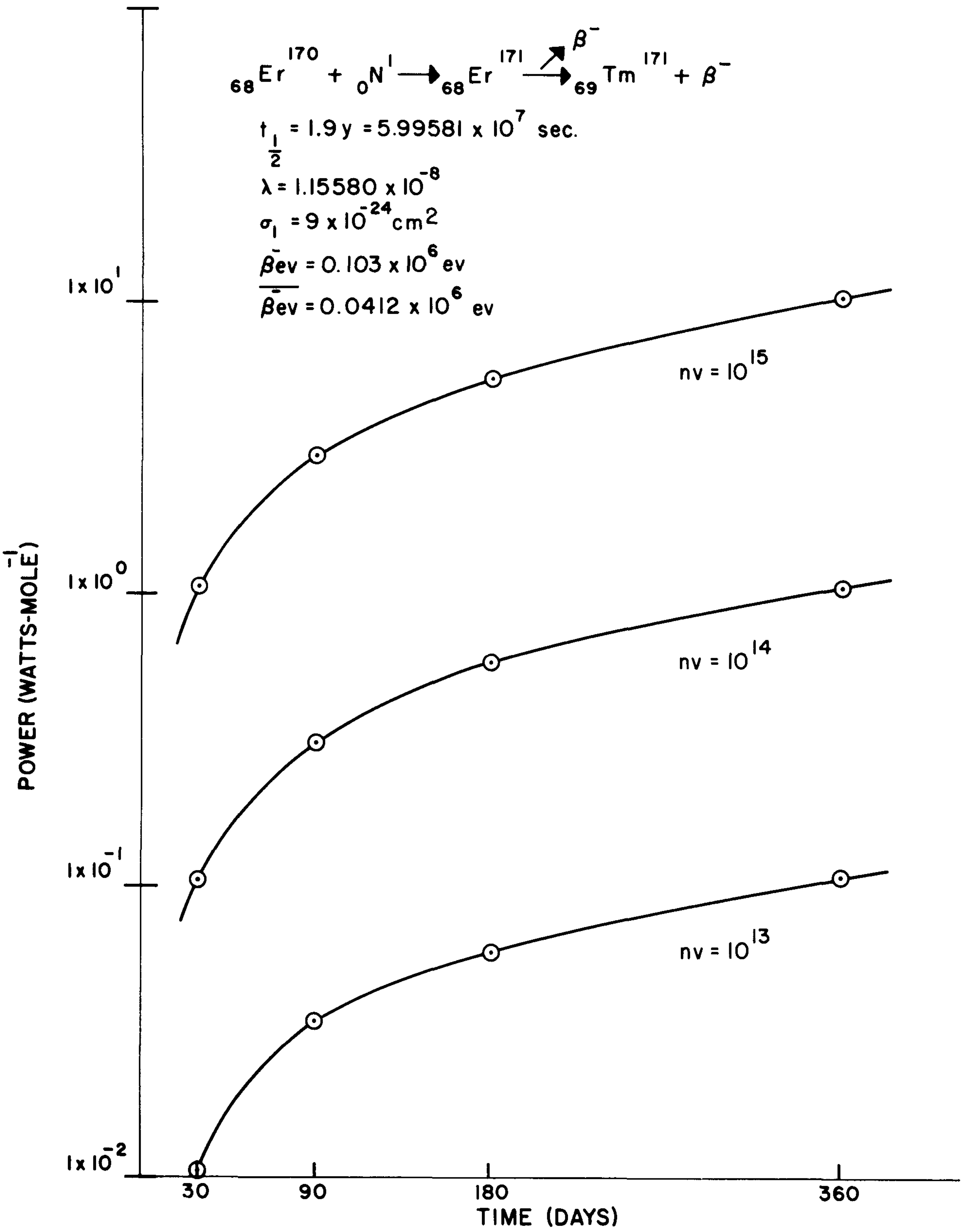


FIGURE 15 - FORMATION OF THULIUM-171

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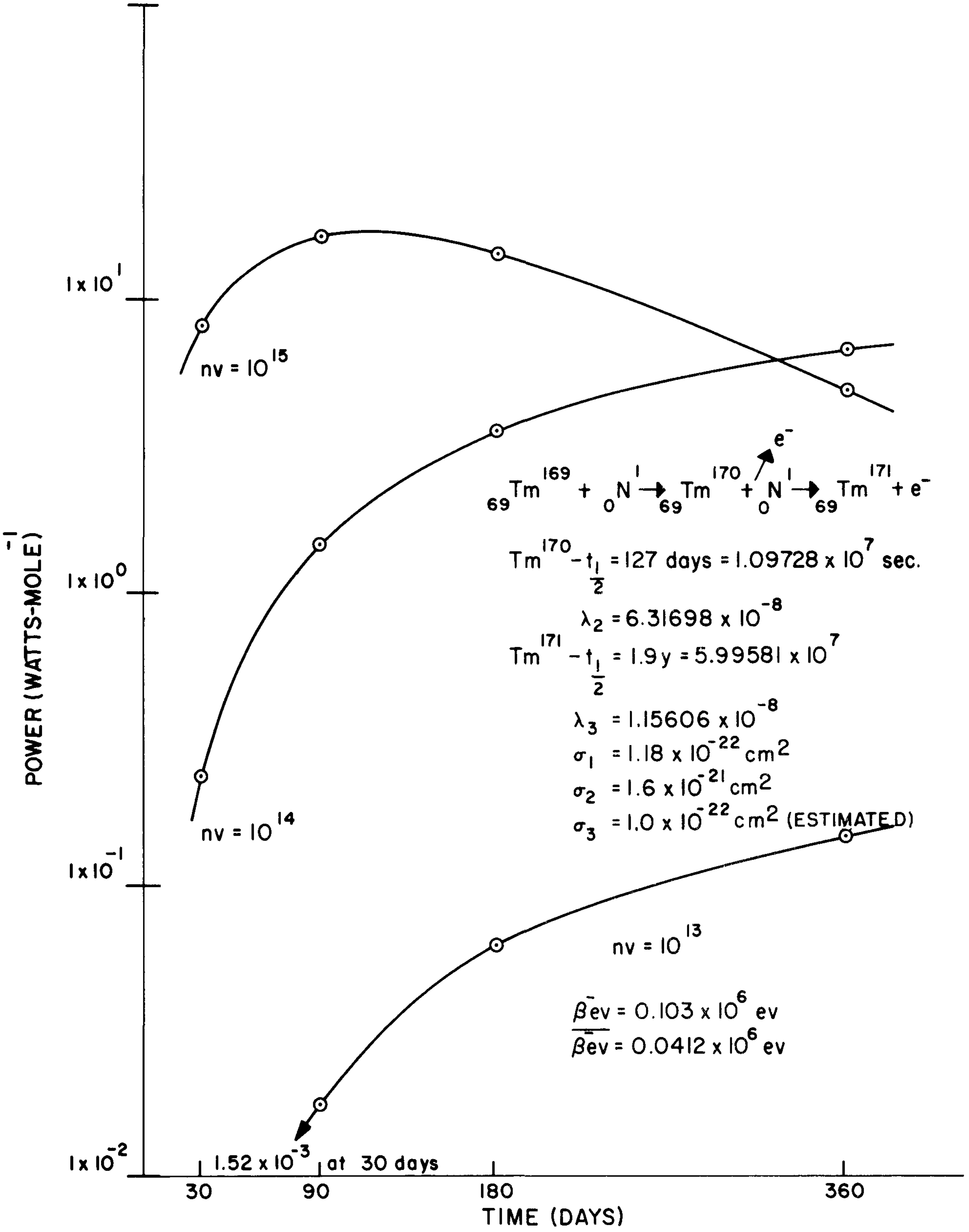

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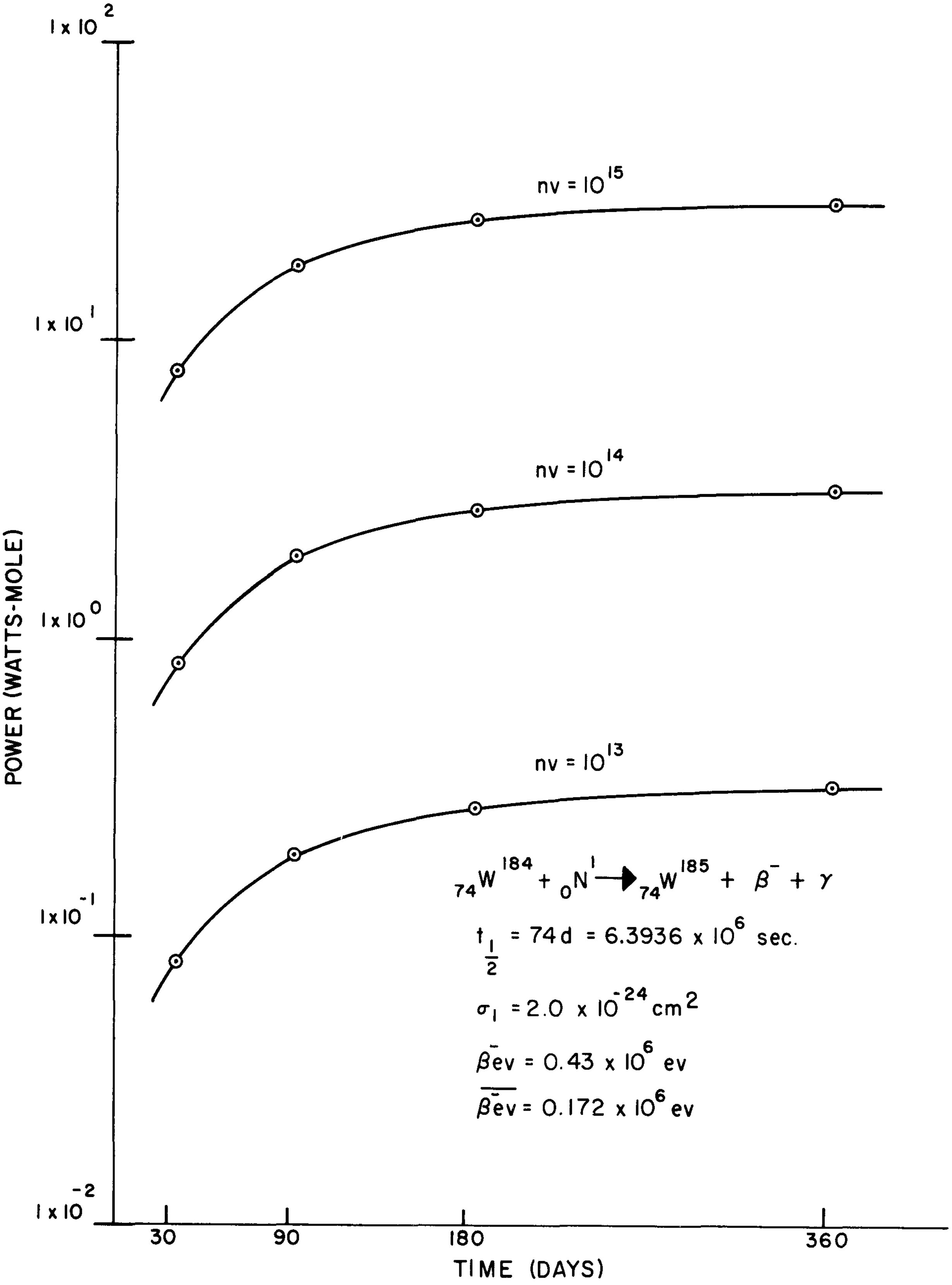
FIGURE 16 - FORMATION OF THULIUM-171



$_{74}W^{184} + {}_{0}N^{1} \longrightarrow {}_{74}W^{185} + \beta^{-} + \gamma$

$t_{\frac{1}{2}} = 74\,d = 6.3936 \times 10^{6}$ sec.

