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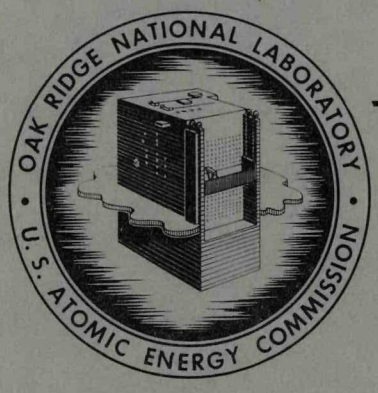
# A STUDY OF THULIUM-171 AS A POWER SOURCE FOR CIRCULATORY SUPPORT SYSTEMS

J. K. Poggenburg

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ISOTOPES DEVELOPMENT CENTER

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J. K. Poggenburg

Isotopes Division

MAY 1969



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CONTENTS

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J. K. Poggenburg


## ABSTRACT

Thulium-171, which has been proposed as a candidate for use as a power source in circulatory support systems because of its apparent low shield weight requirements and potential availability, was studied and compared with another candidate, $^{238}Pu$. One of the desirable features of $^{238}Pu$ for this application is its low dose rate of 35 mrem/hr at 10 cm from the center line of a 1-lb source. The initial amount of $^{170}Tm$ activity in a $^{171}Tm$ source, based on a total source and shield weight allowance of 1 lb, would have to be in the ratio of $10^{-4}$ to $10^{-5}$ for the dose rate to be comparable to that for the $^{238}Pu$ source. The technical and economic factors associated with producing a $^{171}Tm$ product which meets this requirement were considered for each of three possible reactor target materials: natural erbium, natural thulium, and enriched $^{170}Er$. The limited production capabilities and extremely high estimated cost – exceeding $30,000 per watt – led to the conclusion that $^{171}Tm$ is not a practicable power source for large-scale use in the Artificial Heart Program.


## INTRODUCTION

At present only two sources of power, the biological fuel cell and the radioisotopic heat source, appear capable of being totally implanted within the human body for circulatory support systems. While a biological fuel cell would be an ideal source of power, the limited technology makes its realization a future possibility. For the interim, radioisotopic heat sources could meet the requirements of an implantable power source. From studies[1] sponsored by the National Heart Institute, it was concluded that a circulatory pumping power of 2 to 5 W will provide reasonable allowance for normal patient activity. If a 15% conversion efficiency is assumed, the thermal source must supply ~30 W.[2] The projection for annual patient load is of the order of 10,000, which implies the need for a total annual power of ~300 $kW_{th}$. For a short-lived isotope like $^{171}Tm$, the annual requirement would be 600 $kW_{th}$ to allow for radioactive decay loses.

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[1] Hittman Associates, Inc., Final Summary Report on Six Studies Basic to the Consideration of the Artificial Heart Program, CFSTI, PB 173 483.

[2] D. W. Cole, W. E. Mott, and L. A. Sagan, "Factors Relating to the Application of Radioisotopes to Circulatory Support Systems," Intersociety Energy Conversion Engineering Conference, Boulder, Colo., August 13-16, 1968 (to be published by IEEE).

Criteria for choice of an implantable isotopic power source in a circulatory support system include small size, light shielding, a maximum weight of 1 lb for the source and shield together, minimum radiation exposure to the host and immediate associates, high integrity, and availability in quantities commensurate with the projected demand at a reasonable cost. Plutonium-238, an alpha emitter with a half-life of 87.4 y, is a prime candidate because of its low shield weight requirements, practical experience in its application as a power source for aerospace missions, and future availability at a cost projected to be ~$500 per thermal watt. Thulium-171, a beta emitter with a half-life of 1.9 y, has been proposed as a candidate for use in circulatory support systems because of its apparent low shield weight requirements and potential availability; however, no production experience is available to define product quality, production rates, or product cost.

This report contains the results of a study of $^{171}Tm$ that was made to evaluate the feasibility of its use in a circulatory support system. In this study the properties of $^{238}Pu$ served as the criteria for radiation exposure, shield weight, and total heat source weight. The scope of the study includes two principal factors, the determination of product quality requirements for $^{171}Tm$ based on a comparison of radiation characteristics with those of $^{238}Pu$ and the economic factors of large-scale $^{171}Tm$ production.

## RADIATION CHARACTERISTICS AND PRODUCT SPECIFICATIONS

The total weight of an isotopic power source and its associated shielding is an important, if not overriding, consideration when human implantation is involved. For such an application a maximum source weight of 1 lb has been established as a criterion. This stringent limitation on total source and shield weight indicates the use of isotopes such as $^{171}Tm$ or $^{238}Pu$ which have low radiation intensities. The unavoidable presence of $^{170}Tm$ contamination in $^{171}Tm$ produced by the reactor irradiation of either thulium or erbium targets affects the production rate and the cost of $^{171}Tm$ product with suitable radiation properties, and this section discusses the limits of $^{170}Tm$ contamination permissible in $^{171}Tm$ for human implantation.

In addition to its low radiation intensity, another advantage of $^{171}Tm$ is the existence of a suitable fuel form, $Tm_2O_3$, which has a very low biological toxicity.[3] The oxide is stable, melts at 2380°C, and, by analogy with other rare-earth oxides, should be compatible with several source container materials. A $^{171}Tm_2O_3$ source has a maximum possible specific power of 0.175 W/g and, on the basis of a density of 8.0 g/cm$^3$ (90% of theoretical density), a power density of 1.40 W/cm$^3$. In addition, the $Yb_2O_3$ decay product is a rare earth and is chemically similar to the parent. However, the radiation properties of $^{171}Tm$ are dominated by $^{170}Tm$ produced

[3] R. J. Everett, Radiological Health Aspects of Thulium-170, -171 Oxide, SC-RR-66-2679 (February 1967).

concomitantly in the product during reactor irradiation of either erbium or thulium targets (discussed in "Evaluation of $^{171}$Tm Production Methods," this report).

Thulium-171 has a half-life of 1.9 y and decays 98% to ground state by beta emission with a maximum energy of 0.097 MeV and 2% to an excited state of 0.067 MeV. The gamma transition to ground state is highly converted, and as a result photon emission occurs only in ~0.2% of the total disintegrations. The 0.067-MeV photon is just above the K-absorption edge of thulium (0.059 MeV) and therefore is readily self-absorbed. Thulium-170 has a half-life of 128.5 d and decays by beta emission with a maximum energy of 0.97 MeV. It has a 23% branching to an excited state at 0.084 MeV, and although the 0.084-MeV gamma transition is highly converted, photon emission occurs in ~3% of the disintegrations. A more energetic and more intense flux of bremsstrahlung photons results from the 0.97-MeV beta. Due to the effect of these radiations (see Table 1) on dose rates, $^{170}$Tm contamination is the controlling factor in shielding a $^{171}$Tm source even though $^{170}$Tm represents only $10^{-5}$ to $10^{-6}$ of the $^{171}$Tm disintegration rate. The amount of $^{170}$Tm which can be tolerated in a source must be specified, because this limit determines the cost and capabilities of producing a suitable $^{171}$Tm product. A practical basis for setting the $^{170}$Tm limit is the stipulation that a $^{171}$Tm power source should not yield a dose rate greatly exceeding that of $^{238}$Pu having the same total source and shield weight.

Table 1. Properties of Thulium Nuclides

| | $^{171}$Tm | $^{170}$Tm |
|---|---|---|
| Half-life | 1.9 y | 128.5 d |
| Radiations, MeV | | |
| Beta emission, $E_{max}$ | 0.097 (98%)<br>0.030 ( 2%) | 0.970 (77%)<br>0.886 (23%) |
| Photon emission | Bremsstrahlung<br>0.067 gamma (0.2%)<br>x rays | Bremsstrahlung<br>0.084 gamma (3%)<br>x rays |
| Specific power of pure nuclide, W/g | 0.2 | 11.6 |

The composition of the $^{238}$Pu used as the basis of this study is 81% $^{238}$Pu containing 1.2 ppm $^{236}$Pu that would be used for this type of application* (see Appendix A). Plutonium-238 combines a relatively high specific power,

*A 92% $^{238}$Pu product has been produced containing only 0.3 ppm $^{236}$Pu.

0.56 W/g, with a long half-life, 87.4 y, which allows a useful lifetime in power sources of greater than 10 years. The natural radioactivity of the pure isotope consists primarily of alpha radiation having an average energy of ~5.5 MeV. Accompanying this alpha radiation is characteristic gamma radiation (see Table XIV, Appendix A). However, the decay daughters of $^{236}Pu$ introduce penetrating gamma radiation which must be considered. An increasing dose rate with time is due to the buildup of daughters from $^{236}Pu$, especially $^{208}Tl$. Less than 2% of the dose rate comes from nuclides other than $^{238}Pu$ and the daughters of $^{236}Pu$. The neutron emission rate is 2755 neutrons $sec^{-1}$ $g^{-1}$ on a total plutonium basis, or 3410 neutrons $sec^{-1}$ $g^{-1}$ on a pure $^{238}Pu$ basis. A relative biological effectiveness (RBE) value of 10 is used to convert rad dose rates to rem dose rates for the neutron emission.

All dose rate comparisons which follow are made on the basis of a source geometry of a right circular cylinder with height equal to diameter, encapsulation in rhenium, and uniform tungsten shielding on sides and ends. Epsilon-phase plutonium metal was assumed with a density of 16.51 $g/cm^3$. A void volume equal to the plutonium volume was assumed to allow for collection of helium from alpha decay without excessive pressure buildup in the source. Dose rates are calculated at a point on the midplane 10 cm from the center line. The detailed source and fuel characteristics for the comparisons are given in Tables 2 and 3.

The contribution of $^{170}Tm$ bremsstrahlung to the dose rate expected from a 60-$W_{th}$ $^{171}Tm$ source is illustrated in Fig. 1. The data for the curves in this figure were calculated by H. H. Van Tuyl, Pacific Northwest Laboratory, and are given in Appendix A. The reliability of bremsstrahlung calculations for $^{170}Tm$ was tested experimentally at Savannah River Laboratory,[4] and it was found that the calculational techniques can predict actual dose rates reasonably well, but tend to overestimate them by about 20%.

For the 1-lb case in Table 2, $^{238}Pu$ imparts a dose equivalent to an average dose rate of 35 mrem/hr at 10 cm integrated over a 10-year mission. A 60-W $^{171}Tm$ source of equivalent average dose rate for a 2-year mission has a $^{170}Tm/^{171}Tm$ activity ratio of $3.4 \times 10^{-6}$ at the beginning of service. The initial dose rates at 10 cm for the $^{238}Pu$ and $^{171}Tm$ sources would be 21 and 143 mrem/hr, respectively.

While the 1-lb case is emphasized in this report, dose rates have also been considered for other source weights (see Table 2). If, in the development of an implantable power source, weight is a more important constraint than dose rate, then it would be possible with a $^{238}Pu$ source to reduce the weight from 1 to 1/4 lb while increasing the mission average dose rate from 35 to 44 mrem/hr. The 1-lb source is a minimum weight system for $^{171}Tm$ consisting of 370 g of $Tm_2O_3$ and 84 g of rhenium as the container. A 1-lb $^{171}Tm$ source giving a mission average dose equivalent to the 1/4-lb $^{238}Pu$ source would have a maximum $^{170}Tm/^{171}Tm$ activity ratio of $4.4 \times 10^{-6}$.

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[4] S. M. Sanders, Jr., W. J. Kerrigan, and E. L. Albenesius, Radiation Shielding for Small Power Sources of $^{170}Tm$, $^{171}Tm$, DP-1158 (January 1969).

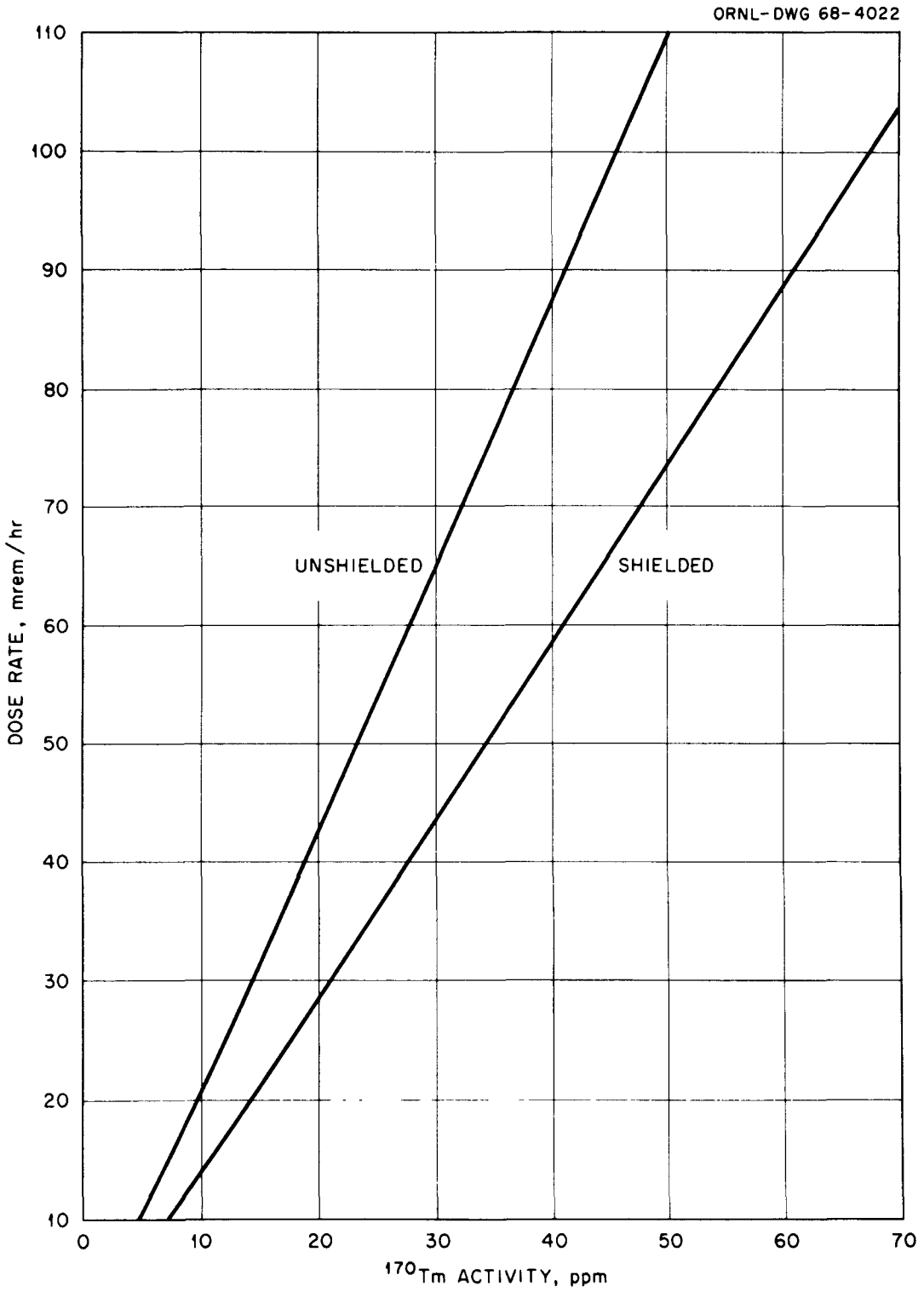


Fig. 1. Dose Rate as a Function of $^{170}Tm$ Content in a 60-$W_{th}$ $^{171}Tm_2O_3$ Source. Unshielded Case: 0.2-cm Re Liner; Shielded Case: 0.2-cm Re Liner Plus U Shield.

