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# **Analysis of Production Mechanisms for Several Radioisotopes and Isomers, and Analysis of Radiation from a Bulk Emitter**

**by Jarrod C. Marsh, James J. Carroll, and Christopher J. Chiara**

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*DEVCOM Army Research Laboratory*

## REPORT DOCUMENTATION PAGE

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| <b>14. ABSTRACT</b><br>The ARL Nuclide Science Team analyzed potentially useful isotopes, including those with metastable isomeric states, for a heat source and their production methods, and radiation outputs are discussed. Production pathways for various candidates were analyzed using cross sections extracted from the literature. <sup>242m</sup> Am is the easiest to produce using current neutron-capture techniques assuming steps are taken to minimize burnup in the reactor before sample extraction. <sup>170</sup> Tm and <sup>242m</sup> Am have similar production rates. Dose rates were estimated using Geant4 simulations of radiation emitted within a simple volume of radioactive material to examine self-shielding estimates related to the external dose rate. The simulations confirm nonlinear scaling between the physical size of the radioactive source and the external dose. This demonstrates that the thermal output can be improved by increasing the size of the source without significantly increasing the external dose. |                   |                                    |                                         |                                                                                                                                                                                                                                                 |                                               |                   |                 |               |                   |
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## 1. Introduction

The Nuclide Science Team, Energy Conversion Branch, U.S. Army Combat Capabilities Development Command Army Research Laboratory, analyzed production reactions for several radioisotopes and isomers of interest for potential energy and power applications. This work was supported by an Operational Energy Capability Improvement Fund (OECIF)-funded Air Force Research Laboratory (AFRL) project. The two standard radioisotopes of primary interest were  $^{170}\text{Tm}$  and  $^{244}\text{Cm}$ , which are envisioned to be present in the forms of  $\text{Tm}_2\text{O}_3$  and  $\text{Cm}_2\text{O}_3$ , respectively. The relevant decay properties (all quoted nuclear data properties, unless otherwise indicated, are taken from the Evaluated Nuclear Structure Data File [ENSDF]<sup>1</sup>) of these radioisotopes are provided in Table 1.

**Table 1. Decay properties of  $^{170}\text{Tm}$  and  $^{244}\text{Cm}$ .**

| Radioisotope      | Half-life<br>( $T_{1/2}$ ) | Decay mode(s)<br>(intensity, %) | Emitted radiation<br>energy, keV<br>(intensity, %) | Daughter<br>( $T_{1/2}$ )      |
|-------------------|----------------------------|---------------------------------|----------------------------------------------------|--------------------------------|
| $^{170}\text{Tm}$ | 128.6 days                 | $\beta^-$ (99.869)              | $E_\beta = 968.1$ (81.9)                           | $^{170}\text{Yb}$ (stable)     |
|                   |                            |                                 | $E_\beta = 883.8$ (18.1)                           |                                |
|                   |                            |                                 | $E_\gamma = 84.25$ (2.48)                          |                                |
|                   |                            |                                 | $E_{ce} < 84.25$ (15.61)                           |                                |
|                   |                            | $\epsilon$ (0.131)              | $E_\gamma = 78.7$ (0.016)                          | $^{170}\text{Er}$ (stable)     |
|                   |                            |                                 | $E_{ce} < 78.7$ (0.115)                            |                                |
| $^{244}\text{Cm}$ | 18.1 years                 | $\alpha$ (100)                  | $E_\alpha = 5804.8$ (76.9)                         | $^{240}\text{Pu}$ (6561 years) |
|                   |                            |                                 | $E_\alpha = 5762.6$ (23.1)                         |                                |
|                   |                            |                                 | $E_\gamma = 42.82$ (0.025)                         |                                |
|                   |                            |                                 | $E_{ce} < 42.82$ (23.075)                          |                                |
|                   |                            |                                 | $E_\alpha \leq 5644$ (<0.02)                       |                                |
|                   |                            |                                 | $E_\gamma, E_{ce} = \text{various}$ (<0.02)        |                                |
|                   |                            | SF (0.00014)                    | Various (<0.00014)                                 | Various                        |

Note: Emission of gamma rays (denoted by  $\gamma$ ) and conversion electrons (denoted by ce) are prompt after the parent radioactive decays; X-ray emission is not included but will be present. Radioactive decay modes are as follows:  $\beta^-$  = electron decay;  $\epsilon$  = atomic electron capture decay by the nucleus;  $\alpha$  = alpha decay; and SF = spontaneous fission. For conversion electrons, the energies will depend on which atomic shell was the origin of the ejected electron; the total intensity of all conversion electrons is given.