$\sigma_1 = 2.0 \times 10^{-24}\,cm^2$

$\bar{\beta}ev = 0.43 \times 10^{6}\,ev$

$\overline{\bar{\beta}ev} = 0.172 \times 10^{6}\,ev$

FIGURE 17 - FORMATION OF TUNGSTEN-185

8810

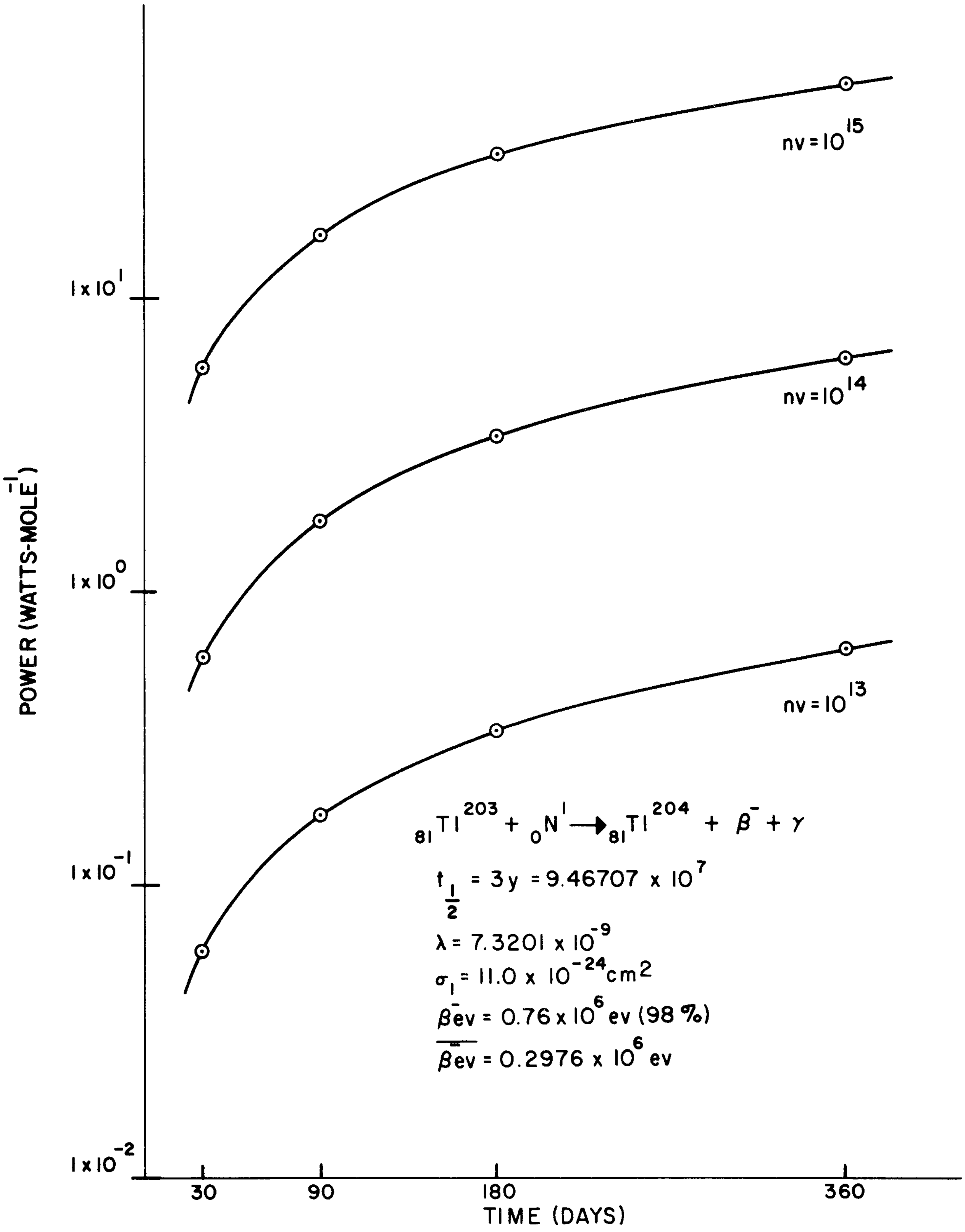


FIGURE 18 - FORMATION OF THALLIUM-204

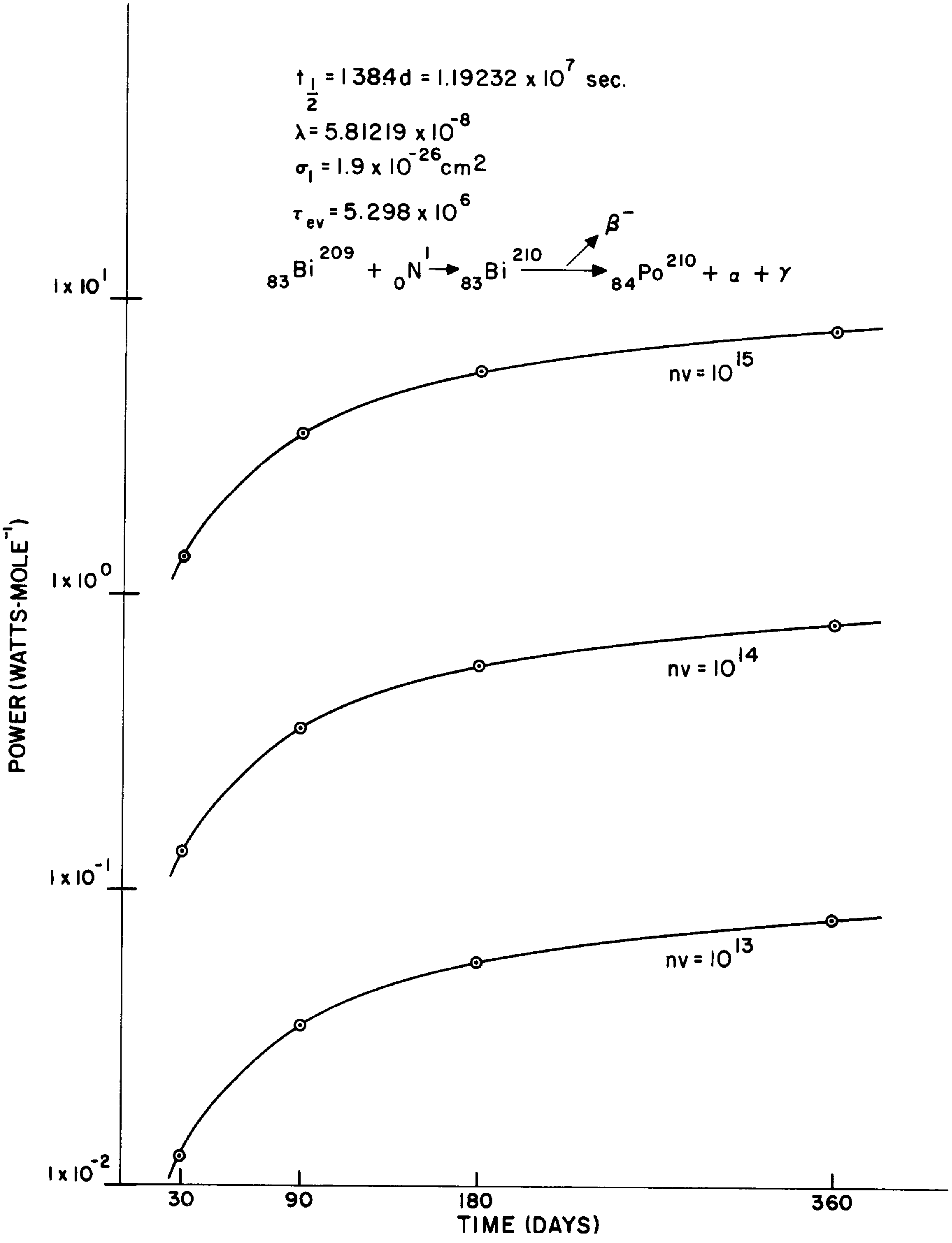


FIGURE 19 - FORMATION OF POLONIUM-210

8812

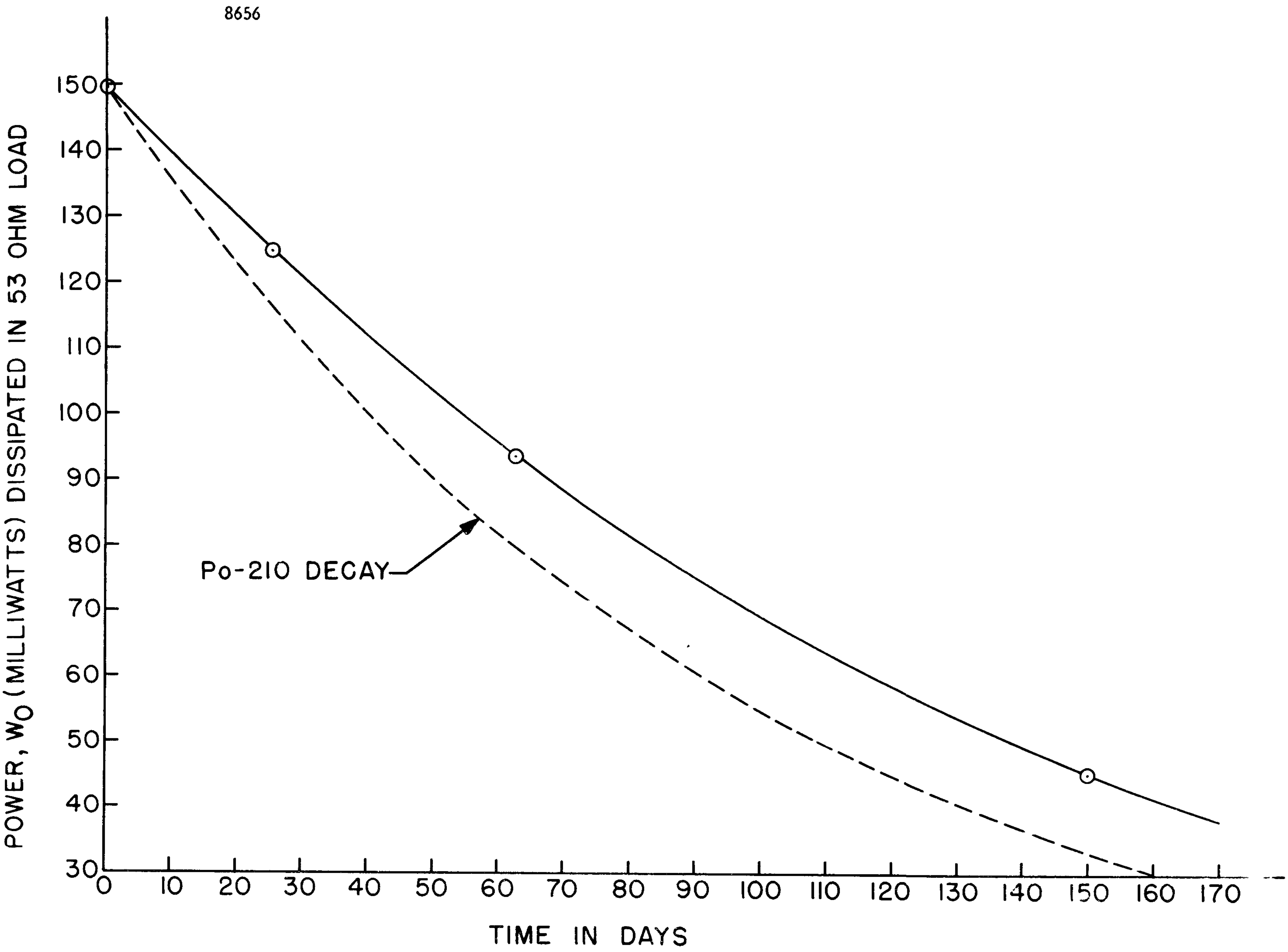


FIGURE 20 - CALCULATED POWER OUTPUT OF GENERATOR NO. 2 VS ELAPSED TIME AFTER LOADING A 701-CURIE POLONIUM-210 HEAT SOURCE

THALLIUM-204

Of all the above isotopes, the one new isotope that seemed to indicate the most promise because of availability was thallium-204. Natural thallium has a cross section of 3.29 barns, most of which is the 30 per cent occurrence of thallium-203 with a σ of 11.0 barns. The maximum beta energy of the thallium-204 is 0.765 Mev. It has at least seven times the half-life of polonium-210 and would, therefore, be more suitable for long term use, although the power density is less than $^1/_{130}$ of that of polonium.

Arrangements were made with the Phillips Petroleum Company of Arco and the United States Atomic Energy Commission to irradiate a button of natural thallium to investigate this isotope of thallium as a useful heat source (to permit direct measurements of this beta emmitter.) A slug was prepared for the Materials Testing Reactor, which consisted of a thallium button weighing 139.9109 grams, heliarc welded into a 2S aluminum jacket. It was flanked on both sides by bismuth buttons, also heliarc welded into aluminum jackets. These three jacketed buttons were then inserted into the aluminum slug, which served as protection from contamination by the cooling water and as a handling jacket, whose dimensions were so calculated to adequately absorb the heat from the radioactively occurring in the slug from absorbed particles and gammas. The slug was irradiated for a total of 1321 megawatt days through two cycles of the Materials Testing Reactor.