Table 2. Source Characteristics for Various Total Weight Allowances

Minimum practical source weights
Case A: 0.256 lb for $^{238}Pu$; 1 lb for $^{171}Tm$
Case B: 1.00 lb for each
Case C: 2.00 lb for each

| | $^{238}Pu$ | $^{171}Tm$ |
|---|---|---|
| Initial power, $W_{th}$ | 30 | 60 |
| Mission life, years | 10 | 2 |
| Fuel form | Metal | $^{171}Tm_2O_3$ |
| Fuel weight, lb, all cases | 0.148 | 0.815 |
| Capsule weight, lb | | |
| Cases A and B | 0.108 | 0.185 |
| Case C | 0.108 | 0.671 |
| Shield weight, lb | | |
| Case A | None | None |
| Case B | 0.744 | None |
| Case C | 1.744 | 0.514 |
| Dose rate,[a,b] mrem/hr | | |
| Case A | 44 | 44 |
| Case B | 35 | 35 |
| Case C | 29 | 29 |
| $^{170}Tm/^{171}Tm$ activity ratio | | |
| Case A | $4.4 \times 10^{-6}$ | |
| Case B | $3.4 \times 10^{-6}$ | |
| Case C | $81 \times 10^{-6}$ | |

[a] Mission average dose rate at a point on the midplane of the source 10 cm from the center line.

[b] See Appendix B.

Table 3. Fuel Characteristics

| Fuel | Purity | Density |
|---|---|---|
| Plutonium[a,b] | 81 wt % $^{238}Pu$[c] | 16.51 g/cm$^3$ |
| Thulium | 95% $Tm_2O_3$ (5% $Yb_2O_3$) | 90% of theoretical |

[a] Bomb-reduced [Progress Report on Refined $^{238}Pu$, CMB-1370 (Nov. 22, 1967)], $\epsilon$-phase metal.

[b] Neutron emission rate: 2755 neutrons $sec^{-1}$ $g^{-1}$ of Pu.

[c] Isotopic composition (wt %): $^{239}Pu$, 15; $^{240}Pu$, 2.9; $^{241}Pu$, 0.8; $^{242}Pu$, 0.1; and $^{236}Pu$, $1.2 \times 10^{-4}$.

If a shielded-source weight of 2 lb is permissible, the mission average dose rate for $^{238}Pu$ would be 29 mrem/hr. A shielded $^{171}Tm$ source with similar weight and a $^{170}Tm/^{171}Tm$ activity ratio of $3.4 \times 10^{-6}$ would produce a mission average dose rate of 1.2 mrem/hr. On the other hand, the higher weight allowance could be used to relax the $^{171}Tm$ purity requirement, and a $^{171}Tm$ source with an equivalent mission average dose rate of 29 mrem/hr would have a $^{170}Tm/^{171}Tm$ activity ratio of $81 \times 10^{-6}$.

The above analysis indicates that the $^{170}Tm/^{171}Tm$ activity ratio could range from $3.4 \times 10^{-6}$ based on the 1-lb shielded $^{238}Pu$ case to $81 \times 10^{-6}$ based on an unlikely 2-lb shielded $^{238}Pu$ case. Therefore in the following section production costs for $^{171}Tm$ are considered over this range of activity ratios.

## EVALUATION OF $^{171}Tm$ PRODUCTION METHODS

### Preliminary Considerations and Assumptions

#### Nuclear Reactions and Cross Sections

Thulium-171 can be produced in a nuclear reactor by three methods. One method consists of irradiating natural erbium containing 14.88% $^{170}Er$ which captures a neutron and forms $^{171}Er$. Erbium-171 decays to $^{171}Tm$ by beta emission with a half-life of 7.5 h:

$$^{170}Er \xrightarrow{n,\gamma} {}^{171}Er \xrightarrow[7.5\ h]{\beta^-} {}^{171}Tm$$

The second method proceeds by the same reaction using isotopically enriched $^{170}Er$. In the third method, made possible by favorable cross sections for neutron capture, natural $^{169}Tm$ forms $^{171}Tm$ by successive capture of two neutrons:

$$^{169}Tm \xrightarrow{n,\gamma} {}^{170}Tm \xrightarrow{n,\gamma} {}^{171}Tm$$

Technical and economic problems are associated with each of these production methods in obtaining a $^{171}Tm$ product of desired purity; this section examines these problems in detail.

The nuclear reactions of significance in the three production methods are shown in Table 4. Stable nuclides are underscored, and natural abundances are given. The reaction cross sections used in the study are listed in Table 5.

In addition to the principal reaction [$^{170}Er(n,\gamma)$], in the irradiation of natural erbium the $^{168}Er(n,\gamma)^{169}Er$ reaction is significant because $^{169}Er$ decays to $^{169}Tm$, which captures a neutron to become $^{170}Tm$. The $^{167}Er(n,\gamma)^{168}Er$ reaction is important because of its large cross section, which seriously affects neutron consumption. The unknown cross section for the $^{169}Er(n,\gamma)^{170}Er$ reaction is important only in the production yield of $^{171}Tm$ from erbium, because $^{171}Tm$ production is increased by this reaction while $^{170}Tm$ production is decreased. It would have to be at least 1000 barns in order for half the $^{169}Er$ to capture a neutron rather than to decay to $^{169}Tm$. The $^{171}Er(n,\gamma)^{172}Er$ reaction leads to a decrease in $^{171}Tm$ production from erbium, but a 10% loss by this route would require a cross section of ~3000 barns, whereas an estimate based on an experiment in which $^{172}Tm$ was produced by reactor irradiation of $^{170}Er$ indicated an effective cross section of only ~250 barns, which would result in a $^{171}Tm$ loss of <1%. The fast-neutron reaction $^{171}Tm(n,2n)^{170}Tm$ is important because it places a lower limit on the $^{170}Tm/^{171}Tm$ ratio which is attained. The $^{170}Yb(n,p)^{170}Tm$ reaction appears unimportant based on its estimated cross section. The cross sections for the ytterbium isotopes are of concern only in the general neutron economy.

## Basic Reactor Considerations

To achieve satisfactory rates of production of $^{171}Tm$ from either erbium or thulium requires a neutron flux $\geq 10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$. Since such a high flux is not obtained in commercial power reactors, special reactors are necessary for $^{171}Tm$ production. In order to calculate production data, the production capabilities and cost of a hypothetical 1000-MW reactor are considered. Capital costs for such a reactor have been projected at about $100 million, and annual operating costs at about $15 million.[5] A 1000-MW reactor burns ~1 kg of $^{235}U$ per day at a cost of $12,500 per day. Since the cost of fuel is about one-third the total cost and the

[5] B. I. Spinrad, "Limitations of Steady State, High Flux Reactors, Current and Future," p. 316 in Proceedings of the U. S. Atomic Energy Commission – European Nuclear Energy Agency Seminar, September 19-23, 1966, Santa Fe, Mexico. Intense Neutron Sources, Conf 660-925.

Table 4. Nuclear Reactions Involved in the Production of $^{171}Tm$ from Erbium or Thulium

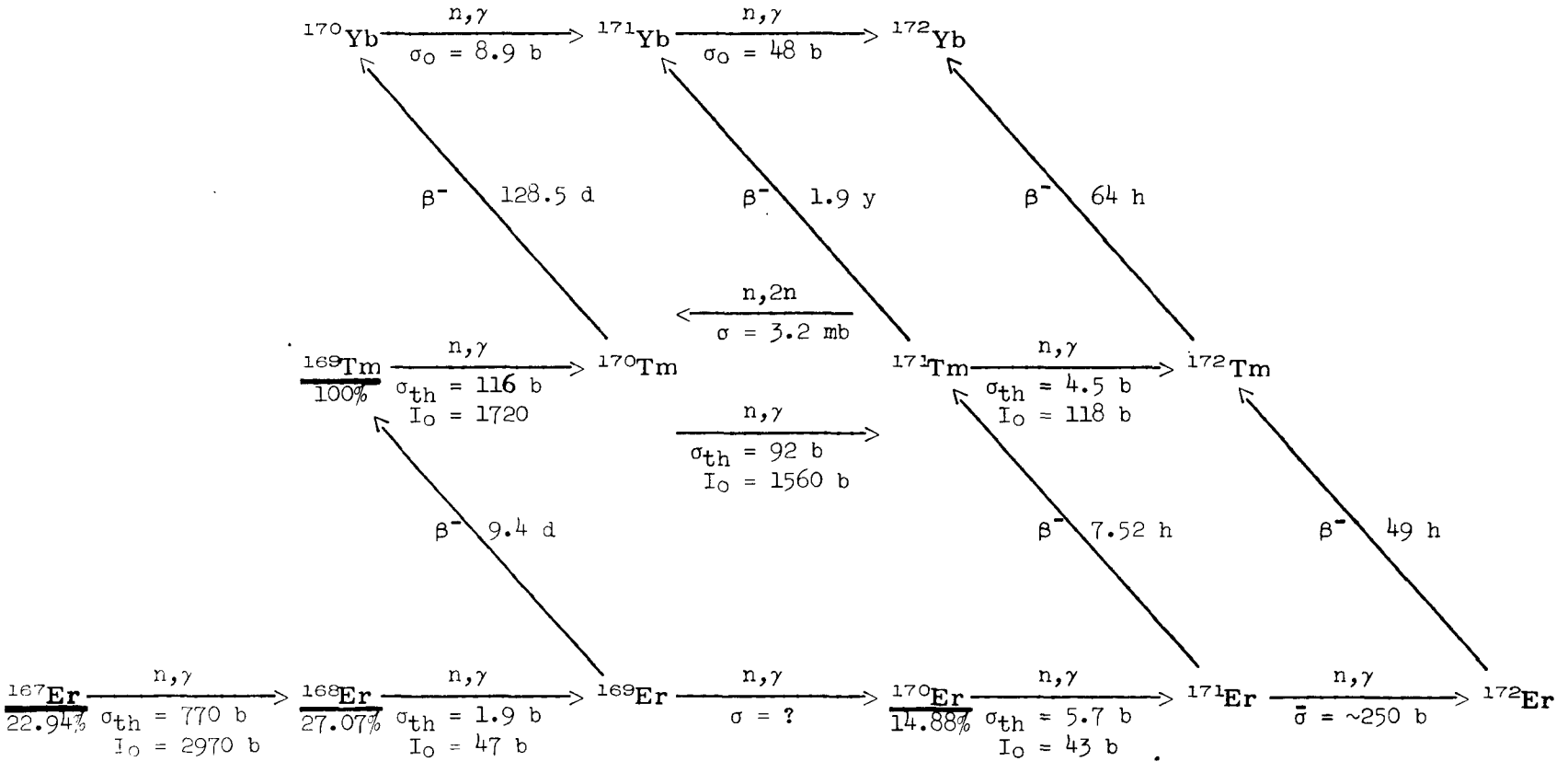

Table 5. Neutron Cross Sections for Thulium-171 Production

| Reaction | Cross Section, barns | | | Method of Determination |
|---|---|---|---|---|
| | Thermal | Resonance Integral | Other | |
| $^{168}Tm(n,\gamma)^{170}Tm$ | 116 ± 3 | 1720 ± 40 | | Mass spectrometry[a] |
| $^{170}Tm(n,\gamma)^{171}Tm$ | 92 ± 4 | 1560 ± 60 | | Mass spectrometry[a] |
| $^{171}Tm(n,\gamma)^{172}Tm$ | 4.5 ± 0.2 | 118 ± 6 | | Mass spectrometry[a] and activation analysis |
| $^{167}Er(n,\gamma)^{168}Er$ | 770 ± 40 | 2970 ± 70 | | Mass spectrometry[a] |
| $^{168}Er(n,\gamma)^{169}Er$ | 1.90 ± 0.03 | 47 ± 5 | | Activation analysis[a] |
| $^{170}Er(n,\gamma)^{171}Er$ | 5.70 ± 0.15 | 43 ± 5 | | Activation analysis[a] |
| $^{171}Er(n,\gamma)^{172}Er$ | | | ~250 (effective) | Experimental estimate,[b] reactor spectrum |
| $^{170}Yb(n,\gamma)^{171}Yb$ | | | 8.9 ± 1[c,d,e] | |
| $^{171}Yb(n,\gamma)^{172}Yb$ | | | 48 ± 3[c,d,e,] | |
| $^{172}Yb(n,\gamma)^{173}Yb$ | | | ~0.25[c,d,e] | |
| $^{171}Tm(n,2n)^{170}Tm$ | | | $4.8 \times 10^{-3}$[f] | Calculated[g] |
| $^{170}Yb(n,p)^{170}Tm$ | | | $0.02 \times 10^{-3}$[f] | Estimated[h] |

[a] Unpublished data of R. E. Lewis, derived from monitored irradiations of isotopically enriched samples with and without 40-mil cadmium shields. Limits shown give the statistical precision indicated by the standard deviation from the mean of a number of measurements.

[b] P. G. Hansen et al., Nucl Phys. 71: 481 (1965).

[c] M. D. Goldberg et al., Neutron Cross Sections, BNL-325, Supp. 2 (1966).

[d] S. F. Mughabghab, Brookhaven National Laboratory, private communication, Feb. 19, 1968.

[e] Neutron energy: 2200 m/sec.

[f] Fast neutron.