On its return to Mound Laboratory, the outer aluminum jacket was removed, and the three buttons in their aluminum jackets were calorimetered separately. Since the cross section of bismuth and the resulting calorimetry of the polonium formed is well known, (Polonium, TID-5221) an accurate average value of the reactor neutron flux was obtained for the slugs' locality. Using the derived expression (1) in MLM-CF-57-4-34 and ignoring the burnup of thallium-204 in the reactor:

$$\text{Watts} = \frac{1.602 \times 10^{-19}\, \lambda_2\, \overline{\beta}\, N^0\sigma,\, n\nu}{\lambda_2 + n\nu\,(\sigma_2 - \sigma_1)} \left[e^{-n\nu\sigma_1 t} - e^{-[\lambda_2 + n\nu\sigma_2]\, t} \right]$$

where

$t_{1/2} = 3.0$ years

$\lambda_2 = 6.325911 \times 10^{-4}$ years $^{-1}$

$\sigma_1 = 11. \times 10^{-24}$ cm^2

$\sigma_2 = 0$ (assumed)

$n\nu = 8.849 \times 10^{1}\ t^{-1}\ \text{cm}^{-2}$

$T = 34.0$ days

$N^0 = 1.2159 \times 10^{23}$ molecules

The measured wattage of thallium was 0.6419, calculated back to the time of removal from the reactor.

This permitted the calculation of $\overline{\beta}$ as 0.159 Mev or 21.25 per cent of $\overline{\beta}$ maximum.

In all of the calculations and for the isotopic growth rate curves (Figure 3 through Figure 19) $\overline{\beta}$ was assumed to be 0.4 x $\overline{\beta}$ (maximum). For thallium-204 the ratio of $\overline{\beta}$ (experimental to $\overline{\beta}$ (assumed) is 0.53; therefore, multiplying the thallium-204 curve values (Figure 18) by 0.53 will correct them to the experimental results.

The thallium-204 sample is being calorimetered periodically and eventually an accurate half-life can be calculated. The average $\overline{\beta}$ energy is seldom less than one fourth of the maximum energy. This indicates that the three year half-life used in the calculations is low.

Measurements on the natural occurring irradiated thallium, by comparison with those on the polonium buttons, indicate that it would have one watt per mole of heat output after 360 days in the reactor at 8.85 x 10^{15} n/cm^2/sec. Calculations indicate that a temperature rise of 400°C can be obtained with approximately 500 pounds of thallium as a sphere. Such a heat source would have about 1000 watts heat output. Approximately three watts per mole could be obtained by isotopic separation. Higher reactor flux levels would give substantial increases in heat output. The surface to volume ratio and the specific activity combine to make natural thallium heat sources of less than 1000 watts impractical if reasonable battery efficiencies are to be obtained.

GENERATOR

Under this project a complete theory of the thermoelectric generator for conductors was developed. Application was made of this theory in design of the thermoelectric generators throughout their construction. This theory reduces to those equations for conductors of Telkes and Joffee when their simplifying assumptions are added, and stands up well under experimental measurement with these thermoelectric generators.

Successful s uration of the Joule, Peltier and Thompson effects were measured using Generator Number 3, although the measured Thompson effect varied more than would be expected from normal experimental error.

The curve for the nuclear battery-thermocouple type[4] has been expanded to include higher $R_S W_S$ values and a line indicating a theoretical maximum efficiency for a given ΔT value of 600°C has been added (Figure 21). This was a family of curves generated by the expression:

$$E = \frac{1.43\ B\ R_S\ W_S}{(\alpha_1 + \alpha_2)\ (\Delta T + 2T_2)} \quad \frac{f_s}{(f_S + 1)^2} \quad \text{where}$$

B - thermoelectric power of the thermocouple

E - overall efficiency

R_S - thermal resistance of insulation hot to cold junction

W_S - heat generated by radioactive capsule

$\alpha_1 \alpha_2$ - Wiedeman Franz Constant

ΔT - temperature difference hot to cold junction °K

R_h - thermal resistance of capsule to hot junction

R_t - thermal resistance of leads hot to cold junction

f_s - $R_t/(R_h + R_S)$

T_2 - temperature of cold junction °K

R_h is here considered to be very small - essentially zero. The broad maximums indicate that thermal resistance through leads to that of insulation is not critical for maximum efficiency, provided that the ratio was greater than 0.3.

therefore, if

$$W_S R_S = 4.185 \ \frac{(f_s + 1)}{(\ f\ )} \cdot \Delta T,$$ ΔT may be determined by assigning values to $R_S W_S$ and f.

By choosing a maximum value for ΔT compatible with structural materials to be used and design parameters (for example - use of constantan limited the design to ΔT of 600°C) maximum values for f can be calculated for a given $W_S R_S$. If it is assumed that ΔT is limited to 600°, T_2 of 300°K, β equal to 7.7×10^{-5}, α_1 equal to 8.0×10^{-9}, α_2 equal to 10.0×10^{-9}, the corresponding efficiency can be calculated. The theoretical maximum efficiency attained if all the heat goes through the leads (or $f_S \longrightarrow 0$) under these conditions is 0.986.

This chart is useful for initial design estimates because W_S is usually known and a good estimate or R_S can be made. The f value at maximum efficiency, as at the intersept of maximum ΔT permitted, will give optimum performance for that generator.

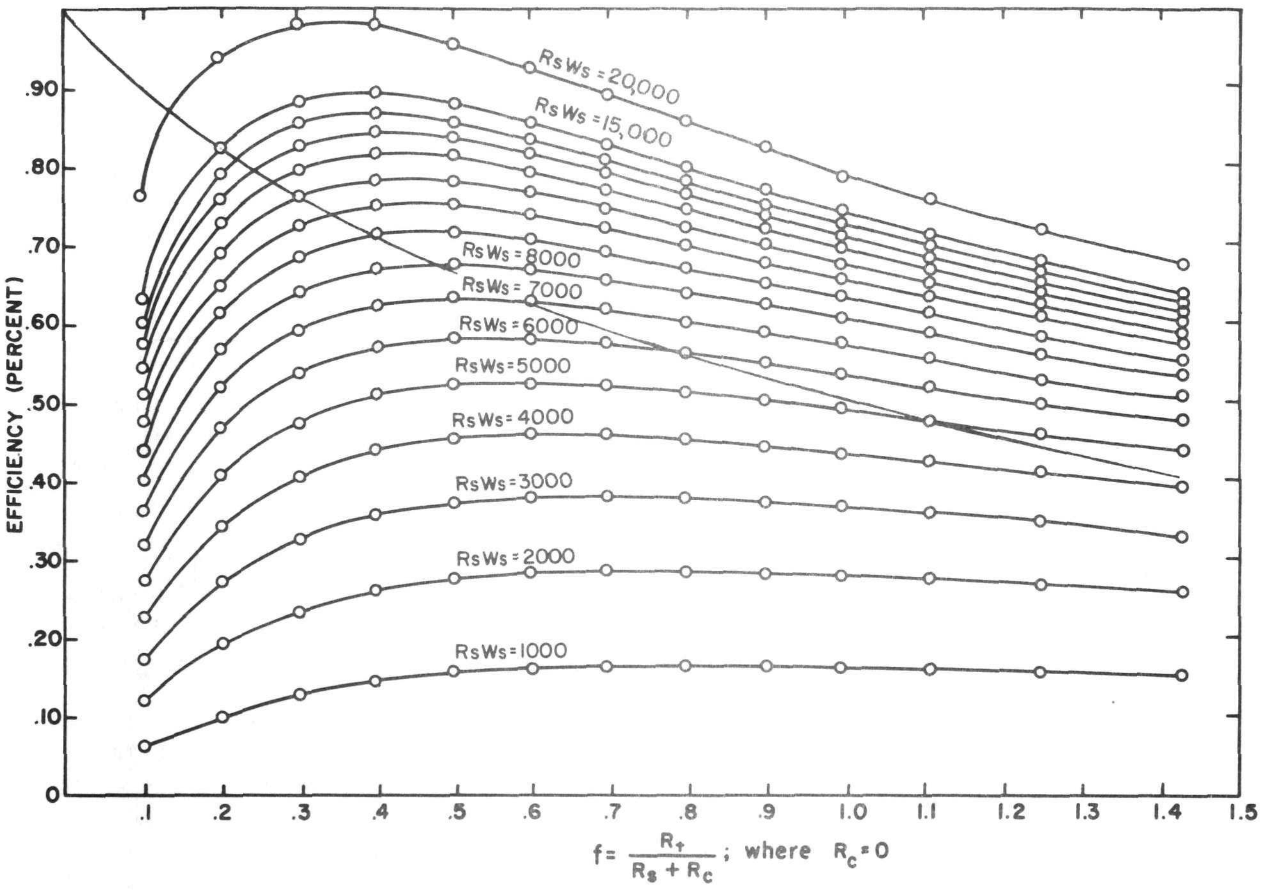


FIGURE 21 - GRAPH OF EFFICIENCY VS RATIO OF THERMAL RESISTANCE OF LEADS TO INSULATION

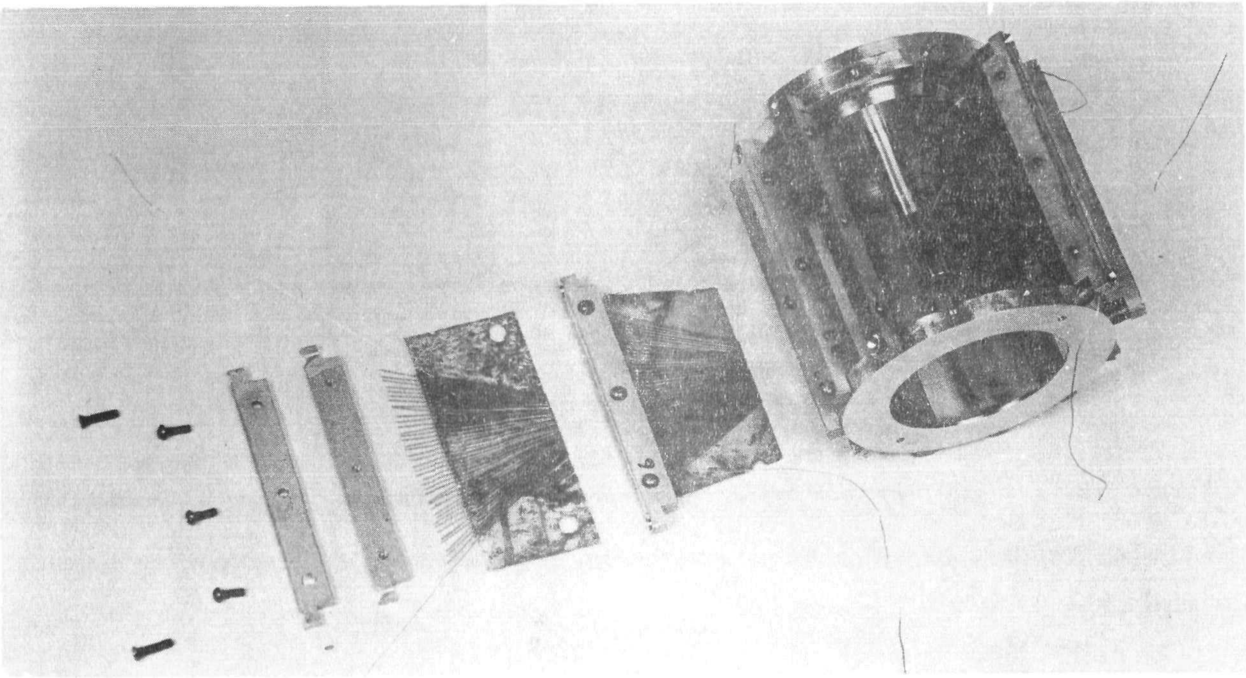


FIGURE 22 - THERMOELECTRIC GENERATOR NO. 3

THEORY OF THERMOELECTRIC GENERATOR

The theory of the thermoelectric generator as presented in MLM-984 (1954)[1] has several weak points. First, the loading effect was presented incorrectly and was eliminated because of the low anticipated efficiencies (MLM-984, Equation 4). The choice of relative cross-sectional area of the two thermocouple materials was also incorrect for materials with different Wiedemann-Franz constants. For completeness, the thermal resistances from the cold junctions to the ambient should have been considered. The heat loss from the surface of the thermocouple materials as presented in MLM-984, Equation 20 should have been included in the thermal circuit and not made a separate entity. It was not made clear that half the heat should flow through the leads *only* in the situation where the source power (W_s) is not large enough to achieve the maximum operating temperature (which will probably be the situation with most radioactive heat sources). The theory that follows is meant to supersede the theory given in MLM-984. Symbols used are listed in Table 2.

The thermal circuit of the generator is given in Figure 23. This circuit should be adaptable to any particular physical design. In some cases the relative values of the thermal resistances might be such that R_c or R_h could be set equal to zero, or R_s and R'_s might be deleted or combined. In any case the thermal circuit should have small values for R_c and R_h, large values for R_s and R'_s and a value for R_t such that the temperature from hot to cold junction (T_1T_2) is as large as practical (400-500°C for chromel-constantan). In other words, as great a portion of W_s should flow through R_t as is consistent with the maximum (T_1-T_2). There are three sources of heat shown in the thermal circuit. W_s is the total source power. W_h is the heat loss from the hot junctions due to loading and is the algabraic sum of Peltier cooling and one-half each of the Joule and Thompson heating. W_c is the heat henerated at the cold junctions due to loading and is the sum of the Peltier heating and one-half of the Joule and Thompson heating. The Joule and Thompson heating is divided 50-50 between hot and cold junctions only for linear thermocouple materials such as wire. For cylindrical materials a greater portion of the heat would be assigned to the hot junctions. Shilliday (1957) has derived the form factor for cylindrical materials.

Table 2

SYMBOLS

T_1	hot junction temperature (°K)
T_2	cold junction temperature (°K)
T_s	source container temperature (°K)
T_a	ambient temperature (°K)
$(T_1-T_2)_L$	temperature drop, hot to cold junctions, generator loaded (°C)
$(T_1-T_2)_O$	temperature drop, hot to cold junctions, no load (°C)
R_c	thermal resistance, cold junctions to ambient (°C-watt^{-1})
R_t	thermal resistance hot to cold junctions, of thermocouple materials, adiabatic surface conditions (°C-watt^{-1})
R_h	thermal resistance, source container to hot junctions (°C-watt^{-1})
R_s	thermal resistance, source container to ambient (°C-watt^{-1})
R'_s	thermal resistance, hot junction to ambient, which governs the loss of heat from the surface of the thermocouple materials (°C-watt^{-1})
n	number of thermocouples, hot or cold
a_1, a_2	cross-sectional area, a single lead, thermocouple material 1 or 2 (cm^2)
ρ_1, ρ_2	resistivity of thermocouple material 1 or 2 (ohm-cm)
k_1, k_2	thermal conductivity of thermocouple materials 1 or 2 (watt-cm^{-1}-°C^{-1})

α_1 α_2	Wiedemann-Franz constant of thermocouple materials 1 or 2 (volts^2 - $^\circ$C^{-1} - $^\circ$K^{-1})
L	length of thermocouple materials (cm)
W_s	source power (watts)
W_o	load power (watts)
W_h	power absorbed at hot junction by virtue of generator current; Peltier minus one-half the total Joule and Thompson heat (watts)
W_c	power supplied at cold junctions by virtue of generator current; Peltier plus one-half the total Joule and Thompson heat (watts)
Eff	efficiency of generator, W_o/W_s
$(\mathrm{Eff})_o$	efficiency of generator if there were no loading effect
R_L	load resistance (ohms)
R_g	internal resistance of generator (ohms)
e	EMF of a single junction (volts)
E_o	open-circuit EMF of generator (volts)
E_L	loaded EMF of generator (volts)
V_L	voltage across load (volts)
I	generator current (amperes)
β	thermoelectric power of a junction, $E_o/n\ (T_1-T_2)_o$ (volts-$^\circ$C^{-1})
$\dfrac{W_s}{1+f}$	heat through thermocouple materials (watts)
$\dfrac{fW_s}{1+f}$	heat lost (watts)
f_1	$R_t/(R_s' + R_c)$
f_2	R_c/R_s'
f_3	$R_s'(R_t + R_c)/(R_s' + R_t + R_c)\ (R_s + R_h)$
f_4	R_h/R_s
$(1 + f)$	$(1 + f_1)\ (1 + f_2)\ (1 + f_3)\ (1 + f_4)$
d	O.D. of source (cm)
h	length of source (cm)
D	O.D. of generator (cm)
A_s	surface area of source (cm^2)

Peltier and Thompson Effects Peltier and Thompson effects have been discussed extensively in the literature. Roeser[15] (1941) reviewed Thompson's original paper[16] (1857), and recommended the relationships derived by Thompson from the laws of thermodynamics. These relationships are:

$$\text{(Peltier effect) } P = T\left(\frac{de}{dT}\right) \text{ volts-junction}^{-1} \qquad (1)$$

$$\text{(Thompson effect) } \alpha_A - \alpha_B = T\left(\frac{d^2e}{dT^2}\right) \text{ volts - }^\circ\text{C}^{-1}\text{ - junction}^{-1} \qquad (2)$$

Bridgeman also has derived Equations 1 and 2, and his derivation is presented by Zemansky[17] (1943). Many papers have appeared recently on the application of the Peltier effect in refrigeration, and Shilliday's paper[18] (1957) is well referenced. The Peltier effect is confirmed by these papers. The Peltier effect was first used in calorimetric applications by Callendar[19] (1910) and more recently by Calvet[20] (1956) and by

Mann[21](1954). The Thompson equation has never been proved experimentally because of its relatively small effect in comparison to the heat conduction, Joule heat, and Peltier heat. However, if we accept the Peltier effect as given by Equation 1, then Equation 2 is mandatory in order to maintain a balance of energy. It was for this reason that Thompson hypothesized Equation 2. In our application this means that if we subtract all the energy leaving the generator from all the energy entering the generator, the result should equal the energy developed in the load, i.e., $W_S + W_C - W_h = W_S - W_O$. Thus in our thermal circuit, under steady-state conditions, W_S enters and leaves the system by the superposition theorem, and so we must have the condition that $W_C - W_h = - W_O = - I^2R_L$ (3)

In Annex A it is shown that Equation 3 is satisfied when the exact Peltier and Thompson effects are applied as given by Equations 1 and 2. Fortunately, Equation (3) is also satisfied by assuming that de/dT is a constant at all temperatures between T_2 and T_1 (which eliminates the Thompson effect) and numerically equal to β. Thus let

$$\frac{d^2e}{dT^2} = 0 \tag{4}$$

and

$$\left.\frac{de}{dT}\right|_{T_1} = \left.\frac{de}{dT}\right|_{T_2} = \beta = \frac{E_O}{n(T_1-T_2)_O} = \frac{E_L}{n(T_1-T_2)_L} \tag{5}$$

By substituting Equation 5 in Equation 1 and observing the 50-50 split of the Joule heat, the heat absorbed at the hot junctions is

$$W_h = n\beta I(T_1)_L - \frac{I^2R_g}{2} = \frac{E_L I(T_1)_L}{(T_1 - T_2)_L} - \frac{I^2R_g}{2} \tag{6}$$

and the heat generated at the cold junctions is

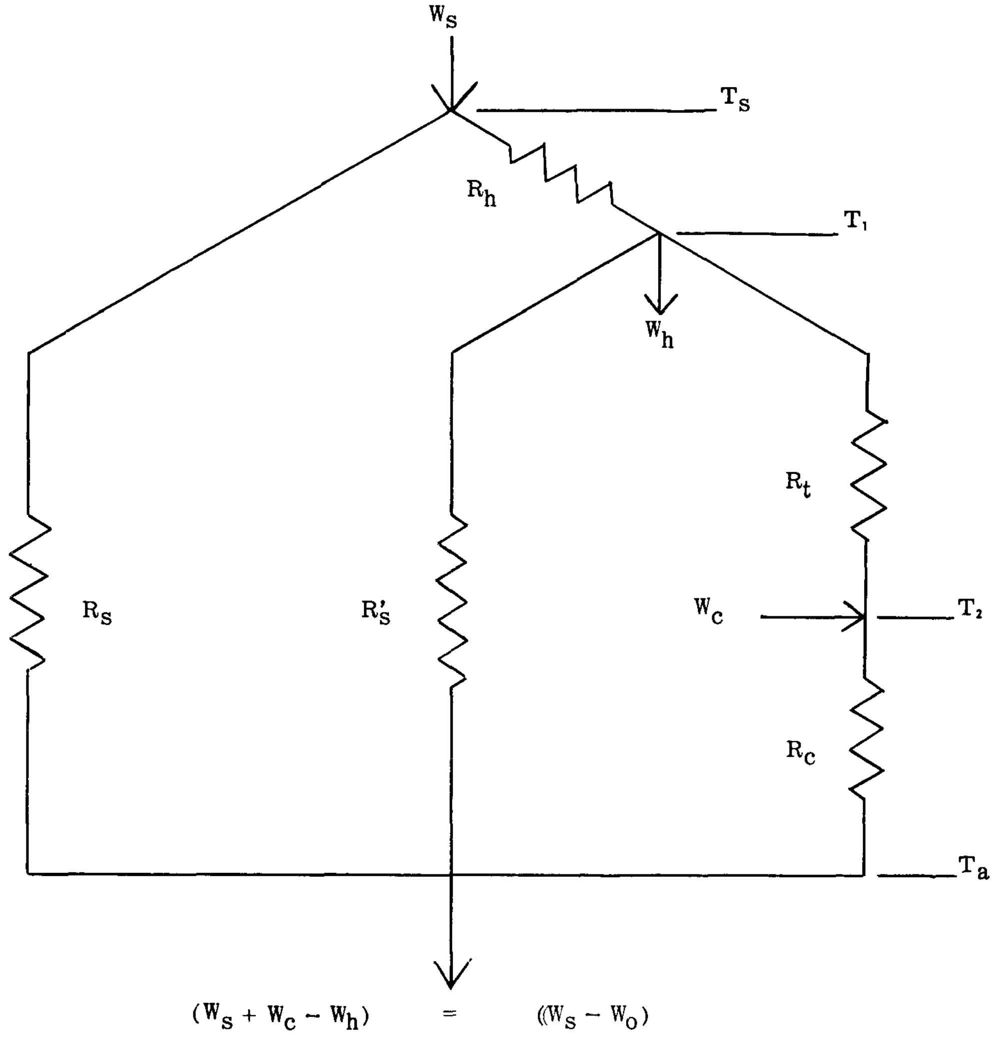
$$W_c = n\beta I(T_2)_L + \frac{I^2R_g}{2} = \frac{E_L I(T_2)_L}{(T_1 - T_2)_L} + \frac{I^2R_g}{2} \tag{7}$$

W_h in Equation 6 subtracted from W_c of Equation 7 gives

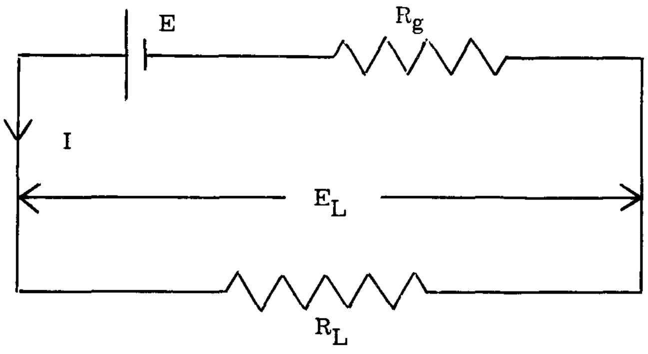
$$W_c - W_h = E_L I + I^2R_g = - W_o \tag{8}$$

which proves that Equation 6 and 7 satisfy the necessary conditions of Equation 3. The only error made by assuming Equations 4 and 5 is in the resulting temperatures (T_1 and T_2). These will be off by a slight amount depending on how the exact de/d_T changes in going from T_2 to T_1. If de/d_T varies linearly with temperature then there will be no error in using Equations 6 and 7.

Heat Flow Distribution in Thermal Circuit By virtue of the superposition therorem for electrical circuits, each source of heat (W_S, W_h, and W_c) can be treated separately and the results added to give the net distribution of heat flow and the temperatures within the thermal circuit under loaded conditions.