[g] S. Pearlstein, Nucl. Data A3: 327 (1967).

[h] J. C. Roy and J. J. Hawton, Table of Estimated Cross Sections for (n,p), (n,α), and (n,2n) Reactions in a Fission Neutron Spectrum, AECL-1181 (December 1960).

ratio of neutrons released per neutron absorbed by $^{235}U$ is 2.08, the cost for 1 g of neutrons is about $7000. More than half these neutrons either will be absorbed by the structural components of the reactor or will leak out of the reactor, so that the cost per gram of productive neutrons will be in the range of $15,000 to $20,000.

The power output of a reactor is proportional to the fission rate of the fuel, and the maximum is fixed by the reactor design. The fission rate is directly proportional to both the neutron flux and the quantity of fuel present. In order to increase the flux within the design power limitation, the density of fuel must be reduced. This scheme naturally results in shorter fuel cycles and increased operating expenses. Under conditions of lighter fuel loading the reactor becomes even more susceptible to flux depression brought about by neutron consumption in a target. Thus one is faced with the problem of counterbalancing the conflicting requirements of high-flux operation against efficient isotope production. The loading capacity of a reactor for isotope production is governed not only by volume limitations but also by the flux depression caused by the target itself.

Some general conclusions can be drawn on how first-order effects of neutron absorption act to limit the target capacity of a reactor. Of the 2.08 neutrons released per neutron absorbed in $^{235}U$, 1.08 are in excess of the number required to sustain the fission rate, and only half of these neutrons are available for productive absorption in the target. The probability for a neutron absorption is usually expressed in terms of a cross section $\sigma$ in units of barns ($10^{-24}$ $cm^2$). As an approximation, the neutron absorption rate in a material is given by the product of flux and cross section ($\Phi\sigma$). Clearly, if a flux is to be maintained, the neutron absorption rate in the target must not exceed the rate at which neutrons are made available for that purpose. If the flux is to be maintained at $10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$, then the absorption rate in a target of cross section $\sigma$ (barns) is about $\sigma A \times 10^{-4}$ neutron per day per mole of target,* where A is Avagadro's number, $6.02 \times 10^{23}$ atoms per mole. A 1000-MW reactor with a fission rate of about 1 kg of $^{235}U$ per day makes available for target consumption about 2A neutrons per day. The target loading capacity of the reactor is therefore about $2/\sigma \times 10^4$ moles. One sees that the load capacity of the reactor varies inversely as the target cross section, and inversely also as the operating flux.

## Radioisotopic Refinement of Product

The optimum rate for $^{171}Tm$ production in a 1000-MW reactor by any of the three methods does not necessarily result in a product with a $^{170}Tm/^{171}Tm$ activity ratio of $10^{-4}$ to $10^{-5}$; therefore methods of radioisotopic refinement to decrease the $^{170}Tm$ content of the product must be considered. Three possible methods are reactor irradiation, natural decay, and electromagnetic separation.

---

*Using the approximation 1 day = $10^5$ sec, the daily consumption at $10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ is $10^{-4}$ $\sigma$ neutron/atom of target or $\sigma A \times 10^{-4}$ neutron per day per mole.

The economic discussions become clearer by considering first the following formulations. The total cost of a product after processing is the initial cost plus the processing cost:

$$U_o\$_o = U_i\$_i + P_oU_o ,$$

where

$U_i$ = units in,
$U_o$ = units out,
$\$_i$ = cost per unit in,
$\$_o$ = cost per unit out,
$P_o$ = process cost per unit out.

The cost per unit product is, therefore,

$$\$_o = \$_i\,(U_i/U_o) + P_o ,$$

where $U_i/U_o$ is the reciprocal of the process yield. In comparisons involving a very large initial unit cost, the process yield can be much more important than the process cost per unit out.

Reactor Irradiation.– The $^{170}Tm/^{171}Tm$ ratio can be decreased by neutron irradiation in a reactor, since the capture cross section of $^{170}Tm$ is 24 times larger than the cross section for the burnout of $^{171}Tm$. To obtain significant advantage over natural decay, the neutron flux must be $>5 \times 10^{14}$ neutrons $cm^{-2}$ $sec^{-1}$. Furthermore, the effectiveness of reactor irradiation is reduced if $^{169}Tm$ is a substantial fraction of the product; thus reactor irradiation is most advantageous for refinement of a $^{171}Tm$ product chemically separated from an enriched $^{170}Er$ target. The product from irradiation of natural erbium contains comparable amounts of $^{169}Tm$ and $^{170}Tm$ from neutron capture of the lower erbium isotopes and decay of $^{169}Er$. In the case of production from $^{169}Tm$ this process would be equivalent to prolonging the production irradiation.

The limitations on reactor refinement due to the fast-neutron $^{171}Tm(n,2n)^{170}Tm$ reaction must be considered. The cross section for this reaction imposes the limit that the

$$\text{activity ratio of } ^{170}Tm/^{171}Tm \cong 2 \times 10^{-4}\,(\phi_f/\phi_{th}) ,$$

where $\phi_f/\phi_{th}$ is the ratio of the fission-spectrum average flux to the thermal neutron flux. This ratio could be as high as 0.5 for an exceptionally hard neutron spectrum, but a value of ~0.1 is more typical of reactor spectra. Even so, this reaction must be considered in any practical application. A hard spectrum is clearly undesirable for this refinement.

Irradiation in a flux of $1.5 \times 10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ for one year changes the $^{170}Tm/^{171}Tm$ activity ratio by a factor of ~0.002, while the $^{171}Tm$ is reduced to about one-half of its initial value. The cost of the one-year irradiation results in a factor-of-2 increase in the unit cost plus an incremental irradiation and processing cost in the range of $750 (±$150) per watt. The cost per watt of product for reducing the $^{170}Tm/^{171}Tm$ activity ratio by a factor of 500 is

$$\$_o = 2\ \$_i + 750 .$$

Natural Decay. – Since $^{170}Tm$ has a shorter half-life than $^{171}Tm$, the product can be aged to decrease the $^{170}Tm$ content. In one year $^{170}Tm$ decays to 0.140 of its original value, while $^{171}Tm$ is reduced to 0.694 of its initial value and the $^{170}Tm/^{171}Tm$ ratio changes by a factor of 0.20. The cost of refining $^{171}Tm$ by this method, if storage costs are negligible, increases the unit cost by a factor of 1.44 each year as a result of decay. In terms of the preceding formulation, the cost per watt of product for reducing the $^{170}Tm/^{171}Tm$ activity ratio by a factor of 5 is

$$\$_o = 1.44\ \$_i .$$

Electromagnetic Separations. – The cost of electromagnetic separations is difficult to estimate because no calutron refinement experience on a large scale is available with shielded facilities and radioactive operations. Tentative extrapolations can be made from calutron separations at ORNL of a few radioisotopes on the curie level. The amount of shielding required depends greatly on the anticipated $^{170}Tm$ contamination, and the length of a separation cycle could be determined solely by considerations of radiation safety. Calutron operating experience has shown that ~15% collection efficiency can be anticipated for each cycle. Overall recovery after processing and decay losses is estimated to be 65%. The separation factor for adjacent-mass rare earths is ~50, so that the $^{170}Tm/^{171}Tm$ ratio will be altered by a factor of ~0.02 after each pass. Since isotope separators are current-limited devices, the collection rate for the $^{171}Tm$ product is a linear function of its atom concentration in the charge; thus the lower the $^{171}Tm$ content, the higher the operating cost allocation per unit product, because $^{171}Tm$ must carry the full cost.

An estimate of the operating cost for a shielded calutron based on two-arc ion-source operation with 30-mA total ion current is $2500 (±20%) per day. This type of operation, though not current practice, appears to be feasible. Since 1 milliampere-day is required to collect approximately 1 mmole of material, ~2.5 g would be collected per day of operation if 50% operating time is assumed. If f is the weight fraction of $^{171}Tm$ in the feed, then the separation cost per product unit is $1000/f per gram, or $5000/f per watt. The unit cost of the charge material is multiplied by a factor of 1.5 to account for an assumed process loss of 35%. Refinement by electromagnetic separation would be limited to starting materials with a low $^{170}Tm/^{171}Tm$ ratio, where f is near unity. The cost per watt of product for reducing the $^{170}Tm/^{171}Tm$ activity ratio by a factor of 50 is

$$\$_o = 1.5\ \$_i + 5000/f .$$

Comparison of Refinement Techniques. — The radioisotopic refinement methods effecting a change in the $^{170}Tm/^{171}Tm$ ratio by a factor of 0.02, which is the factor expected for a single pass in a calutron, are compared in Table 6. Reactor costs are based on the assumption that the $^{171}Tm$ product output would be ~20 kW/year. Reactor irradiation is estimated to be the most economical method if the cost of the initial material is above $500 per watt. If the initial material cost is less than that, then natural decay would be more economical. For example, if the initial material costs $600/W, the product enriched by natural decay costs $1460/W, whereas the product of an irradiation costs $1370/W. For an initial cost of $400/W, the decay product costs $980/W, whereas the irradiation product costs $1080/W.

Table 6. Radioisotopic Refinement to Change the $^{170}Tm/^{171}Tm$ Ratio by a Factor of 0.02

| Method | $P_o$ Process Cost Per Watt of Product | $U_o/U_i$ Yield Factor | Process Time |
|---|---|---|---|
| Natural decay | Assumed negligible | 0.41 | 2.44 years |
| Reactor irradiation[a] | ~$500 | 0.69 | 0.62 years |
| Calutron | ~$5500 | ~0.65 | ~0.5 W/day[b] |

[a] Limited to material with low $^{169}Tm$ and $^{170}Tm$ content; flux ~$1.5 \times 10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$.

[b] Rate per machine.

## Production of $^{171}Tm$ from Natural Erbium

The production of $^{171}Tm$ from natural erbium (Table 7) proceeds by two reaction routes (underscored isotopes are stable):

$$\underline{\underline{^{170}Er}} \xrightarrow{n,\gamma} {}^{171}Er \xrightarrow{\beta^-} {}^{171}Tm$$

and

$$\underline{\underline{^{167}Er}} \xrightarrow{n,\gamma} \underline{\underline{^{168}Er}} \xrightarrow{n,\gamma} {}^{169}Er \xrightarrow{\beta^-} \underline{\underline{^{169}Tm}} \xrightarrow{n,\gamma} {}^{170}Tm \xrightarrow{n,\gamma} {}^{171}Tm$$

The first reaction route predominates at short exposures; with longer irradiation times the second reaction route is significant because of the larger natural abundances of $^{167}Er$ and $^{168}Er$ and because of the relatively large neutron-capture cross sections of $^{169}Tm$ and $^{170}Tm$. The very large cross section of $^{167}Er$ makes it significant in the second reaction route

and also places severe restrictions on reactor loading. The second reaction route produces $^{170}Tm$ contamination and causes the large $^{170}Tm/^{171}Tm$ activity ratio in the product, which makes radioisotopic refinement necessary.

Table 7. Isotopic Composition of Natural Erbium

| Mass No. | Amount, % |
|---|---|
| 162 | 0.136 |
| 164 | 1.56 |
| 166 | 33.4 |
| 167 | 22.9 |
| 168 | 27.1 |
| 170 | 14.9 |

The advantage of using natural erbium as a target material is its present availability in large quantities at comparatively low cost. The price for the oxide is ~$180/kg. Chemical separation of $^{171}Tm$ from the erbium target and ytterbium decay products would be required, probably by ion exchange, for which capital costs would be ~$3 million and annual operating costs about $0.5 million.

Calculation of the production of $^{171}Tm$ from natural erbium after a one-year irradiation in a flux $>10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ ($10^{15}$ thermal plus 10% resonance-energy neutron contribution) indicates that 10.6 neutrons are captured for every atom of $^{171}Tm$ obtained. The cost of $^{171}Tm$ calculated by using the estimate of $15,000 to $20,000 per gram of neutrons thus ranges from $4700 to $6200 per watt.

Estimation of the reactor loading capacity from the relation given previously ($2/\sigma \times 10^4$ moles) yields the figure 20 kg for natural erbium with cross section of 160 barns. Figure 2 shows a $^{171}Tm$ yield of 7% after one-year exposure at $10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$, and the annual output of $^{171}Tm$ is therefore only about 0.3 kW for a 1000-MW reactor, at a cost of $50,000/W. It would be advantageous to expose the natural erbium to a preliminary lower flux (~$5 \times 10^{14}$ neutrons $cm^{-2}$ $sec^{-1}$) to burn out $^{167}Er$ and reduce the target cross section, thereby increasing the loading capacity of the higher flux production reactor. By this scheme it should be possible to approach a production reactor output of 2 kW annually at a product cost of ~$7500/W. The product would, however, have a $^{170}Tm/^{171}Tm$ activity ratio of ~1, and further refinement would be necessary. To reach the $10^{-4}$ to $10^{-5}$ range of activity ratio, three calutron refinement stages would be required, at a cost of $35,000 to $45,000 per watt.