THERMAL CIRCUIT



ELECTRICAL CIRCUIT

FIGURE 23

The portion of W_s that flows through R_t is

$$\frac{W_s\, R_s}{R_s + R_h + \dfrac{R'_s\,(R_t + R_c)}{R'_s + R_t + R_c}} \times \frac{R'_s}{R_t + R_c + R'_s} \text{ watts} \tag{9}$$

The following substitutions are made in Equation 9 (see Table 1)

$$R_t = f_1\,(R'_s - R_c) \tag{10}$$

$$R_c = f_2\, R'_s \tag{11}$$

$$\frac{R'_s\,(R_t + R_c)}{R'_s + R_t + R_c} = f_3\,(R_s + R_h) \tag{12}$$

$$R_h = f_4\, R_s \tag{13}$$

Equation 9 is thereby reduced to

$$\frac{W_s}{(1 + f_1)\,(1 + f_2)(1 + f_3)\,(1 + f_4)} \tag{14}$$

By definition, let

$$(1 + f) = (1 + f_1)\,(1 + f_2)\,(1 + f_3)\,(1 + f_4) \tag{15}$$

The term $(1 + f)$ can be obtained for a given generator by calculating R_t and measuring $(T_1 - T_2)_0$ for a given W_s. Thus

$$(T_1 - T_2)_0 = \frac{W_s\, R_t}{1 + f} = \frac{W_s\, R_t}{(1 + f_1)\,(1 + f_2)\,(1 + f_3)\,(1 + f_4)} \tag{16}$$

The reason for choosing f_1, f_2, f_3, and f_4 as defined by Equations 10, 11, 12, and 13 is so that, for $R'_s = \infty$, $f_1 = 0$; $R_c = 0$, $f_2 = 0$; $R_s = \infty$, $f_3 = 0$; and for $R_h = 0$, $f_4 = 0$. (In MLM-984, $f_2 = 0$, $f_3 = f$, $f_4 = M$, and $f_1 = \pi naHLR_0$ by Eq. 20). The definition for f does not change under any condition.

The portion of W_h that flows through R_t is

$$\frac{\dfrac{W_h\, R'_s\,(R_h + R_s)}{R'_s + R_s + R_h}}{\dfrac{(R_h + R_s)\, R'_s}{R'_s + R_s + R_h} + R_t + R_c} \tag{17}$$

which can be reduced by substitution of Equations, 10, 11, 12, and 13 to

$$\frac{W_h}{(1 - f_1)\,(1 + f_2)\,(1 + f_3)} \tag{18}$$

The temperature drop from hot to cold junctions caused by W_h is

$$\Delta (T_1 - T_2)_L = \frac{R_t W_h}{(1 + f_1)(1 + f_2)(1 + f_3)} \qquad (19)$$

The small portion of W_c that passes through R_t is omitted, and so the full loading effect caused by the current (I) is given by Equation 20.

$$(T_1 - T_2)_O - (T_1 - T_2)_L = \frac{R_t W_h}{(1 + f_1)(1 + f_2)(1 + f_3)} \qquad (20)$$

The absolute values of T_1 and T_2 are raised by W_c to the extent of $R_c W_c$ (°C). W_h lowers T_2 by $W_h R_c/(1 + f_1)(1 + f_2)(1 + f_3)$ °C and lowers T_1 by $W_h (R_t + R_c)/(1 + f_1)(1 + f_2)(1 + f_3)$ °C. The temperatures of T_1 and T_2 under load conditions are then

$$(T_1)_L = T_a + \frac{W_s(R_t + R_c)}{(1 + f)} + R_c W_c - \frac{W_h (R_t + R_c)}{(1 + f_1)(1 + f_2)(1 + f_3)} \,^{\circ}\mathrm{K} \qquad (21)$$

and

$$(T_2)_L = T_a + \frac{W_s R_c}{(1 + f)} + R_c W_c - \frac{W_h R_c}{(1 + f_1)(1 + f_2)(1 + f_3)} \,^{\circ}\mathrm{K} \qquad (22)$$

Subtracting Equation 22 from Equation 21, gives the temperature drop from hot to cold junctions under load.

$$(T_1 - T_2)_L = \frac{R_t [W_s - (1 + f_4) W_h]}{(1 + f)} \,^{\circ}\mathrm{C} \qquad (23)$$

Derivation of Efficiency Equations An expression for the efficiency (Eff = W_O/W_S) will now be derived. The current through the load is

$$I = \frac{n\beta (T_1 - T_2)_L}{R_L + R_g} \qquad (24)$$

and the power dissipated in the load is

$$W_O = I^2 R_L \qquad (25)$$

The efficiency is then

$$\mathrm{Eff} = \frac{W_O}{W_S} = \frac{n^2 \beta^2 (T_1 - T_2)^2_L R_L}{W_S (R_L + R_g)^2} \qquad (26)$$

Equation 26 has no practical value since $(T_1 - T_2)_L$ cannot be calculated until the efficiency is known and can only be measured in a generator by a transient method. The efficiency in terms of $(T_1 - T_2)_O$ will be much more useful.

Substitution of Equation 23 in Equation 26 gives

$$\mathrm{Eff} = \frac{n^2\beta^2 R_L R_t^2 [W_S - (1 + f_4) W_h]}{W_S (R_L + R_g)^2 (1 + f)^2} \qquad (27)$$

Equation 27 also cannot be solved directly since W_h depends upon the efficiency.

By solving Equation 16 for R_t and substituting for one of the R_t's in Equation 27, and equivalent statement of the efficiency is obtained.

$$\mathrm{Eff} = \frac{n^2\beta^2 R_L R_t \quad (T_1 - T_2)_O}{(R_L + R_g)^2 (1 + f)} \times \left[- \frac{(1 + f_4) W_h}{W_s} \right]^2 \tag{28}$$

If W_h is assumed to be small (no loading), then Equation 28 reduces to a definition of (Eff)o

$$\mathrm{Eff} = \frac{n^2\beta^2 R_L R_t (T_1 - T_2)_O}{(R_L + R_g)^2 (1 + f)} \tag{29}$$

The corrective term in Equation 28 also contains Eff which can be obtained as follows: W_h can be expressed in terms of W_O by combining Equations 6, 24, and 25.

$$W_h = W_O \left[\left(\frac{T_1}{T_1 - T_2} \right)_L \left(\frac{R_L + R_g}{R_L} \right) - \frac{R_g}{2R_L} \right] \tag{30}$$

Equation 29 and 30 substituted in Equation 28 gives a nearly exact and yet useful equation for the efficiency.

$$\mathrm{Eff} = (\mathrm{Eff})_O \left[1 - \mathrm{Eff}\,(1 + f_4) \left[\left(\frac{T_1}{T_1 - T_2} \right)_L \quad \left(\frac{R_L + R_g}{R_L} \right) \frac{R_g}{2R_L} \right] \right]^2 \tag{31}$$

Equation 31 could be solved uniquely for the efficiency, but this would involve a quadratic equation. A reiterative method, starting with the evaluation of (Eff)o, is easier. Equation 31 contains a ratio of temperatures under loaded conditions, but a negligible error will occur if unloaded temperatures are used.

Since from equation 26 $\quad \mathrm{Eff} \approx K_1 (T_1 - T_2)_L \qquad (32)$

and

$$\left(\frac{T_1}{T_1 - T_2} \right)_L \left(\frac{R_L + R_g}{R_L} \right) >> \frac{R_g}{2R_L} \tag{33}$$

with a given generator and load, Equation 31 can be converted by using this approximation for the corrective terms to

$$\mathrm{Eff} = (\mathrm{Eff})_O \, [1 - K_2 T_1]^2 \tag{34}$$

or since K_2 is small

$$\frac{(\mathrm{Eff})_O - \mathrm{Eff}}{(\mathrm{Eff})_O} = 2 K_2 T_1 \tag{35}$$

Equation 35 shows that the per cent difference between the true efficiency and $(\mathrm{Eff})_O$ increases only as the absolute temperature of the hot junctions. The same statement is true for the difference between E_L and E_O, or for $(T_1 - T_2)_L$ and $(T_1 - T_2)_O$.

The only assumptions made in the derivation of Equation 31 are: (1) no part of W_c passes through R_t and, (2) a second order correction was dropped in applying the Peltier and Thompson effects. Another second order approximation will now be made in Equation 31. Let

$$\mathrm{Eff}^2\,(1+f_4)^2 \left[\left(\frac{T_1}{T_1-T_2}\right)_L \left(\frac{R_L+R_g}{R_L}\right) - \frac{R_g}{2R_L}\right]^2 = 0 \tag{36}$$

With Eff = 0.01, $(T_1 - T_2) = 400°\mathrm{C}$, $T_2 = 300°\mathrm{K}$, $R_L = R_g$, and $f_4 = 0$, the error made in assuming Equation 36 is 0.09 per cent of Eff. Equation 31 can now be written

$$\mathrm{Eff} = \frac{(\mathrm{Eff})_0}{1 - 2\,(\mathrm{Eff})_0\,(1+f_4) \left[\left(\frac{T_1}{T_1-T_2}\right)_L \left(\frac{R_L-R_g}{R_L}\right) - \frac{R_g}{2R_L}\right]} \tag{37}$$

The Optimum Load Resistance The solution of

$$\frac{\partial\,(\mathrm{Eff})}{\partial R_L} = 0 \tag{38}$$

should give nearly the optimum load resistance, since Equation 38 becomes an exact derivative if $(T_1/T_2)_L$ is assumed to be a constant. The solution of Equation 38 for optimum load resistance is

$$R_L = R_g \sqrt{1 + \frac{n^2\beta^2 R_t\,[T_2 + T]}{R_g\,(1+f_1)\,(1+f_2)\,(1+f_3)}} \tag{39}$$

Choice of Relative Wire Size, a_1/a_2 The equation of efficiency (Equations 26, 27, 28, 29, 31, 34, and 37) can be stated in terms of the thermal conductivities and electrical resistivities of the thermocouple materials. R_t and R_g are related to the ρ's and k's by the following equations:

$$R_t = \frac{L}{n\,(k_1 a_1 + k_2 a_2)} \tag{40}$$

$$R_g = nL \left(\frac{\rho_1}{a_1} + \frac{\rho_2}{a_2}\right) \tag{41}$$

$$\frac{R_g}{R_t} = n^2\,(k_1 a_1 + k_2 a_2) \left(\frac{\rho_1}{a_1} + \frac{\rho_2}{a_2}\right) \tag{42}$$

Since R_t/R_g appears in Equation 39, the choice of a_1/a_2 will affect the optimum load resistance. However, we do not want to make our choice of a_1/a_2 dependent upon R_L. Therefore, for simplicity, let $R_L = R_g$, which converts Equations 29 and 37 to

$$(\mathrm{Eff})_0 = \frac{n^2\beta^2 R_t\,(T_1 - T_2)_0}{4\,R_g\,(1+f)} \tag{43}$$

$$\mathrm{Eff} = \frac{(\mathrm{Eff})_0}{1 + 2(\mathrm{Eff}_0)(1+f_4) \left[2\left(\frac{T_1}{T_1-T_2}\right)_L - \frac{1}{2}\right]} \tag{44}$$

An inspection of Equations 43 and 44 shows that for maximum efficiency R_t/R_g should be a maximum, i.e., Equation 42 should be a minimum. Since Equation 42 cannot have both a maximum and a minimum for a_1/a_2 then the solution of

$$d\left(\frac{R_g}{R_t}\right) \Big/ d\left(\frac{a_1}{a_2}\right) = 0 \tag{45}$$

should give the optimum ratio of a_1/a_2 is

$$\frac{a_1}{a_2} = \sqrt{\frac{k_2\rho_1}{k_1\rho_2}} \tag{46}$$

Equation 46 is the same result as that obtained by Telkes[22, 23] (1947) by a different method under different conditions. If the two thermocouple materials have the same Wiedemann-Franz constant ($\rho_1 k_1 = \rho_2 k_2$), then both metals will conduct the same amount of heat and have the same electrical resistance. If the two metals do not have the same Wiedemann-Franz constant, then the metal that conducts the more heat will have the larger electrical resistance.

Substituting Equation 46 in Equation 42 gives the optimum ratio of R_g/R_t.

$$\frac{R_g}{R_t} = n^2 \left(\sqrt{\rho_1 k_1} + \sqrt{\rho_2 k_2}\right)^2 \tag{47}$$

Generator Design for a Specified Load The required number of junctions is dependent on R_L, the desired load power (W_O), the thermoelectric power of the couples, and the type of heat source. It will be assumed that the heat source is such that W_S can be made large enough to produce the maximum operating temperature of either T_S or T_1 which are designated $(T_S)_m$ or $(T_1)_m$. $(T_S)_m$ depends upon the type of source and $(T_1)_m$ depends upon the thermocouple materials. In the case of radioactive sources and chromel-constantan couples $(T_S)_m$ and $(T_S)_m$ are about 770°K. The absolute values of T_S, T_1 and T_2 cannot be determined until the entire generator has been designed, but from approximations of T_2 and R_C one can estimate the maximum allowable temperature difference between hot and cold junctions $(T_1 - T_2)_m$. To produce W_O watts in a load of R_L ohms with a generator of internal resistance $R_g = R_L$ ohms, the generator EMF is

$$E_L = 2\sqrt{W_O R_L} = 2\sqrt{W_O R_g} \quad \text{volts} \tag{48}$$

where at a temperature difference of $(T_1 - T_2)_m$ °C,

$$E_L = n\,\beta\,(T_1 - T_2)_m \text{ volts} \tag{49}$$

Solving Equations 48 and 49 for n gives the required number of junctions.

$$n = \frac{2\sqrt{W_O R_L}}{\beta\,(T_1 - T_2)_m} = \frac{2\sqrt{W_O R_g}}{\beta\,(T_1 - T_2)_m} \tag{50}$$

The same conditions that are used to determine the necessary number of junctions, are also used to calculate the cross-sectional areas of each wire. Substituting the optimum ratio of a_1/a_2 as given by Equation 46 into Equation 41 gives

$$R_g = \frac{n\,L}{a_2}\left[\sqrt{\frac{k_1\,\rho_1\,\rho_2}{k_2}} + \rho_2\right] = R_L \tag{51}$$

Solving Equation 51 for a_2 and substituting Equation 50 for n gives

$$a_2 = \frac{2L\,\sqrt{W_O}\,\left(\sqrt{k_1\,\rho_1\,\rho_2} + \rho_2\,\sqrt{k_2}\right)}{\beta\,\sqrt{k_2 R_L}\,(T_1 - T_2)_m} \tag{52}$$

After obtaining a_2, a_1 can be calculated by Equation 46.

Generator Design for a Specified Heat Source and Load Resistance If the power source and load resistance are specified, the problem of generator design is quite different. The source will have a fixed surface area and, consequently, the R_S of the generator can be no greater than that obtainable with the best insulation. In MLM-984, it was shown that under these conditions (W_S, R_S, R_C assumed constant), and with k_1 and k_2 independent of temperature, that $R_t = R_S + R_C$ for maximum efficiency. With $R_C = 0$ this meant half the heat should flow through the wires. Later, in Figure 3 in MLM-CF-57-8-27 "Nuclear Battery-Thermocouple Type, Second Quarterly Report, April 1, 1957 to June 30, 1957," we showed the shift in the 50-50 distribution resulting from assuming that k_1 and k_2 were varying with temperature according to the Wiedemann-Franz law.[4] The shift in the 50-50 distribution is not valid because of assuming R_S constant. Actually, as the temperature of a generator is increased, R_S and R_t both decrease, and the per cent of heat through the leads remains nearly constant.

Of course, with a large W_S, the temperature would increase beyond the $(T_1 - T_2)_m$.

Brooks[24] (1941), in a discussion of a paper by Gier and Boelter on a thermopile design, also states that half the heat should flow through the thermocouple wires. Calvet[20] (1956), in designing a calorimeter, has likewise derived the 50-50 splitting of the heat flow and says that Tonnelat[25] (1945) established the relationship. The optimum value for R_t for the present thermal circuit can be established as the solution of

$$\frac{\partial\,(\mathrm{Eff})}{\partial R_t} = 0 \tag{53}$$

Applying Equation 53 to the exact equation of the efficiency, Equation 31, shows that to a first approximation

$$\frac{\partial\,(\mathrm{Eff})}{\partial\,R_t} \approx \frac{\partial(\mathrm{Eff})_O}{\partial\,R_t} \tag{54}$$

Assuming a matched load and the proper wire sizes, Equation 55 is obtained by substituting Equation 47 in Equation 43.

$$(\mathrm{Eff})_O = \frac{\beta^2\,(T_1 - T_2)_O}{4\,(1 + f)\left(\sqrt{\rho_1\,k_1} + \sqrt{\rho_2\,k_2}\right)^2} \tag{55}$$

W_S is fixed so that it must appear in Equation 55 before the partial derivative is taken. This can be done only by substituting Equation 16.

$$(\mathrm{Eff})_O = \frac{\beta^2 W_S R_t}{4\,(1+f)^2 \left(\sqrt{\rho_1 k_1} + \sqrt{\rho_2 k_2}\right)^2} \tag{56}$$

From an inspection of Equation 10, 11, 12, 13, and 15, f_2 and f_4 do not contain an R_t so that Equation 56 can be written

$$(\mathrm{Eff})_O = \left[\frac{\beta^2 W_S}{\left(4\ \sqrt{\rho_1 k_1} + \sqrt{\rho_2 k_2}\right)^2\ (1+f_2)^2 (1+f_4)^2}\right] \frac{R_t}{(1+f_1)^2 (1+f_3)^2} \tag{57}$$

The portion of Equation 57 in brackets is to be considered a constant so that the optimum value for R_t is in the solution of

$$\partial \left[\frac{R_t}{(1+f_1)^2\ (1+f_3)^2}\right] \Big/ \partial R_t = 0 \tag{58}$$

Substituting Equation 10 and 12 in Equation 58 and solving for R_t gives

$$R_t = \frac{R'_S R_S + R'_S R_h + R_S R_c + R_c R_h + R'_S R_c}{R_S + R_h + R'_S} \tag{59}$$

From an inspection of Equation 9, half the heat will flow through R_t when

$$\frac{R_S}{R_S + R_h + \dfrac{R'_S (R_t + R_c)}{R'_S + R_t + R_c}} = \frac{R_t + R_c + R'_S}{2 R'_S} \tag{60}$$

Solving Equation 60 for R_t one obtains

$$R_t = \frac{R_S R'_S - R'_S R_h - R_S R_c - R_c R_h - R'_S R_c}{R_S + R_h + R'_S} \tag{61}$$

Comparing Equations 59 and 60 it is seen less than half the heat flows through R_t for maximum efficiency. Equation 59 reduces to $R_t = R_S$ for $R'_S = \infty$ and $R_c = R_h = 0$ and so half the heat should flow through R_t in this case.

With R_t fixed by Equation 59, $R_g = R_L$, and with R_g/R_t given by Equation 47, the optimum number of junctions is given by

$$n = \frac{1}{\sqrt{\rho_1 k_1} + \sqrt{\rho_2 k_2}} \sqrt{\frac{R_L (R_S + R_h + R'_S)}{(R_S R'_S + R'_S R_h + R_S R_c + R_c R_h + R'_S R_c)}} \tag{62}$$

Since R'_S, and to a certain extent R_c and R_h, depend upon n, Equation 62 must be solved by a reiterative method.

The cross-sectional areas are given by solving Equations 46, 51, and 62 for a_1 and a_2.

Generator Design for a Specified Load Using Sources of Low Power Density For a given radioactive isotope there is a maximum attainable power density depending upon the sealing techniques used, the half-life, the density, and the average energy per disintegration. Hittman[26] (1957) has derived a useful equation for the power density. For polonium-210, the power density is so large that W_S can be considered independent of A_S in most applications. For an isotope such as strontium-90/yttrium-90 with about one-fifth the average energy per disintegration of polonium-210 and 60 times the half-life, the power density of the isotope alone would be 300 times smaller than for polonium-210. A source of W_S watts of this isotope might have such a large A_S and consequently low R_S that the efficiency would be too low to be practical. Considering only right cylindrical sources (d = h), W_S would be proportional to d^3.

$$W_S = k_1 d^3 \text{ watts} \tag{63}$$

The area of the source, under the same conditions, would be proportional to d^2.

$$A_S = k_2 d^2 \text{ cm}^2 \tag{64}$$

The heat flow per unit area of the source would be

$$\frac{W_S}{A_S} = (k_1/k_2)d \text{ watts - cm}^{-2} \tag{65}$$

For a specified insulating material, a given ratio of D to d, and R_t selected by Equation 59, the temperature rise would be proportional to W_S and inversely proportional to A_S.

$$\frac{W_S}{A_S} = k_3 (T_1 - T_2)_O \tag{66}$$

Equating Equations 66 and 65, one finds as a first approximation that the temperature rise, and consequently the efficiency, is proportional to the diameter of the heat source,

$$(T_1 - T_2)_O = (k_1/k_2 k_3)d \tag{67}$$

or by Equation 55

$$(\text{Eff})_O = \frac{\beta^2}{4(1+f)\left(\sqrt{\rho_1 k_1} + \sqrt{\rho_2 k_2}\right)^2} \quad \frac{k_1 d}{k_2 k_3} \tag{68}$$

By the definition of efficiency and Equations 63 and 68

$$W_O \approx W_S (\text{Eff})_O \approx \frac{\beta^2}{4(1+f)\left(\sqrt{\rho_1 k_1} + \sqrt{\rho_2 k_2}\right)^2} \quad \frac{k^2{}_1 d^4}{k_2 k_3} \tag{69}$$

or $$W_O = k_4 d^4 \tag{70}$$

Since the power output of the generator increases as the fourth power of the diameter of the source, the situation is not too adverse even for low power-density sources.

After determining d from Equation 69, R_S can be calculated, and then n may be determined by Equation 62.

DISCUSSION OF GENERATORS

Seven thermoelectric generators were built to check design theory and construction techniques. Generator Number 2 consisted of wheel-type thermopiles of 40 Chromel-Constantan thermocouples stacked coaxially about a common core containing the heat sources. Generator Number 2A of the same design but having smaller and more numerous thermocouples was started but never tested.

Generator Number 3, an improved design of Generator Number 2, was the prototype of all succeeding generators built. This consisted of 12 thermopiles consisting of 37 thermocouples each supported by 0.003 inch mica sheets. These mica cards were vertically mounted and radially spaced on two two-inch aluminum rings so that the edges of the mica cards holding the hot junctions support the radioactive heat source. The cold junctions face outward on the periphery. This design has the advantage that the mica cards are interchangeable and can be removed for repair. The heat source diameter is essentially independent of the mica card dimensions and heat source geometries other than cylinderical are possible. More junctions per area of heat sources can be added with this construction. (See Figure 24).

Extensive measurements were made on Generators 2 and 3 to determine efficiencies, effects of various types of insulation including vacuum, noise levels, ambient temperatures, matched and unmatched loads, thermal circuits and resistances at various power levels[7, 8, 9].

Generator Number 3 became a demonstration unit with an electrically powered synthetic radioactive source and was sent to the United States Army Signal Corps. Research and Development Laboratories at Ft. Monmouth, New Jersey for further checking.

Generators Number 5 and 6 were identical. The separate numbering systems were indicated because the generator was completely dismantled and rebuilt (during construction). It was the first completely engieered radioactive powered thermoelectric battery built. Specifications were to make a generator capable of 30 millwats powered at independent voltages of 1.2 and 4.8 volts after six months of use.[10] The generator had to be sufficiently rugged enough to undergo vibrational and gravitational testing.[10] It was 2.955 inches in diameter by 2.911 inches long. It has an internal resistance of 780 ohms and produced 6.25 volts with an input of 15.612 watts at an efficiency of 0.32 per cent, which was the calculated decay value of the 1200 curie polonium heat source after six months. A test heat source containing 430 curies of polonium-210 was made to check transfer and sealing techniques.

Generator Number 8 was an actual battery constructed similarly to Generator Number 6. The target design was 50 milliwatts output at 4.8 and 1.2 volts after six months with a 1100 curie polonium-210 heat source. Certain modifications were built in the outer case, since it was to be partially used for demonstration purposes.

Heat sources of the 1100 curie size have been made only in the past two years by the Mound Laboratory Neutron Source Group. Although this source was made successfully, there was no way to check the transfer efficiency until after the source was hermetically sealed. Therefore, when the heat source (Figure 24) for Generator Number 8 was made, it was not known until after it had been sealed that it contained only 880 curies rather than the 1100 curies which had been planned.

The gamma energy spectrum of this heat source is shown in Figure 25 and was determined by pulse-height analysis equipment. No adsorbers were used. Since some impurities such as metal oxides, are present, there is a small neutron flux. Nominally, 100 neutrons per second per curie of polonium can be considered to be the minimum attainable value.

Since this source had lower heat output than intended, 74 junction pairs (two complete mica card assemblies) were removed from the higher voltage section. This permitted a better thermal matching of the heat load to the heat source to obtain a maximum electrical power output. Voltage and power output data were taken on generator Number 8 and are shown in Figure 26.