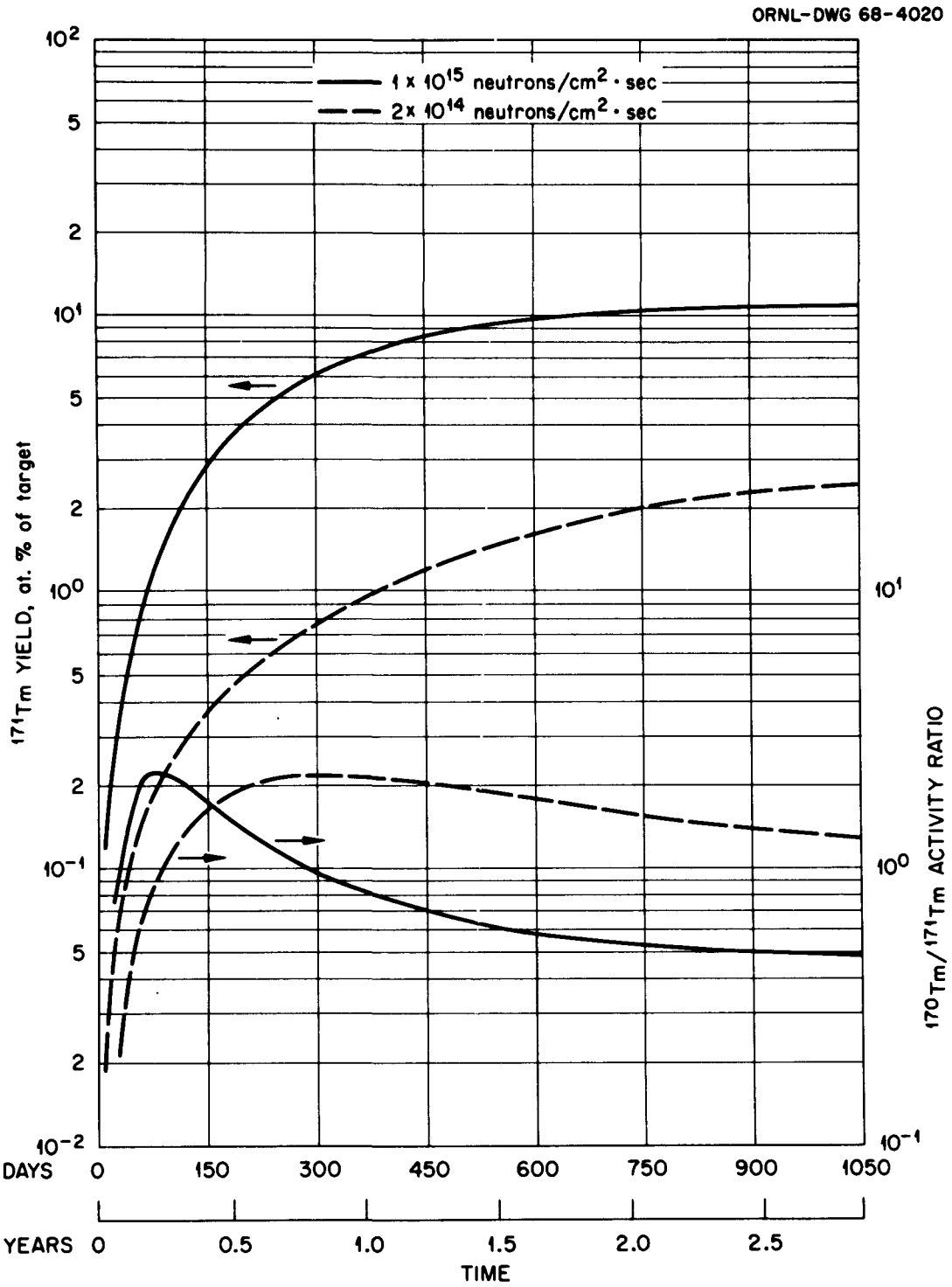


Fig. 2. Yield and Activity Ratio as a Function of Irradiation Time for Natural Erbium.

The variation of $^{171}Tm$ yield from natural erbium targets as a function of time and flux is shown in Fig. 2. This figure gives the atomic percent of $^{171}Tm$ and the $^{170}Tm/^{171}Tm$ activity ratio resulting from continuous irradiation of a natural erbium target at fluxes of $2 \times 10^{14}$ and $1 \times 10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$. A 10% contribution from resonance-energy neutron absorption is assumed. The abundance of thulium isotopes in the target after a one-year irradiation for the two cases is given below:

| Flux, neutrons $cm^{-2}$ $sec^{-1}$ | $2 \times 10^{14}$ | $1 \times 10^{15}$ |
|---|---|---|
| Thulium abundance in target, at. % | | |
| $^{169}Tm$ | 0.91 | 1.15 |
| $^{170}Tm$ | 0.37 | 1.07 |
| $^{171}Tm$ | 0.935 | 7.03 |
| Activity ratio, $^{170}Tm/^{171}Tm$ | 2.1 | 0.8 |
| Atomic ratio, $^{169}Tm/^{171}Tm$ | 0.97 | 0.16 |

If the thulium product from the flux of $10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ were chemically separated from the erbium target and irradiated again in the same flux, it would take ~1.5 years to attain a product with a $^{170}Tm/^{171}Tm$ activity ratio of $10^{-4}$ to $10^{-5}$. The yield of $^{171}Tm$ would then be one-third its initial value. For the reactor refinement method to be effective, the erbium content must be reduced to 0.01%. The separated erbium could then be blended with natural erbium and recycled as feed which would be low in $^{167}Er$ content and not require the suggested first-cycle low-flux irradiation. Eventually, the $^{170}Er$ content would be reduced to the point that further recycling would be impractical. It appears feasible that, by using at least two 1000-MW reactors and this recycled-target process, $^{171}Tm$ of requisite quality could be produced at the rate of 1 kW/year. Capital costs would thus be about $200 million for reactors, plus $3 million for a chemical separation plant. Annual operating costs would be ~$30 million for reactors and ~$0.5 million for the chemical separation plant. The cost of a suitable $^{171}Tm$ product would range from $30,000 to $40,000 per watt.

There is a possible variation on the production of $^{171}Tm$ from natural erbium. Since $^{170}Tm$ contamination results from the capture of a second neutron by the $^{169}Tm$ formed by the decay of $^{169}Er$, it is obvious that for short irradiations (e.g., 0.5 day at a flux of $10^{14}$ neutrons $cm^{-2}$ $sec^{-1}$) the $^{170}Tm$ contamination is extremely low. In principle, then, it is possible to irradiate natural erbium briefly at low flux and to separate chemically a $^{171}Tm$ product having $<10^{-5}$ parts of $^{170}Tm$ activity; however, $^{171}Tm$ would constitute $<10^{-3}$% of the natural erbium target. Because erbium and thulium are practically indistinquishable chemically, the

recovery of low concentrations of $^{171}Tm$ from the erbium target would require a multistage separation facility based on advanced technology, the cost of which would be prohibitive.

## Production of $^{171}Tm$ from Natural Thulium

Because of the high cross sections of both $^{169}Tm$ and $^{170}Tm$, it is possible to produce $^{171}Tm$ by a double neutron-capture reaction:

$$^{169}Tm \xrightarrow{n,\gamma} {}^{170}Tm \xrightarrow{n,\gamma} {}^{171}Tm$$

The main problems with this reaction are the need for long-term irradiations in order to deplete the $^{169}Tm$ and $^{170}Tm$ nuclides and the high flux that must be used to minimize loss through decay of $^{170}Tm$ during irradiation.

Thulium is one of the less abundant rare earths, and its commercial availability is presently limited. The latter drawback does not appear to be a serious problem, however, because potential production capacity could meet future demands. The present price of ~$3000 per kilogram of oxide may be expected to drop ultimately to about $1600 per kilogram upon greater demand.

Figure 3 illustrates the activity ratio attained as a function of time under continuous irradiation at various effective fluxes (i.e., under the assumption of no flux depression or self-shielding of the target). These corrections to the nominal flux will, in fact, be significant. Also shown in Fig. 3 is the limiting effect that a fast-flux component has on the attainable activity ratio. The effect of resonance neutron capture in decreasing the irradiation time required and increasing the yield is shown in Fig. 4. The activity ratio and corresponding percent yield of $^{171}Tm$ from the target are plotted as a function of time for continuous irradiation at an effective thermal flux of $10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ with assumed effective resonance flux components of 0, 1, and 10%. Because of the large resonance integrals of $^{169}Tm$ and $^{170}Tm$, a high resonance-energy flux is desirable. The self-shielding effect is also severe for the same reason, and target dilution is required to achieve a large effective contribution from the resonance flux and to minimize flux depression. The relation of the $^{171}Tm$ yield from the target to the desired activity ratio and to the effective flux at which it is produced is shown in Fig. 5.

The irradiated targets will require chemical processing to separate the thulium from the ytterbium decay products, probably by electrolytic reduction with sodium amalgam. Such a technique has been tested and found to be effective in rapidly removing ytterbium from thulium. This process would be considerably cheaper than ion exchange, and processing costs would be negligible compared with irradiation costs.

The reactor capacity for the initial natural thulium target ($\sigma \cong 100$ barns) is about 30 kg of thulium. The average cross section will decrease as $^{169}Tm$ and $^{170}Tm$ are converted to $^{171}Tm$, but the output based on the

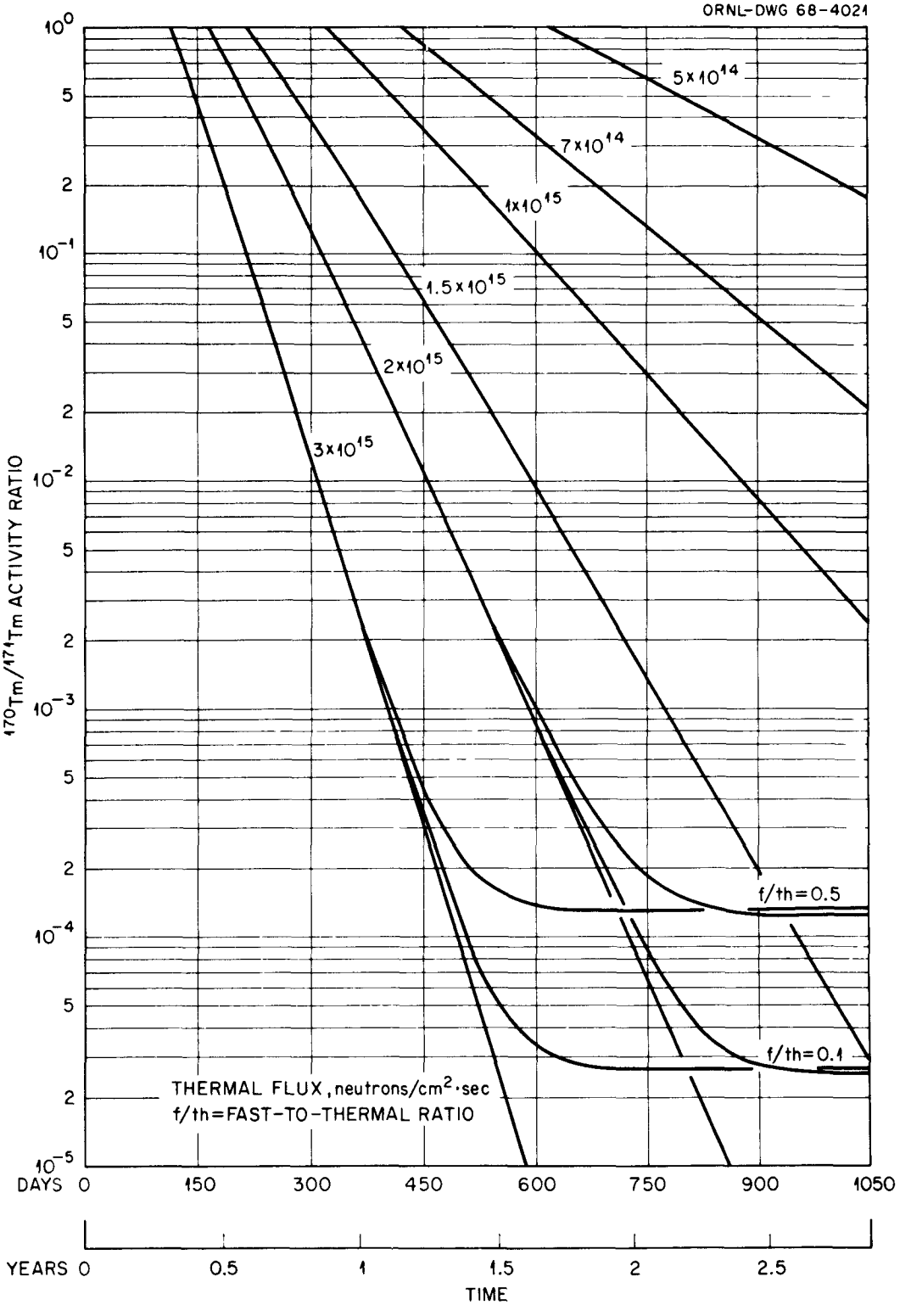


Fig. 3. Activity Ratio as a Function of Irradiation Time for Natural $^{169}Tm$.

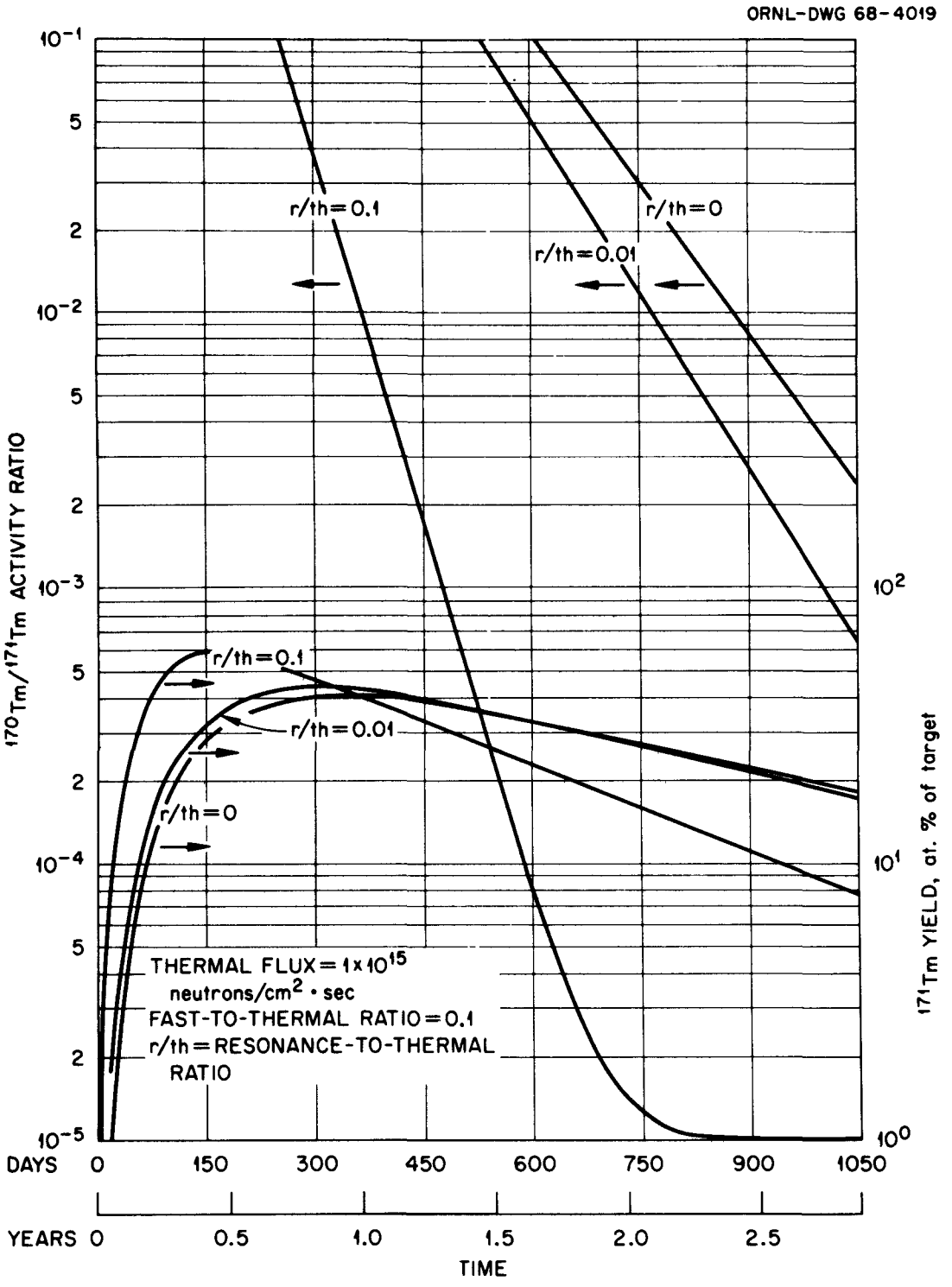


Fig. 4. Yield and Activity Ratio as a Function of Irradiation Time for Natural $^{169}Tm$.