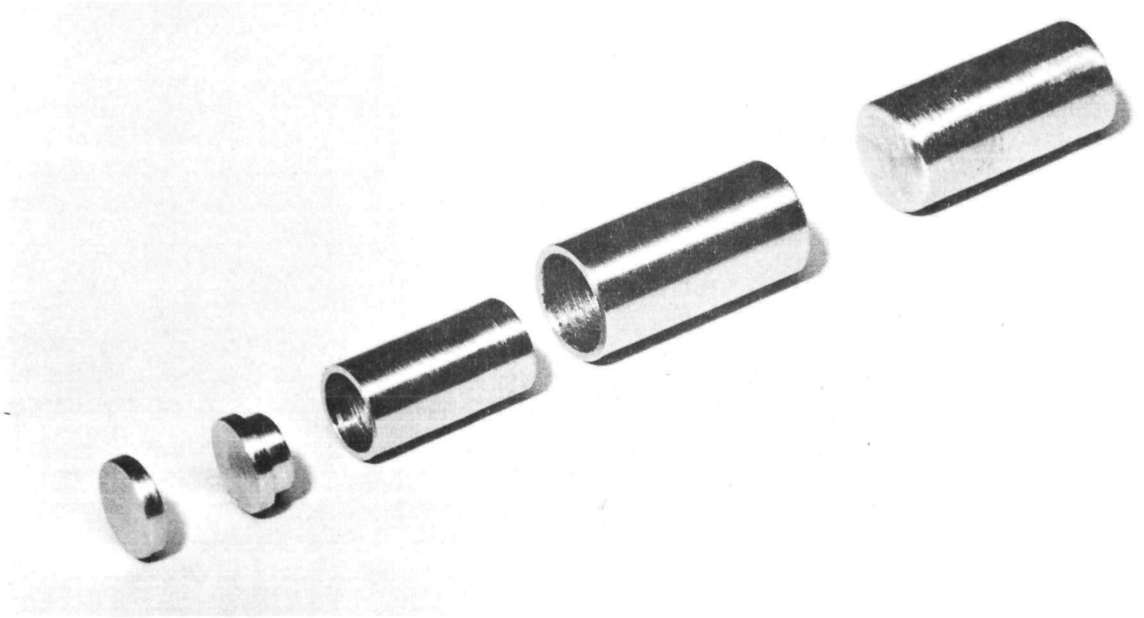


FIGURE 24 - HEAT SOURCE

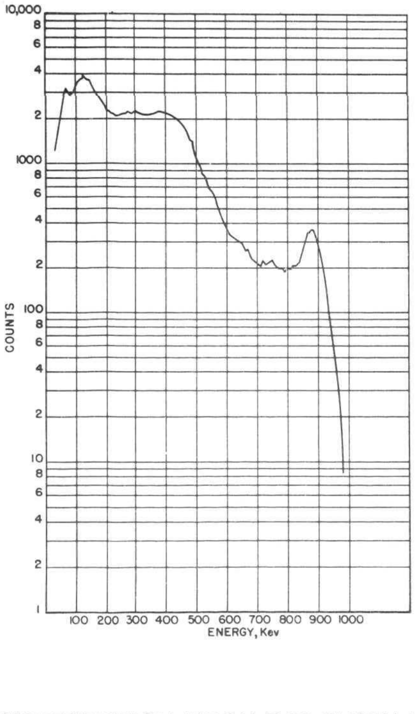


FIGURE 25 - GAMMA DISTRIBUTION FROM POLONIUM BATTERY HEAT SOURCE

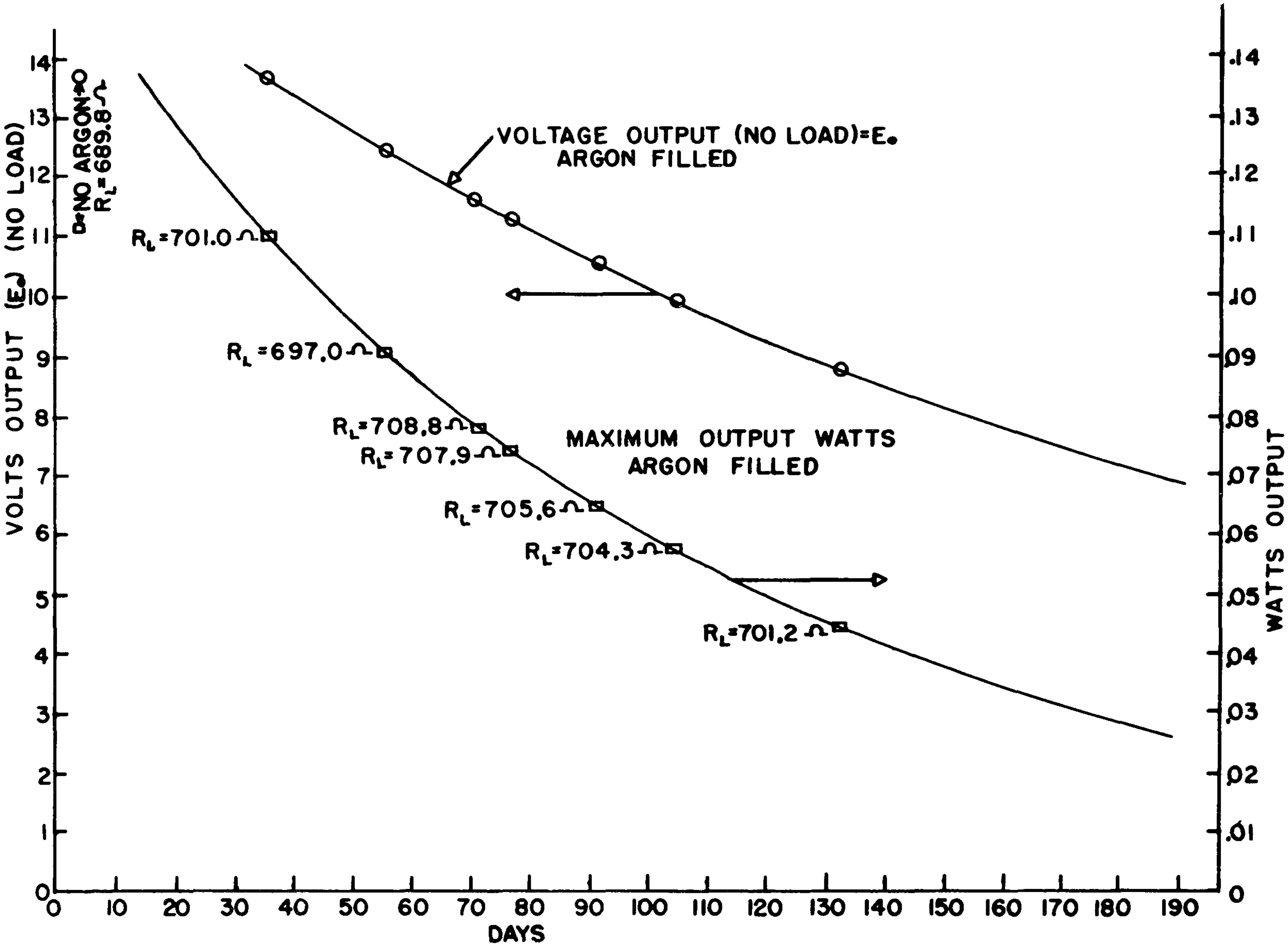


FIGURE 26 - NUCLEAR BATTERY PERFORMANCE

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OVERALL CONCLUSIONS

Thermoelectric generators powered by the heat of radioactive decay are feasible and have been successfully demonstrated. A generator has been constructed that weighs less than half a pound that will put out continuously for six months more than 50 milliwatts at 4.8 and 1.2 volts.

Isotopes which can be presently considered for use as heat sources are strontium-90, cerium-144, polonium-210, and thallium-204. The strontium and cerium are fission products and need only to be separated from waste material for use.Polonium and thallium are reactor-produced substances which could be manufactured for this use if the demand became large enough. Costs pf preparation and handling of these sources would be high, but facilities exist for their separation and encapsulation. Polonium is the easiest to handle and has the highest specific activity. but the half-life is too short to be used for any but short-term applications (Figure 27. Thallium, strontium and cerium are beta emitters, which means they have lower energies per disintergration and lower specific activities and are associated with a large gamma and brehmstrahlung spectrum. Thallium does have an advantage in that it can be encapsulated before irradiation, which makes it easier to handle. Strontium and cerium, being fission products, are already in existance and will require no reactor space.

Isotopes showing good nuclear characteristics that will probably become available in quantity in the near future are americanium-241 and cerium-242. These are by-products of reactor use and will only require modification of existing facilities and process streams for separation. Americanium has a very low specific activity. Cerium has a somewhat longer half-life than polonium.

The radium-228, actinium-227 complex could be made from radium-226 under present conditions. However, not very large quantities of radium are available, and special loading facilities would have to be arranged. This complex and the thallium have the advantage that they may be reinserted into a pile and be reactivated after decay of the heat source.

Plutonium-238, lead-210 and polonium-208 are three isotopes which have excellent nuclear characteristics for heat sources, but have no economical method of production at present. Future production methods leading to any of these should cause a reexamination of their feasibility for heat sources. Polonium-208 with a pure alpha and a moderate half-life would be almost ideal for short-lived producers while plutonium-238 with its weak gamma and long half-life and lead-210 with its polonium-210 daughters would make excellent standard sources.

The theory developed has been found very satisfactory for conductors and a similar theory without the assumption $de/_{dt} = 0$ made in MLM-CF 58-1-40 appendix could be developed for semiconductors. The theory was used to predict performance to be expected from our batteries and did so extremely well.

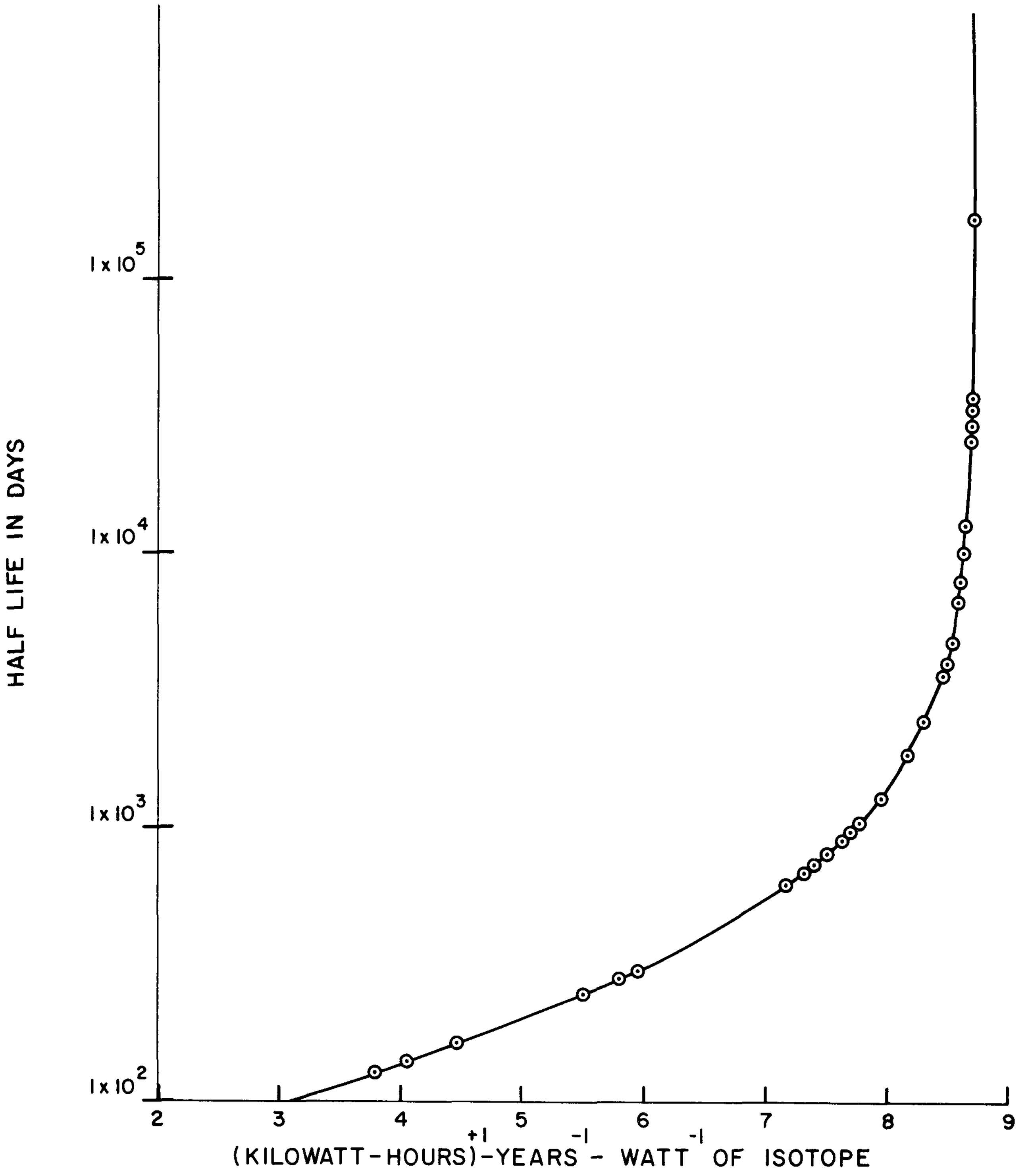

FIGURE 27 - POWER AVAILABLE FROM HALF-LIVES

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The Peltier heat absorbed in the hot junctions is, by Equation 1

$$nIT_1 \left.\frac{de}{dT}\right|_{T_1} \quad \text{watts} \tag{1a}$$

The Peltier heat generated in the cold junctions is

$$nIT_2 \left.\frac{de}{dT}\right|_{T_2} \quad \text{watts} \tag{2a}$$

The Joule heat generated internally is

$$I^2 R_g \quad \text{watts} \tag{3a}$$

The Thompson heat generated from hot to cold junctions is, by Equation 2.