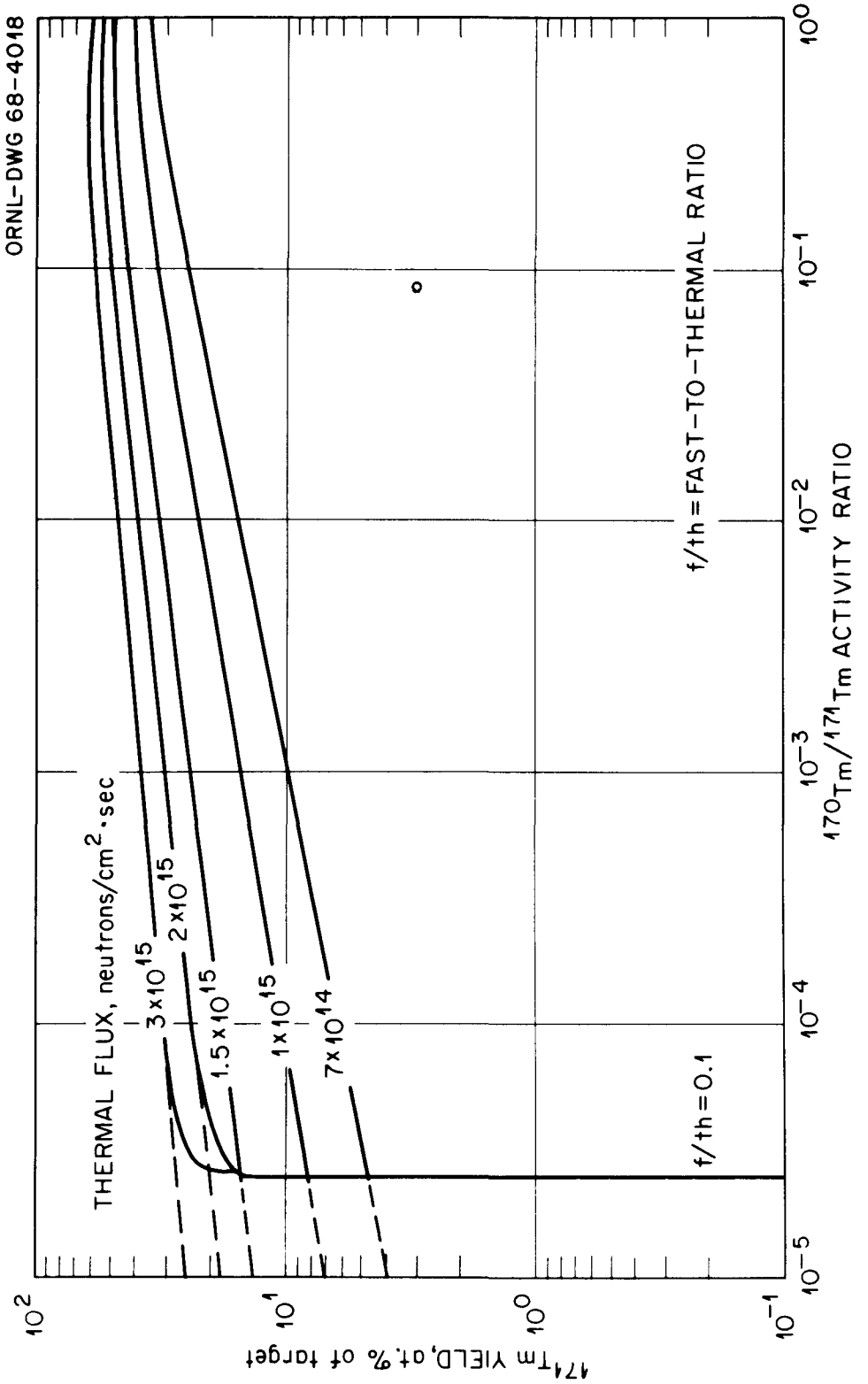


Fig. 5. Yield from Natural $^{169}$Tm as a Function of Activity Ratio at Various Thermal Fluxes and a Fixed Fast-to-Thermal Flux Ratio of 0.1.

30-kg loading at an effective flux of ~$10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ with about 10% yield in ~3.5 years is about 0.2 kW annually. If the flux is doubled, the loading capacity is halved, but the yield will be ~20% for an irradiation time of about 1.75 years and the $^{171}Tm$ output will increase to about 0.3 kW annually. Higher flux favors higher output, but this advantage is offset by shorter fuel cycles and increased costs. After a substantial fraction of $^{169}Tm$ and $^{170}Tm$ is burned out, the loading capacity is increased, but this gain must also be traded off against costs of target processing and fabrication. An annual output of ~0.25 kW of product $^{171}Tm$ from a single reactor operating at ~2 x $10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ appears to be feasible, with a price range of $50,000 to $75,000 per watt, based on annual operating expenses of $15 million for the reactor.

## Production of $^{171}Tm$ from Enriched $^{170}Er$

Obtaining $^{171}Tm$ from enriched $^{170}Er$ by the reaction

$$\underline{\underline{^{170}Er}} \xrightarrow{n,\gamma} {}^{171}Er \xrightarrow{\beta^-} {}^{171}Tm$$

is potentially the most economical and productive method, if the present limitations and costs of highly enriched target material can be substantially altered by technological breakthroughs in isotopic separations. While the prospect for such a development is not immediate, it offers the most promising means of substantially reducing costs and increasing production capacity.

To obtain a product with a $10^{-4}$ to $10^{-5}$ $^{170}Tm/^{171}Tm$ activity ratio, the target enrichment of $^{170}Er$ must be >99.99%. The best enrichment presently obtainable is only 96% from a single calutron pass, with a reasonable prospect for 99% enrichment; a two-stage calutron enrichment would be required with present technology. The requisite $^{170}Er$ target is not currently available and is, in fact, the central problem in this production scheme.

Study of neutron economy in the production of $^{171}Tm$ from $^{170}Er$ reveals that the number of neutrons required for each atom of $^{171}Tm$ formed ranges from slightly more than the theoretical minimum of one to about two, the number depending on the length of the irradiation cycle before chemical separation. This number yields a cost estimate of $450 to $600 per watt for irradiation costs based on neutron costs. The favorable neutron economy means that the target "payload" is highest for $^{170}Er$. With a 5.6-barn cross section the loading capacity of the reactor is about 600 kg at $10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$. With 90-day cycles the annual output from one reactor would be about 18 kW of $^{171}Tm$ of product quality, which leads to an irradiation cost estimate of ~$830 per watt based on assumed operating costs of $15 million per year. This is the largest reactor output among the three modes of production. Another advantage is that much lower fluxes can be tolerated, 5 x $10^{14}$ neutrons $cm^{-2}$ $sec^{-1}$, for example, but volume limitation rather than flux depression is the limiting factor in reactor capacity.

With the highly enriched $^{170}Er$ no further radioisotopic refinement is necessary, but chemical separation to recover the valuable target material is required. This production scheme would require the largest chemical separation facility, probably employing ion exchange, and capital costs may run above $5 million, with annual operation costing ~$1 million. With an 18-kW annual production, however, the unit chemical processing cost is about $55 per watt.

The annual $^{170}Er$ target requirements of a single 1000-MW reactor will be ~110 kg, by assuming ~10% loss in irradiation. Such requirements cannot reasonably be met with existing facilities for isotopic separations. With a two-stage enrichment using calutrons operating with two arcs at 15% process efficiency, 85% retention, and 30 mA total ion current, each machine will produce 0.58 g of $^{170}Er$ per day in the first refinement stage. For second-stage refinement, single-arc operation has been found to result in better mass separation than is obtained with two arcs: each machine will produce ~1.92 g of $^{170}Er$ per day. Thus the need for ~110 kg/year implies the need for ~160 machines for the second-stage refinement and ~760 for first-stage refinement. Since they would be in operation ~80% of the time, the number of machines required will be ~1150. No such capacity is available, and a rough estimate of capital costs for such a plant is $1 billion. A reasonable estimate for operating costs of a new facility is $600 per machine per day. If ~25% of the $^{170}Er$ is lost during second-stage enrichment, the target material would cost ~$1600 per gram or $8100 per watt of $^{171}Tm$ product.

The total $^{171}Tm$ product cost, per watt, resulting from irradiation of enriched $^{170}Er$ would be ~$9000, as detailed below:

| | |
|---|---|
| Enriched target cost | $8,100 |
| Chemical separation cost | 55 |
| Reactor irradiation cost | 830 |
| | $8,985/W |

There is no advantage in using $^{170}Er$ of single-stage enrichment. The $^{171}Tm$ product would have to be refined further by being irradiated for ~1.4 years at a flux of $>1.5 \times 10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ and the cost would be ~$14,000 per watt.

The foregoing estimates are based on an optimized system for large-scale production. The capacity for such production is nonexistent, and capital costs seem prohibitive if one acknowledges the interim role of radioisotopic power in the heart program. Production on a smaller, more conservative, scale would result in an increase in unit cost, probably by a factor of 2 or 3. Any method of increasing isotopic enrichment of $^{170}Er$ that may be lower in capital and operating costs could substantially improve the outlook for this method of production. At present there are no known compounds of erbium that are volatile under conditions other than vacuum and elevated temperatures that might afford a practical basis for isotopic separation.

## Comparison of Production Methods

The three production methods are summarized and compared in Table 8. The method presently offering the best reactor yield and price is that of recycling natural erbium. The price range for a product with a $^{170}Tm/^{171}Tm$ activity ratio of $10^{-4}$ to $10^{-5}$ is \$30,000 to \$40,000 per watt. The use of enriched $^{170}Er$ target holds the greatest promise for increased production capacity and prices under \$10,000 per watt. A major advance in technology for separating erbium isotopes could reduce the cost to a few thousand dollars per watt.

The use of one 1000-MW reactor was chosen for the basis of comparison, except for the natural erbium method, which requires at least two such reactors. These results are necessarily broad-range estimates. In order to narrow the range, more detailed study is required, with consideration of the characteristics of particular reactors, but the present study defines the obvious problems for large-scale production.

The annual requirement for $^{171}Tm$ was set at 600 kW. Even the most productive method developed in this report would require more than thirty 1000-MW reactors and a capital outlay of \$30 billion to meet this requirement. The production of $^{171}Tm$ from recycled natural erbium is technically feasible at present with a 1000-MW production reactor, but the 600-kW objective would require 600 such reactors.

## CONCLUSIONS

The present capacity for producing $^{171}Tm$ is severely limited by the fact that special reactors capable of fluxes in excess of $10^{15}$ neutrons $cm^{-2}sec^{-1}$ must be employed. In order to realize the low dose rates desired for the circulatory support system, the $^{170}Tm$ contamination in a heat source must be extremely low. The $^{170}Tm/^{171}Tm$ activity ratio must be in the range of $10^{-4}$ to $10^{-5}$ so that dose rates equivalent to those expected from a $^{238}Pu$ source will not be exceeded. The lowest cost range for a suitable $^{171}Tm$ product within present reach is \$30,000 to \$40,000 per watt using a recycled natural erbium target. Increased productive capacity and a price substantially under \$10,000 per watt could be attained if a significant breakthrough were made in producing highly enriched $^{170}Er$ cheaply. Even if this were achieved, the cost would greatly exceed current estimates of ~\$500/W for $^{238}Pu$. Because of both the limited capacity to produce a heat source product meeting stringent dose rate limitations and the high cost estimate of such a product, it is not practical to consider $^{171}Tm$ for large-scale use in the Artificial Heart Program.

Table 8. Comparison of $^{171}Tm$ Production Methods

| | Natural Erbium | Natural $^{169}Tm$ | Enriched $^{170}Er$ |
|---|---|---|---|
| | | Reactor Production | |
| Minimum facilities | At least two 1000-MW reactors | One 1000-MW reactor | One 1000-MW reactor |
| Procedures | Preliminary low flux to remove $^{167}Er$ or blending of recycled Er remaining after chemical separation of Tm<br><br>Irradiation at a flux of $>10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ for ~1 year to produce $^{171}Tm$<br><br>Chemical separation of Tm and recycle of Er target in first step<br><br>Reirradiation of Tm at a flux of $>10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ to remove $^{170}Tm$ | Irradiation at a flux of $>10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ for ~2.5 years; probable chemical processing and compaction of targets for final refinement stage | Flux of $<10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ (very high flux not required) |
| | | Target Material | |
| Requirements | ~180 kg/year | ~8 kg/year | ~110 kg/year of 99.99% enrichment |
| Availability | Unlimited commercial oxide | Potentially unlimited commercial oxide | Not available; separation plant required |
| | | Chemical Processing | |
| Method | Ion exchange separation of Er, Tm, and Yb | Sodium-amalgam extraction of Yb from Tm | Ion exchange separation of $^{170}Er$, Tm, and Yb |
| | | $^{171}Tm$ Product | |
| $^{170}Tm/^{171}Tm$ ratio | $10^{-4}$ to $10^{-5}$ | $10^{-4}$ to $10^{-5}$ | $10^{-4}$ to $10^{-5}$ |
| Production capacity, kW/1000 MWyr | ~1 | ~0.25 | ~18 |
| Cost range per W | $30,000-40,000 | $50,000-75,000 | ~$9,000-10,000 |
| | | Capital Costs,[a] millions | |
| Reactors | $200 | $100 | $ 100 |
| Chemical processing | 3 | 2 | 5 |
| Isotopic separations | ___ | ___ | 1000 |
| Total capital | $203 | $102 | $1105 |
| | | Annual Operating Costs,[a] millions | |
| Reactor | $ 30 | $ 15 | $ 15 |
| Target materials | 0.03 | 0.01 | 165 |
| Chemical processing | 0.5 | 0.4 | 1 |
| Total operating | $ 30.53 | $ 15.41 | $ 181 |

[a]Costs are approximate.

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# Appendix A

Unclassified Extraction from: Division of Isotope Development Programs, Quarterly Report, January - April 1968, edited by R. L. Moore, HNWL-781 (Confidential).

## DOSE RATE AND SHIELDING CALCULATIONS FOR ARTIFICIAL HEART POWER SOURCES

H. H. Van Tuyl

Calculations were made during the quarter to compare dose rates from promethium, plutonium, and thulium heat sources--all candidates for artificial heart application.

The initial calculations were made on the basis of beginning-of-life powers of 15, 30, and 50 W without regard to half-life or mission lifetimes. Therefore, to compare sources on an end-of-life basis, it would be necessary to use different beginning-of-life powers dependent on the nuclide and mission lifetime. Selected data for 30 W sources are presented as graphs of dose rate versus shield thickness, total weight, and total volume, (Figures 20-22). The first 0.1 cm of shield is rhenium to provide a good compatibility liner. The balance of the shield is either uranium or lithium hydride as appropriate to obtain maximum dose rate reduction for the added shield weight.

The principal conclusions from these curves are that a moderately low dose rate can be achieved with $^{238}Pu$ sources with low total weight, and comparable dose rates can be achieved with high quality $^{147}Pm$ or $^{171}Tm$ sources of slightly higher weights. If lower dose rates are required, however, substantially lower weights can be obtained by using either $^{147}Pm$ or $^{171}Tm$ sources since they yield radiations which are less penetrating.

The most recent calculations are based on an end-of-life power comparable to that from a $^{238}Pu$ source that was initially 30 W, but which has decayed for ten years. Thulium and promethium sources of twice the initial $^{238}Pu$ power would thus have mission lifetimes of about one half-life. Comparisons of the basis of equal end-of-life power are shown in Table XIII. Beginning-of-life power was 30 W for $^{238}Pu$ and 60 W for $^{147}Pm$