$$nI \int_{T_1}^{T_2} T \frac{d^2e}{dT^2}\, dT \quad \text{watts} \tag{4a}$$

As described in the text the wattage of Equations (3a) and (4a) will be divided 50-50 between hot and cold junctions. Therefore, the net heat absorbed at the hot junction is

$$W_h = nIT_1 \left.\frac{de}{dT}\right|_{T_1} - \frac{I^2 R_g}{2} - \frac{nI}{2} \int_{T_1}^{T_2} T \frac{d^2e}{dT^2}\, dT \tag{5a}$$

and the heat generated at the cold junctions is

$$W_c = nIT_2 \left.\frac{de}{dT}\right|_{T_2} + \frac{I^2R_g}{2} + \frac{nI}{2} \int_{T_1}^{T_2} T \frac{d^2e}{dT^2}\, dT \tag{6a}$$

Subtracting Equation (5a) from (6a) gives

$$W_c - W_h = nI \Bigl(T_2 \left.\frac{de}{dT}\right|_{T_2} - T_1 \left.\frac{de}{dT}\right|_{T_1} + \int_{T_1}^{T_2} T \frac{d^2e}{dT^2}\, dT + I^2R_g \tag{7a}$$

By the method of integration by parts, $\int udv = uv - \int vdu$, and letting $u = \partial e/\partial T$ and $dv = dT$ the following identity can be established.

$$e\,(T_1) - e\,(T_2) \equiv T_1 \left.\frac{de}{dT}\right|_{T_1} - T_2 \left.\frac{de}{dT}\right|_{T_2} - \int_{T_1}^{T_2} T \frac{d^2e}{dT^2}\, dT \tag{8a}$$

where e can be any continuous function of T.

Substituting Equation (8a) in Equation (7a) gives

$$W_c - W_h = nI \left[e(T_2) - e(T_1) \right] + I^2 R_g \tag{9a}$$

$$= - IE_L + I^2 R_g \tag{10a}$$

$$= - W_O \tag{11a}$$

Equation (11a) is identical to Equation 3 and proves that Equation 1a, 2a, 3a and 4a satisfy the law of conservation of energy.

The Peltier and Thompson EMF's have been calculated for chromel-constantan couples from experimental data (Figure 4 in MLM-CF-57-10-31, "Nuclear Battery-Thermocouple Type, Third Quarterly Report, July 1, 1957 to September 30, 1957"). The results are shown in Figures 28 and 29. The Thompson EMF's are about one-tenth the Peltier.

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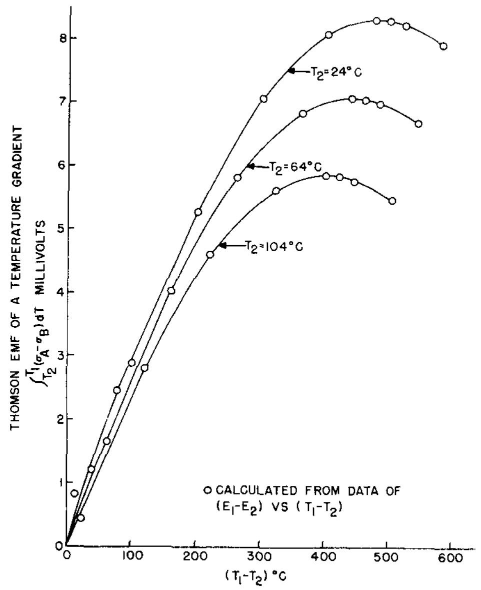

FIGURE 28 - THOMSON EMF DEVELOPED BY THE GRADIENT (T_1-T_2) BETWEEN TWO COUPLES OF CHROMEL-P-CONSTANTAN

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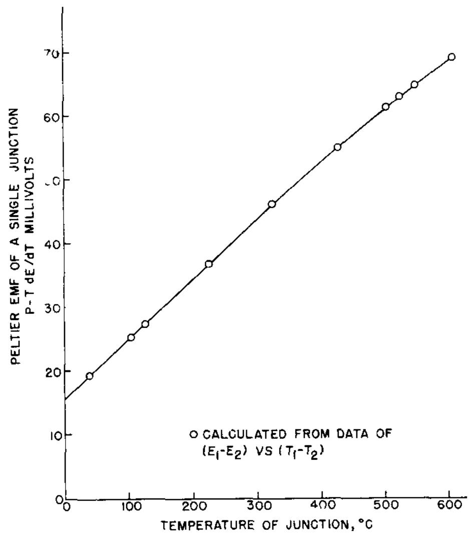

FIGURE 29 - PELTIER EMF OF A CHROMEL-P-CONSTANTAN JUNCTION AS A FUNCTION OF JUNCTION TEMPERATURE

$$\mathrm{Eff} = \frac{\mathrm{Eff}_O}{1 + 2\,\mathrm{Eff}_O\,(1+f_4)\left[\left(\frac{T_1}{\Delta T}\right)_L \left(\frac{R_L + R_g}{R_L}\right) - \frac{R_g}{2R_L}\right]}$$

$$= \frac{1}{\frac{1}{\mathrm{Eff}_O} + 2\,(1+t_4)\left[\left(\frac{T_1}{\Delta T}\right)_L \left(\frac{R_L + R_g}{R_L}\right) - \frac{R_g}{2R_L}\right]}$$

$$= \frac{1}{\frac{(R_L + R_g)^2\,(1+f)}{n^2\beta^2 R_L R_t \Delta T_O} + 2\,(1+f_4)\left[\left(\frac{T_1}{\Delta T}\right)_L \left(\frac{R_L + R_g}{R_L} - \frac{R_g}{2R_L}\right)\right]}$$

$$\frac{1}{\mathrm{Eff}} = \frac{(R_L + R_g)^2\,(1+f)}{n^2\beta^2 R_L R_t \Delta T_O} + 2\,(1+f_4)\left[\left(\frac{T_1}{\Delta T}\right)_L \left(1 + \frac{R_g}{R_L}\right) - \frac{R_g}{2R_L}\right]$$

$$= \frac{\left(R_L + 2R_g + \frac{R_g{}^2}{R_L}\right)(1+f)}{n^2\beta^2\, R_t\, \Delta T_O} + 2\,(1+f_4\left[\left(\frac{T_1}{\Delta T}\right)_L\left(1 + \frac{R_g}{R_L} - \frac{R_g}{2R_L}\right]\right.$$

$$\frac{\left(\partial \frac{1}{\mathrm{Eff}}\right)}{\partial R_L} = \frac{(1+f)\left(1 - \frac{R_g{}^2}{R_L{}^2}\right)}{n^2\,\beta^2 R_T\,\Delta T_O} + 2\,(1+f_4)\left[-\frac{R_g}{2R_L^2}\left(\frac{2T_1}{\Delta T_L} - 1\right)\right] = 0$$

$$= \frac{(1+f)\left(1 - \frac{R_g{}^2}{R_L{}^2}\right)}{n^2\,\beta^2\,R_t} - (1+f_4)\left[\frac{R_g}{R_L{}^2}\left(\frac{2\,\Delta T_o\,T_1}{\Delta T_L} - \Delta T_O\right)\right] = 0$$

$$= \frac{(1+f)\left(R_L{}^2 - R_g{}^2\right)}{n^2\,\beta^2\,R_t\,R_L{}^2} - \frac{(1+f_4)}{R_L{}^2}\left[R_g\left(\frac{2\,\Delta T_o T_1}{\Delta T_L} - \Delta T_O\right)\right]$$

Since $\Delta T_O \approx \Delta T_L$

$$R_L{}^2\,R_g{}^2 = \frac{n^2\beta^2 R_t}{(1+f)}\,(1+f_4)\,\left[R_g\,(2T_1 - \Delta T_O)\right]$$

$$R_L{}^2 = R_g{}^2 + \frac{n^2\,\beta^2\,R_t\,R_g{}^2\,(2T_1 - \Delta T_O)}{(1+f_1)\,(1+f_2)\,(1+f_3)\,R_g}$$

$$R_L = R_g \left[1 + \frac{n^2 \beta^2 R_t (2T_1 - \Delta T_0)}{(1 + f_1)(1 + f_2)(1 + f_3) R_g} \right]^{1/2}$$

$$= R_g \left[1 + \frac{n^2 \beta^2 R_t (T_1 + T_2)}{(1 + f_1)(1 + f_2)(1 + f_3) R_g} \right]^{1/2}$$

$$f_1 = \frac{R_t}{R_s' + R_c}$$

$$(1 + f_1) = \frac{R_s' + R_c + R_t}{R_s' + R_c}$$

$$f_3 = \frac{R_s' (R_t + R_c)}{(R_s' + R_t + R_c)(R_s + R_h)}$$

$$(1 + f_3) = \frac{(R_s' + R_t + R_c)(R_s + R_h) + R_s' (R_t + R_c)}{(R_s' + R_t + R_c)(R_s + R_h)}$$

$$\frac{R_t}{(1 + f_1)^2 (1 + f_3)^2} = \frac{R_t}{\left[\dfrac{R_s' (R_t + R_c) + (R_s' + R_t + R_c)(R_s + R_h)}{(R_s' + R_t + R_c)(R_s + R_h)}\right]^2 \left[\dfrac{R_s' + R_c + R_t}{R_s' + R_c}\right]^2}$$

$$= \frac{R_t}{\left[\dfrac{R_s' (R_t + R_c) + (R_s' + R_t + R_c)(R_s + R_h)}{(R_s + R_h)\ (R_s' + R_c)}\right]^2}$$

$$= \frac{R_t}{\left[\dfrac{R_s' R_t + R_s' R_c + (R_s' + R_c)(R_s + R_h) + R_t (R_s + R_h)}{(R_s + R_h)(R_s' + R_c)}\right]^2}$$

$$= \frac{R_t}{\left[\dfrac{R_t (R_s' + R_s + R_h)}{(R_s + R_h)(R_s' + R_c)} + \dfrac{R_s' R_c + (R_s' + R_c)(R_s + R_h)}{(R_s + R_h)(R_s' + R_c)}\right]^2}$$

If we let

$$K_1 = \frac{R_s' + R_s + R_h}{(R_s + R_h)(R_s' + R_c)} \qquad \text{and} \qquad K_2 = \frac{R_s' R_c + (R_s' + R_c)(R_s + R_h)}{(R_s + R_h)(R_s' + R_c)}$$

Then

$$\frac{R_t}{(1 + f_1)^2 (1 + f_3)^2} = \frac{R_t}{(K_1 R_t + K_2)^2} = y$$

$$\frac{dy}{dR_t} = \frac{(K_1 R_t + K_2)^2 - R_t\, 2\, (K_1 R_t + K_2) K_1}{(K_1 R_t + K_2)^4} = 0$$

$$R_t = \frac{K_1 R_t + K_2}{2K_1} = \frac{R_t}{2} + \frac{K_2}{2K_1}$$

$$R_t = \frac{K_2}{K_1} = \frac{R_s' R_c + (R_s' + R_c)(R_s + R_h)}{(R_s + R_h)(R_s' + R_c)} \Bigg/ \frac{R_s' + R_s + R_h}{(R_s + R_h)(R_s' + R_c)}$$

$$= \frac{R_s' R_c + (R_s' + R_c)(R_s + R_h)}{(R_s' + R_s + R_h)}$$

$$= \frac{R_s' R_c + R_s' R_s + R_s' R_h + R_c R_s + R_c R_h}{R_s' + R_s + R_h}$$

$$W_O\ (t) \propto E^2{}_{(t)} \propto (\Delta T_{(t)})^2 \propto W_S{}^2\ (t)$$

and

$$W_{S(t)} = W_S\ (t = o)\quad e^{-\lambda t}$$

$$W_{O\ (t)} = W_{O\ (t = o)}\ e^{-2\lambda t}$$

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