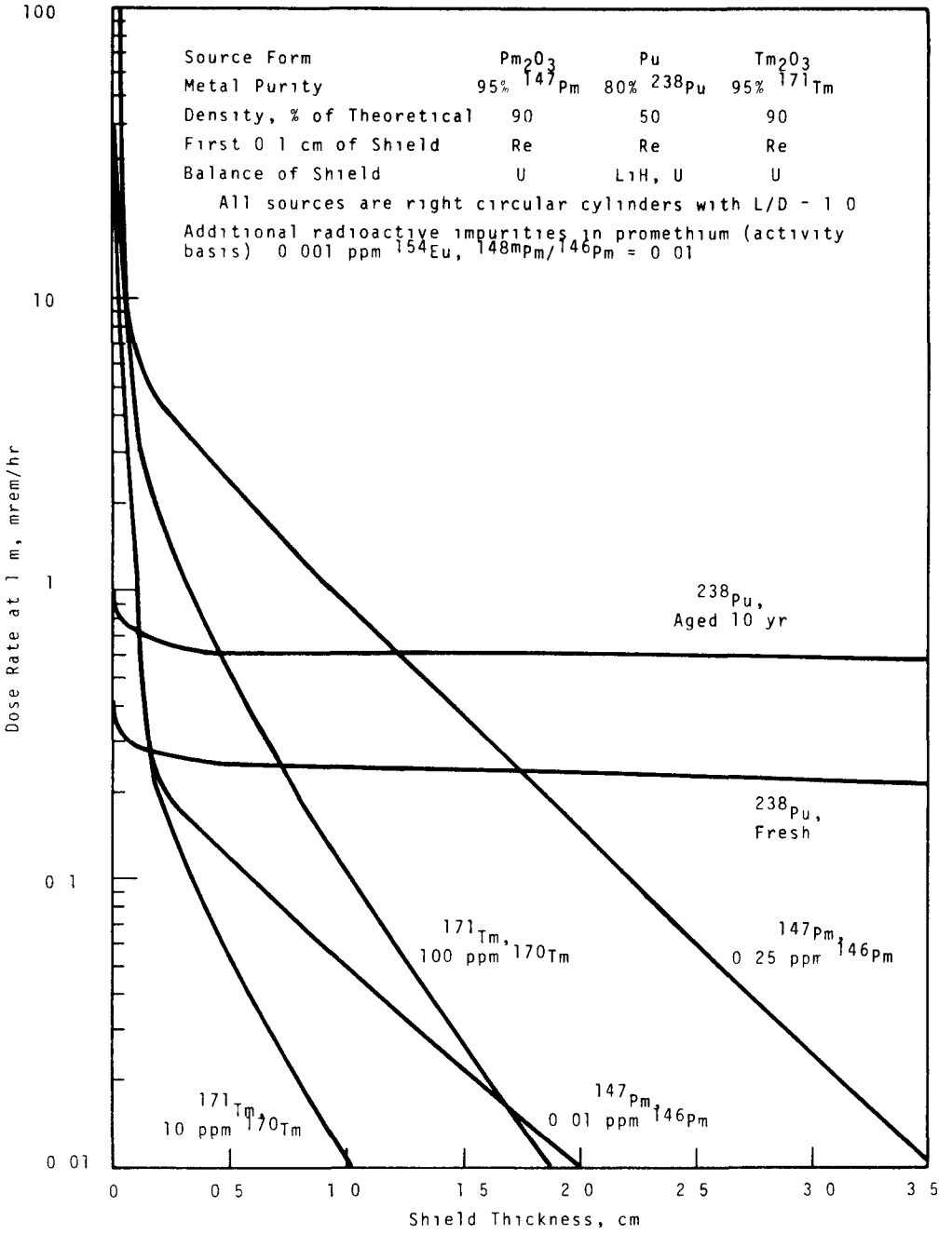


*FIGURE 20. Dose Rate from 30 W Heat Sources as a Function of Shield Thickness*

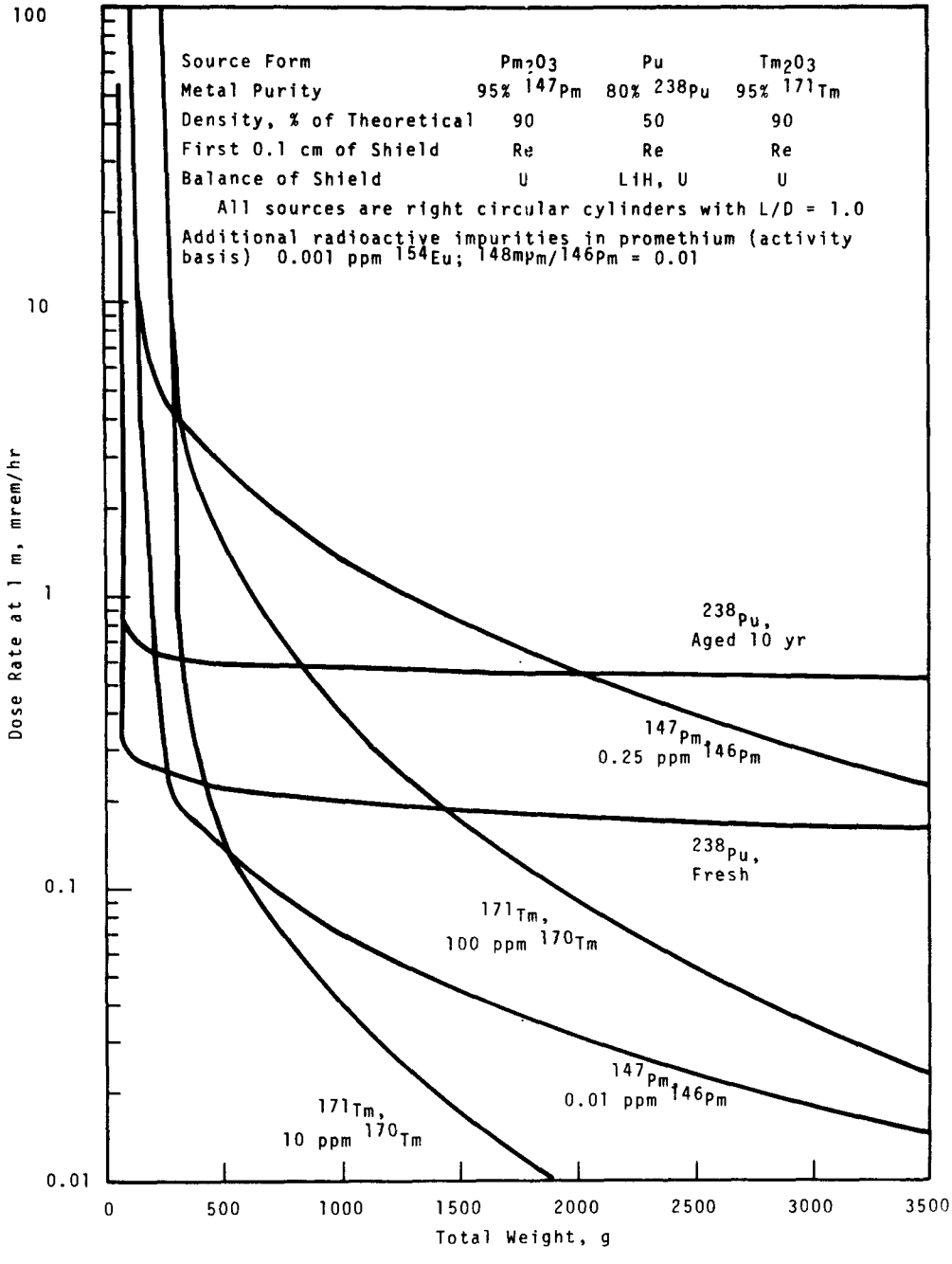


*FIGURE 21. Dose Rate from 30 W Heat Sources as a Function of Total Weight*

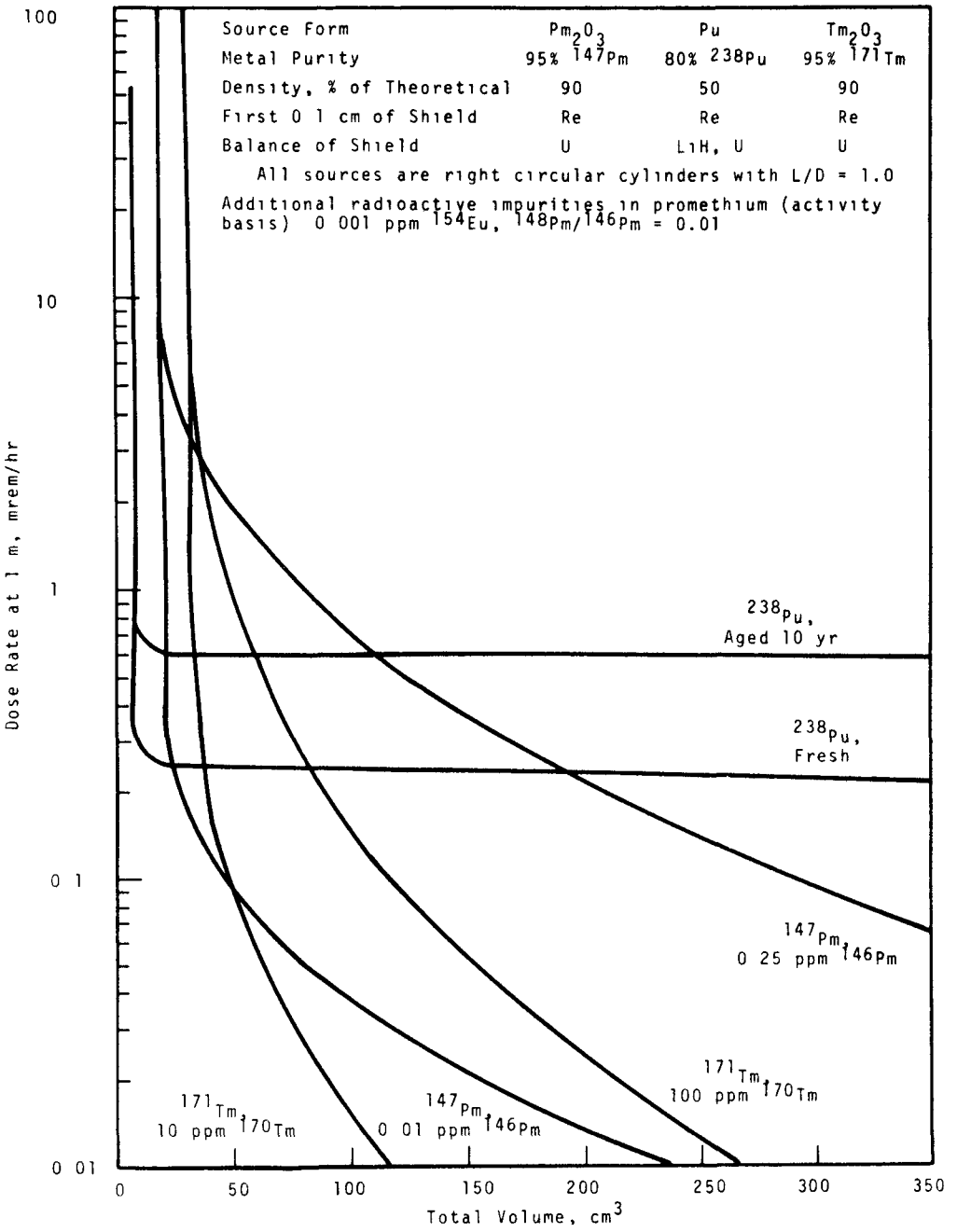


*FIGURE 22. Dose Rate from 30 W Heat Sources as a Function of Total Volume*

*TABLE XIII. Comparison of Candidate Radioisotope Power Sources*

| Age | RBE | Dose Rate, mrem/hr | $^{170}Tm$, ppm | | $^{146}Pm$, ppm | |
|---|---|---|---|---|---|---|
| | | | Lined | Shielded | Lined | Shielded |
| **Lined But Unshielded Plutonium** | | | | | | |
| Fresh | 5 | 17 | 8 | 12 | 0.0018 | 0.010 |
| | 9.05 | 23 | 11 | 16 | 0.0042 | 0.014 |
| | 10 | 25 | 12 | 17 | 0.0051 | 0.015 |
| Aged | 5 | 55 | 1300 | 1900 | 0.028 | 0.047 |
| | 9.05 | 61 | 1400 | 2100 | 0.032 | 0.052 |
| | 10 | 62 | 1500 | 2100 | 0.032 | 0.053 |
| Average | 5 | 38 | 72 | 100 | 0.014 | 0.027 |
| | 9.05 | 43 | 83 | 120 | 0.017 | 0.031 |
| | 10 | 44 | 85 | 120 | 0.017 | 0.032 |
| **Lined and Shielded Plutonium** | | | | | | |
| Fresh | 5 | 11 | 5 | 8 | -- | 0.0065 |
| | 9.05 | 17 | 8 | 12 | 0.0018 | 0.010 |
| | 10 | 18 | 9 | 13 | 0.0022 | 0.011 |
| Aged | 5 | 33 | 770 | 1100 | 0.015 | 0.028 |
| | 9.05 | 38 | 900 | 1300 | 0.018 | 0.032 |
| | 10 | 39 | 930 | 1300 | 0.019 | 0.033 |
| Average | 5 | 23 | 43 | 62 | 0.0068 | 0.017 |
| | 9.05 | 28 | 54 | 77 | 0.0092 | 0.020 |
| | 10 | 29 | 56 | 81 | 0.0097 | 0.021 |
| **Lined and Shielded Plutonium with 56g LiH** | | | | | | |
| Fresh | 5 | 10 | 5 | 7 | -- | 0.0059 |
| | 9.05 | 16 | 7 | 11 | 0.0014 | 0.0095 |
| | 10 | 17 | 8 | 12 | 0.0018 | 0.010 |
| Aged | 5 | 31 | 750 | 1100 | 0.014 | 0.026 |
| | 9.05 | 36 | 860 | 1200 | 0.017 | 0.031 |
| | 10 | 37 | 890 | 1300 | 0.018 | 0.031 |
| Average | 5 | 22 | 41 | 59 | 0.0063 | 0.016 |
| | 9.05 | 26 | 51 | 73 | 0.0082 | 0.019 |
| | 10 | 28 | 53 | 76 | 0.0092 | 0.020 |

and $^{171}Tm$. Mission lifetimes were 10, 2.6, and 2 yr, respectively. Dose rates from plutonium sources are listed, and the quantity tabulated for $^{147}Pm$ and $^{171}Tm$ is the ppm by activity of the principal impurity ($^{146}Pm$ and $^{170}Tm$, respectively), which will result in the same dose rate as the specified $^{238}Pu$ source. The "shielded" values, which include both the rhenium liner and tungsten shield, result in a lower dose rate for the same source composition. Therefore, a higher impurity content can be tolerated for the same dose rate.

The thickness of compatibility liner was chosen to provide shielding from the soft radiations associated with the source, so that a thinner liner would yield a substantially higher dose rate. Thicknesses of 0.1 cm for $^{238}Pu$ and 0.2 cm for the other two heat sources were chosen. A shield of tungsten thick enough to make the total source plus shield weight equal to two pounds was added to obtain the shielded cases.

Dose rates are compared for the sources at the beginning of life, at the end of life, and on the basis of average dose rate during the mission lifetime. Equivalent time bases are used throughout the tabulation so that end-of-life dose rates are compared only with other end-of-life dose rates, etc. The mission average dose rate is obtained by integrating the activity from each radionuclide in the source over the duration of the mission. Dividing by the duration of the mission:

$$\bar{A} = \frac{1}{t} \int_0^t \lambda N dt.$$

For simple decay of a nuclide present in amount $N_o$ at the beginning of life,

$$\bar{A} = \frac{\lambda N_o}{t\lambda} (1-e^{-\lambda t}).$$

Similar but more complicated expressions hold for systems involving two or more sequential decays.

In one case, an additional shield of 56 g of lithium hydride was included around the plutonium source. Because it represents thermal storage provided in several engine design concepts, it is not included in the shield weight. Gamma attenuation in this shield is neglected since attenuation is almost exclusively by Compton scattering, which results in secondary radiations essentially equal to the primary radiation. Neutron attenuation was included, however.

The data are tabulated on the basis of RBE values of 5, 9.05, and 10 for the neutrons associated with the $^{238}Pu$ source. The RBE (relative biological effectiveness) is the ratio of biological damage per unit energy absorption for the radiation being considered to the biological damage per unit energy absorption for penetrating gamma radiation. A newer name for the RBE is the quality factor (QF). The value of 9.05 was derived from a curve of RBE versus energy[1] weighted in accordance with the fission spectrum distribution of neutron energies.

On an average mission dose-rate basis, the comparison shows that $^{238}Pu$ is similar to $^{147}Pm$ with 0.02 ppm $^{146}Pm$ and also to $^{171}Tm$ with 75 ppm $^{170}Tm$. The difference between lined and shielded sources is usually not great. However, for some $^{147}Pm$ sources, the dose rate from $^{147}Pm$ is sufficiently high with only a liner that little or no $^{146}Pm$ is allowable without exceeding the $^{238}Pu$ dose rate. For shielded sources, most of the dose rate is due to $^{146}Pm$. Although source age has some effect on allowable $^{146}Pm$ content, it has much more pronounced effect on allowable $^{170}Tm$ content. By the end of life, the $^{170}Tm$ has decayed by more than an order of magnitude which markedly reduces the dose rate of this nuclide. Probably the most meaningful comparison is that based on average dose rate over the mission life.

A factor-of-two change in RBE does not cause a great change in dose rate or allowable impurity content except for beginning-of-life comparisons. This occurs because much of the dose rate from plutonium is due to gamma radiation, and is thus unaffected by RBE. The relative contributions of gammas and neutrons may be derived from the table since the neutron dose rate at an RBE of 5 is equal to the difference between the total dose rate at RBE values of 5 and 10.

For the preliminary calculations shown in the figures, plutonium was assumed to be β phase metal with a void volume equal to the metal volume. (Later work used ε phase metal because of high proposed temperatures.) The neutron emission rate was taken as 2500 n/sec-g for the source containing 80% $^{238}Pu$. This is only slightly higher than the irreducible minimum of 2150 n/sec-g reported by Los Alamos for the neutron generation rate caused by spontaneous fission.[2] The $^{236}Pu$ content was taken as $1.2 \times 10^{-4}$ at.%, or 45 ppm by activity. The increasing dose rate with time is due to buildup of daughters from $^{236}Pu$, especially $^{208}Tl$.

In the tabulated comparison, plutonium of the following isotopic composition was used:

| Isotope | Wt% |
|---|---|
| $^{238}Pu$ | 81 |
| $^{239}Pu$ | 15 |
| $^{240}Pu$ | 2.9 |
| $^{241}Pu$ | 0.8 |
| $^{242}Pu$ | 0.1 |
| $^{236}Pu$ | $1.2 \times 10^{-4}$ |

Less than 2% of the dose rate came from nuclides other than $^{238}Pu$ and the daughters of $^{236}Pu$. Epsilon phase plutonium metal was assumed, with a density of 16.51. A void volume equal to the plutonium volume was assumed to allow for collection of helium from alpha decay without excessive pressure buildup in

the source. However, a pressure on the order of 4000 psi may be developed, depending on the source temperature. The plutonium was assumed to contain no significant impurities. The neutron emission rate was taken as 2755 n/sec-g on a total plutonium basis, or 3410 n/sec-g on a $^{238}Pu$ basis. This is only about 30% over the irreducible minimum from spontaneous fission.

The thulium sources were assumed to contain $^{170}Tm$ as specified (expressed as ppm by activity). If the $^{170}Tm$ content were expressed on a weight basis, lower ppm values would result since 10 ppm by activity is about 1.9 ppm by weight. The thulium sources were assumed to be oxide with a density of 8.015, which is about 90% of theoretical density. Decay of $^{171}Tm$ to ytterbium was assumed to limit the chemical purity of the thulium to 95%. This allows only about two months between the time of thulium separation from ytterbium and the time of beneficial use.

The promethium sources were assumed to contain $^{146}Pm$ as specified, expressed as ppm by activity. Similar but slightly higher values result if the $^{146}Pm$ content is expressed on a weight basis, since 0.25 ppm by activity is about 0.53 ppm by weight. The promethium sources were assumed to be oxide with a density of 6.157, which is about 90% of theoretical density. Decay of $^{147}Pm$ to samarium was assumed to limit the chemical purity of the promethium to 95%. This allows only about two months between the time of promethium separation from samarium and the time of beneficial use. The amounts of $^{148m}Pm$ and $^{154}Eu$ associated with the promethium were assumed to be dependent on the $^{146}Pm$ content since the amounts can be controlled to any desired level by aging and chemical separation, respectively. The $^{148m}Pm$ activity was assumed to be one percent of the $^{146}Pm$ activity, and the $^{154}Eu$ activity was assumed

to be 0.4% of the $^{146}Pm$ activity. The feasibility of purifying $^{147}Pm$ to this extent has previously been demonstrated by E. J. Wheelwright.[3]

Spectra of gammas, bremsstrahlung, and characteristic X-rays of the nuclides considered are listed in Table XIV. The nuclear data were derived mostly from Lederer et al[4] but with some promethium data being obtained from a recent issue of "Nuclear Data."[5] Bremsstrahlung spectra were calculated with program BREMRAD.[6]

All sources were assumed to be right circular cylinders with length equal to diameter. The dose point was as specified, either 10 cm or 1 m from the center of the source, with no intervening matter between the shielded source and the dose point.

Source weights and activities were based on 30.44 Ci/W for $^{238}Pu$, 2788 Ci/W for $^{147}Pm$, and 5618 Ci/W for $^{171}Tm$.

Geometry and self-shielding effects were included in gamma calculations by approximate methods similar to those developed in HW-69533,[7] but they were modified to be applicable to a cylinder rather than a sphere. This circumvented the need for time consuming double numerical integration. The approximate method has been compared with the more tedious numerical integration method for several typical conditions and found to be within 10%.

Absorption coefficients were derived from the data of Hubbell and Berger.[8] Values for other atomic numbers were obtained by three-point logarithmic interpolation of $\mu$ versus Z. Near the X-ray absorption edges, the interpolation is more difficult. To circumvent this problem, all data were converted to an extrapolated K-shell absorption coefficient by multiplying all values below the edge by the ratio of the

*TABLE XIV. Spectral Distribution of Photons from Selected Nuclides*

| Energy, MeV | Abundance, % |
|---|---|
| From $^{238}Pu$ | |
| 0.017 | 7.83 |
| 0.020 | 2.08 |
| 0.0984 | 1.5 (-4)* |
| 0.1113 | 5.0 (-5) |
| 0.0435 | 0.038 |
| 0.100 | 0.008 |
| 0.153 | 0.001 |
| 0.203 | 4.0 (-6) |
| 0.743 | 9.0 (-6) |
| 0.767 | 3.5 (-5) |
| 0.786 | 6.0 (-6) |
| 0.854 | 5.0 (-6) |
| 1.001 | 8.0 (-7) |
| 1.1 | 1.94 (-7) |
| 1.2 | 1.17 (-7) |
| 1.5 | 3.42 (-7) |
| 1.6 | 3.78 (-7) |
| 2.0 | 1.28 (-7) |
| 2.3 | 3.0 (-7) |
| 2.5 | 6.8 (-8) |
| 2.75 | 6.8 (-8) |
| 3.0 | 8.4 (-8) |
| 5.0 | 4.2 (-9) |
| From $^{236}Pu$ | |
| 0.0476 | 0.0031 |
| 0.109 | 0.012 |
| 0.165 | 6.6 (-4) |
| 0.473 | 9.0 (-6) |
| 0.516 | 1.7 (-4) |
| 0.564 | 9.0 (-5) |
| 0.584 | 6.0 (-6) |
| 0.645 | 2.4 (-4) |
| From $^{232}U$ | |
| 0.017 | 30.0 |
| 0.0575 | 0.21 |
| 0.129 | 0.082 |
| 0.270 | 0.0038 |
| 0.3275 | 0.0034 |
| From $^{241}Am$ | |
| 0.0011 | 0.44 |
| 0.0044 | 5.9 |
| 0.0119 | 0.21 |
| 0.0139 | 7.4 |
| 0.0168 | 2.6 |
| 0.0179 | 33.0 |
| 0.0210 | 11.0 |
| 0.1010 | 0.003 |
| 0.1142 | 0.001 |
| 0.0264 | 2.5 |
| 0.0332 | 0.03 |
| 0.0426 | 6.0 (-4) |
| 0.0434 | 0.07 |
| 0.0556 | 0.026 |
| 0.0595 | 36.0 |
| 0.0675 | 0.001 |
| 0.0990 | 0.024 |
| 0.103 | 0.019 |
| 0.125 | 0.005 |
| 0.146 | 2.0 (-4) |
| 0.165 | 4.0 (-5) |
| 0.169 | 1.0 (-4) |
| 0.208 | 6.0 (-4) |
| 0.222 | 4.0 (-5) |
| 0.268 | 3.0 (-5) |
| 0.293 | 2.0 (-5) |
| 0.312 | 3.0 (-5) |
| 0.323 | 1.6 (-4) |
| 0.332 | 1.8 (-4) |
| 0.335 | 6.0 (-4) |
| 0.338 | 1.0 (-5) |
| 0.369 | 3.0 (-4) |
| 0.371 | 1.3 (-4) |
| 0.377 | 1.6 (-4) |
| 0.384 | 3.0 (-5) |
| 0.420 | 4.0 (-5) |
| 0.427 | 3.0 (-5) |
| 0.570 | 1.0 (-5) |
| 0.597 | 1.0 (-5) |
| 0.619 | 8.0 (-5) |
| 0.641 | 1.0 (-5) |
| 0.653 | 5.0 (-5) |
| 0.662 | 5.0 (-4) |
| 0.680 | 6.0 (-6) |
| 0.695 | 1.0 (-5) |
| 0.689 | 4.0 (-5) |
| 0.710 | 9.0 (-6) |
| 0.722 | 2.8 (-4) |
| 0.737 | 1.2 (-5) |
| 0.757 | 1.3 (-5) |
| 0.767 | 8.0 (-6) |
| 0.771 | 1.0 (-5) |

| Energy, MeV | Abundance, % |
|---|---|
| From $^{228}Th$ | |
| 0.015 | 25.0 |
| 0.0845 | 1.6 |
| 0.132 | 0.19 |
| 0.040 | 0.011 |
| 0.041 | 0.40 |
| 0.073 | 33.0 |
| 0.115 | 0.70 |
| 0.167 | 0.12 |
| 0.177 | 0.24 |
| 0.211 | 0.10 |
| 0.216 | 0.29 |
| 0.233 | 0.10 |
| 0.2386 | 47.0 |
| 0.241 | 4.2 |
| 0.253 | 0.40 |
| 0.277 | 3.6 |
| 0.288 | 0.28 |
| 0.300 | 3.6 |
| 0.328 | 0.10 |
| 0.415 | 0.16 |
| 0.453 | 0.10 |
| 0.486 | 0.18 |
| 0.511 | 9.0 |
| 0.583 | 31.0 |
| 0.727 | 7.1 |
| 0.763 | 0.70 |
| 0.785 | 1.0 |
| 0.860 | 4.3 |
| 0.893 | 0.40 |
| 0.953 | 0.10 |
| 1.076 | 0.70 |
| 1.094 | 0.20 |
| 1.513 | 0.29 |
| 1.62 | 1.6 |
| 1.80 | 0.11 |
| 2.615 | 36.0 |
| From $^{239}Pu$ | |
| 0.0043 | 5.0 |
| 0.0116 | 0.055 |
| 0.0136 | 2.0 |
| 0.0164 | 0.70 |
| 0.0172 | 2.4 |
| 0.0202 | 0.82 |
| 0.0175 | 0.48 |
| 0.0207 | 0.17 |
| 0.0984 | 0.0063 |
| 0.1113 | 0.0021 |
| 0.0130 | 3.0 (-4) |
| 0.0386 | 0.015 |
| 0.0462 | 0.0016 |
| 0.0516 | 0.041 |
| 0.0569 | 0.0016 |
| 0.0686 | 0.0014 |
| 0.0776 | 0.0011 |
| 0.098 | 0.001 |
| 0.103 | 4.0 (-6) |
| 0.116 | 0.0018 |
| 0.1293 | 0.010 |
| 0.1442 | 5.0 (-4) |
| 0.1614 | 9.0 (-5) |
| 0.1714 | 1.8 (-4) |
| 0.1791 | 1.2 (-4) |
| 0.1891 | 1.5 (-4) |
| 0.1956 | 1.9 (-4) |
| 0.2035 | 9.0 (-4) |
| 0.2555 | 1.6 (-4) |
| 0.2640 | 6.0 (-5) |
| 0.2976 | 9.0 (-5) |
| 0.3118 | 5.0 (-5) |
| 0.3211 | 8.0 (-5) |
| 0.3239 | 9.0 (-5) |
| 0.3330 | 8.0 (-4) |
| 0.3363 | 1.8 (-4) |
| 0.3417 | 1.2 (-4) |
| 0.3451 | 8.7 (-4) |
| 0.3674 | 1.6 (-4) |
| 0.3687 | 1.4 (-4) |
| 0.3752 | 2.5 (-3) |
| 0.3804 | 5.0 (-4) |
| 0.3829 | 4.0 (-4) |
| 0.3935 | 1.0 (-3) |
| 0.4137 | 2.5 (-3) |
| 0.4226 | 2.0 (-4) |
| 0.4267 | 3.0 (-5) |
| 0.4516 | 3.4 (-4) |
| 0.639 | 1.7 (-5) |
| 0.645 | 2.0 (-5) |
| 0.652 | 1.3 (-5) |
| 0.659 | 1.6 (-5) |
| 0.718 | 4.0 (-6) |
| 0.758 | 6.0 (-6) |
| 0.771 | 1.8 (-5) |

| Energy, MeV | Abundance, % |
|---|---|
| From $^{240}Pu$ | |
| 0.0010 | 0.18 |
| 0.0043 | 2.4 |
| 0.0116 | 0.12 |
| 0.0136 | 4.3 |
| 0.0164 | 1.5 |
| 0.0172 | 5.2 |
| 0.0174 | 0.20 |
| 0.0202 | 1.8 |
| 0.0207 | 0.07 |
| 0.0984 | 1.46 (-4) |
| 0.1113 | 4.9 (-5) |
| 0.0453 | 0.05 |
| 0.1036 | 0.012 |
| 0.160 | 0.0012 |
| 0.642 | 1.4 (-5) |
| 0.688 | 4.0 (-6) |
| From $^{241}Pu$ | |
| 0.0010 | 2.1 (-6) |
| 0.0043 | 2.7 (-5) |
| 0.0116 | 3.1 (-7) |
| 0.0136 | 1.1 (-5) |
| 0.0164 | 4.0 (-6) |
| 0.0173 | 1.7 (-4) |
| 0.0205 | 5.9 (-5) |
| 0.0984 | 1.14 (-3) |
| 0.1113 | 3.8 (-4) |
| 0.145 | 4.2 (-4) |
| From $^{242}Pu$ | |
| 0.0010 | 0.18 |
| 0.0043 | 2.4 |
| 0.0116 | 0.12 |
| 0.0136 | 4.3 |
| 0.0164 | 1.5 |
| 0.0173 | 5.4 |
| 0.0205 | 1.87 |
| 0.0447 | 0.050 |
| From $^{237}U$ | |
| 0.0010 | 1.3 |
| 0.0043 | 17.0 |
| 0.0116 | 0.32 |
| 0.0136 | 12.0 |
| 0.0164 | 4.1 |
| 0.0174 | 18.0 |
| 0.0205 | 6.4 |
| 0.0984 | 42.0 |
| 0.1113 | 14.0 |
| 0.0138 | 0.001 |
| 0.0264 | 2.4 |
| 0.0332 | 0.019 |
| 0.0434 | 0.074 |
| 0.0510 | 0.20 |
| 0.0595 | 36.0 |
| 0.0648 | 1.3 |
| 0.1030 | 0.60 |
| 0.1646 | 2.0 |
| 0.2080 | 23.0 |
| 0.2218 | 0.022 |
| 0.2344 | 0.021 |
| 0.2675 | 0.76 |
| 0.3324 | 1.3 |
| 0.3354 | 0.10 |
| 0.3377 | 0.027 |
| 0.3686 | 0.050 |
| 0.3709 | 0.12 |
| From $^{171}Tm$ | |
| 0.0016 | 0.0065 |
| 0.0063 | 0.0033 |
| 0.0072 | 0.126 |
| 0.0081 | 0.195 |
| 0.0085 | 0.050 |
| 0.0096 | 0.078 |
| 0.0507 | 0.78 |
| 0.0575 | 0.26 |
| 0.0667 | 0.18 |
| 0.01 | 0.464 |
| 0.02 | 0.133 |
| 0.03 | 0.0502 |
| 0.04 | 0.020 |
| 0.05 | 7.53 (-3) |
| 0.06 | 2.46 (-3) |
| 0.07 | 6.12 (-4) |
| 0.08 | 8.51 (-5) |
| 0.09 | 2.06 (-6) |

| Energy, MeV | Abundance, % |
|---|---|
| From $^{170}Tm$ | |
| 0.0016 | 0.12 |
| 0.0063 | 0.047 |
| 0.0072 | 1.8 |
| 0.0081 | 2.3 |
| 0.0085 | 0.71 |
| 0.0096 | 0.93 |
| 0.0507 | 3.8 |
| 0.0575 | 1.3 |
| 0.0840 | 3.7 |
| 0.05 | 4.19 |
| 0.10 | 1.34 |
| 0.15 | 0.613 |
| 0.20 | 0.322 |
| 0.25 | 0.182 |
| 0.30 | 0.107 |
| 0.35 | 0.0644 |
| 0.40 | 0.0390 |
| 0.45 | 0.0235 |
| 0.50 | 0.0140 |
| 0.55 | 8.11 (-3) |
| 0.60 | 4.52 (-3) |
| 0.65 | 2.38 (-3) |
| 0.70 | 1.15 (-3) |
| 0.75 | 4.96 (-4) |
| 0.80 | 1.78 (-4) |
| 0.85 | 4.77 (-5) |
| 0.90 | 6.92 (-6) |
| 0.95 | 1.25 (-7) |
| From $^{147}Pm$ | |
| 0.01 | 1.18 |
| 0.02 | 0.426 |
| 0.03 | 0.216 |
| 0.04 | 0.125 |
| 0.05 | 0.0778 |
| 0.06 | 0.0502 |
| 0.07 | 0.0330 |
| 0.08 | 0.0219 |
| 0.09 | 0.0146 |
| 0.10 | 9.63 (-3) |
| 0.11 | 6.28 (-3) |
| 0.12 | 7.01 (-3) |
| 0.13 | 2.50 (-3) |
| 0.14 | 1.5 (-3) |
| 0.15 | 8.6 (-4) |
| 0.16 | 4.6 (-4) |
| 0.17 | 2.2 (-4) |
| 0.18 | 9.7 (-5) |
| 0.19 | 3.4 (-5) |
| 0.20 | 8.8 (-6) |
| 0.21 | 1.2 (-6) |
| From $^{146}Pm$ | |
| 0.146 | 0.2 |
| 0.4536 | 65.0 |
| 0.590 | 0.60 |
| 0.7362 | 21.0 |
| 0.6333 | 2.0 |
| 0.7474 | 34.0 |
| 1.190 | 0.05 |
| 1.383 | 0.006 |
| From $^{148}Pm$ | |
| 0.40 | 15.0 |
| 0.56 | 101.0 |
| 0.63 | 100.0 |
| 0.73 | 33.0 |
| 0.93 | 19.0 |
| 1.02 | 20.0 |
| 1.46 | 2.0 |
| From $^{154}Eu$ | |
| 0.593 | 6.0 |
| 0.693 | 4.0 |
| 0.724 | 20.0 |
| 0.758 | 4.0 |
| 0.874 | 15.0 |
| 1.00 | 30.0 |
| 1.276 | 38.0 |

* *Numbers in parentheses represent powers of 10 by which preceding number is to be multiplied.*

two absorption coefficients at the discontinuity. This process was repeated as necessary for the L, M, and N edges. A particular absorption coefficient was then obtained by three-point logarithmic interpolation of E versus $\mu$. This type of interpolation was found to be substantially more accurate than linear or semilogarithmic interpolation.

Buildup factors were generally those of Goldstein and Wilkins[9] but for energies below 0.5 MeV, unpublished values developed by Battelle-Northwest were used. Particular values were obtained by three-point logarithmic interpolation of E versus $\mu t$ for three values of $\mu t$, followed by three-point logarithmic interpolation of B versus $\mu t$. Some additional adjustments were necessary for very low or very high values of $\mu t$, but these caused only a slight change in the final result.

In addition to the customary photon buildup factor, the buildup of X-rays from photoelectric capture was also included. The X-rays were divided into eleven groups: two K X-rays, seven L X-rays, and one each M and N X-ray. The photoelectric absorption for each shell was calculated, and the X-rays arising from that shell were distributed in accordance with the expected decay probability. Internal conversion of the X-rays was included by decreasing the amount of each X-ray in accordance with the fluorescence yield, with the converted X-rays giving rise to X-rays in the lower shells. X-ray buildup in the source was calculated for each nuclide, and the X-rays were treated the same as gamma rays arising within the source--including self-absorption in the source. X-rays arising in the shields were calculated for each nuclide and each shield region.

Another buildup factor for conversion electrons from photoelectric capture was <u>not</u> included in these calculations since a few $mg/cm^2$ of a low atomic number absorber will

reduce these to a negligible level. At the surface of a bare heavy metal shield, the dose rate may be increased by a factor of two or more because of photoelectrons. At a distance of 1 m from the source, absorption of the photoelectrons by air would be appreciable.

In a matrix of constant size, density and composition, the dose rate at a given distance from the source is directly proportional to the amount of radionuclide in the source. Thus, the effect of variation in the amount of $^{146}Pm$ in a given $^{147}Pm$ source (for example) can be readily evaluated if the dose rate contribution from $^{146}Pm$ and $^{149}Pm$ are each known for any $^{146}Pm$ content. Likewise, if an allowable dose rate is given, the permissible $^{146}Pm$ content can be evaluated by subtracting the $^{147}Pm$ contribution from the allowable dose rate and determining by simple ratio the amount of $^{146}Pm$ that will contribute the balance of the allowable dose rate. This property of radiations was utilized in obtaining average dose rates, end-of-life dose rates, and dose rates from various initial amounts of $^{146}Pm$ and $^{170}Tm$.

Self-shielding changes with source size, density, or composition; geometry changes with source size. Since these effects are not linear, any change in these variables, or the amount or composition of external shielding, requires a complete recalculation of dose rates for each radionuclide in the source.

The neutron calculations were much less refined than the gamma calculations. The neutron flux at the dose point was calculated from the neutron emission rate of the source and the geometry effect. Neither shielding nor buildup of neutrons in the source was considered, and shielding external to the source was also ignored except for lithium hydride when it was present. A flux-to-dose rate conversion factor

was derived from unpublished data by Bach and Caswell[10] assuming a fission spectrum distribution of neutron energies. This is based on first collision energy transfer, neglecting buildup of scattered neutrons. Since no alteration of the neutron spectrum or intensity was assumed in the shields, the omission of buildup is probably conservative. The neutron dose rate obtained in this way was multiplied by the RBE value and added to the gamma dose rate to obtain the total dose rate in rems per hour.

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# Appendix B

## Dose Rate Data for Sources Weighing 1 lb*

Source: 30-W, cylindrical, $^{238}Pu$, L = D, void volume = 50%, 0.10-cm rhenium liner, tungsten-shielded

| Source Age | $^{238}Pu$ Dose Rate, mr/hr, at 10 cm for RBE = 5 | RBE = 9.05 | RBE = 10 |
|---|---|---|---|
| Fresh | 13 | 19 | 21 |
| Average | 28 | 34 | 35 |
| Aged | 39 | 45 | 46 |

Source: 60-W, cylindrical, $^{171}Tm$, L = D, 0.054-cm rhenium liner and shield

| Source Age | Amount, ppm of $^{170}Tm$ to Give Above Dose Rates RBE = 5 | RBE = 9.05 | RBE = 10 |
|---|---|---|---|
| Fresh | 0.3 | 0.5 | 0.5 |
| Average | 2.7 | 3.3 | 3.4 |
| Aged | 36 | 43 | 45 |

*Private communication from J. C. Sheppard, Battelle Northwest, to W. E. Mott, DID-USAEC, Aug. 13, 1968.

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