The Nuclide Science Team analyses included the following:

- 1) Evaluation of reactions and reaction pathways for production of several different radioisotopes, compared with production of  $^{170}\text{Tm}$ :
  - a. (U)  $^{148}\text{Gd}$
  - b. (U)  $^{192\text{m}}\text{Ir}$
  - c. (U)  $^{242\text{m}}\text{Am}$
- 2) Evaluation of radiation emission from a bulk material containing  $^{170}\text{Tm}$ .

## 2. Production Reactions and Reaction Pathways

### 2.1 $^{148}\text{Gd}$

Each of the radioisotopes listed above for supplemental study was selected due to specific characteristics. In the case of  $^{148}\text{Gd}$ , the key feature is that it decays with 100% probability by emission of an  $\alpha$  particle (see Table 2). Despite their high kinetic energies,  $\alpha$  particles have extremely little penetrating power through matter and are easily stopped in thin layers. In addition, there is only one  $\alpha$ -decay branch for  $^{148}\text{Gd}$  decay, leading to the ground state of the daughter nuclide  $^{144}\text{Sm}$ , so that the decay is not accompanied by subsequent  $\gamma$  emission. Finally,  $^{144}\text{Sm}$  is stable so that there is no further radiation to consider. Taken together, these suggest a reduced need for radiation shielding that would serve as parasitic weight for applications. The half-life of  $^{148}\text{Gd}$  is long at 71.1 years, and this leads to a lower specific power than either  $^{170}\text{Tm}$  or  $^{244}\text{Cm}$  due to the fundamental inverse relationship between emitted power and half-life.

**Table 2. Decay properties of  $^{148}\text{Gd}$ .**

| Radioisotope      | Half-life<br>( $T_{1/2}$ ) | Decay mode(s)<br>(intensity, %) | Emitted radiation energy,<br>keV (intensity, %) | Daughter<br>( $T_{1/2}$ )  |
|-------------------|----------------------------|---------------------------------|-------------------------------------------------|----------------------------|
| $^{148}\text{Gd}$ | 71.1 years                 | $\alpha$ (100)                  | $E_\alpha = 3182.69$ (100)                      | $^{144}\text{Sm}$ (stable) |

Note: Decay modes are as follows:  $\alpha$  = alpha decay.

The most straightforward method for producing any radioisotope, if physically allowed, is by neutron capture since that can be performed in a nuclear reactor with high neutron fluences. However, there is no simple feedstock for such production of  $^{148}\text{Gd}$  because  $^{147}\text{Gd}$  is unstable with a half-life of only  $T_{1/2} = 38.06$  h. Radioisotope production during an irradiation period ( $T_{\text{irr}}$ ) is often governed by Equation 1, which describes how many of the produced nuclei survive until the end of the period (since those nuclei produced earlier in the period might decay before the end of the period):

$$N_{\text{irr}} = \frac{R_{\text{prod}} T_{1/2}}{\ln(2)} \left[ 1 - e^{-T_{\text{irr}} \ln(2)/T_{1/2}} \right], \quad (1)$$

where  $R_{\text{prod}}$  is a constant rate at which desired radioisotopes are produced from some feedstock,  $T_{1/2}$  is the half-life of the produced nuclei, and  $N_{\text{irr}}$  is the number of those nuclei surviving until the end of the production period. Equation 1 assumes that there is no significant reduction in the number of target nuclei during irradiation.

A simple way to account for decay of the feedstock seems to be to replace the constant production rate by an exponentially decaying function after the feedstock

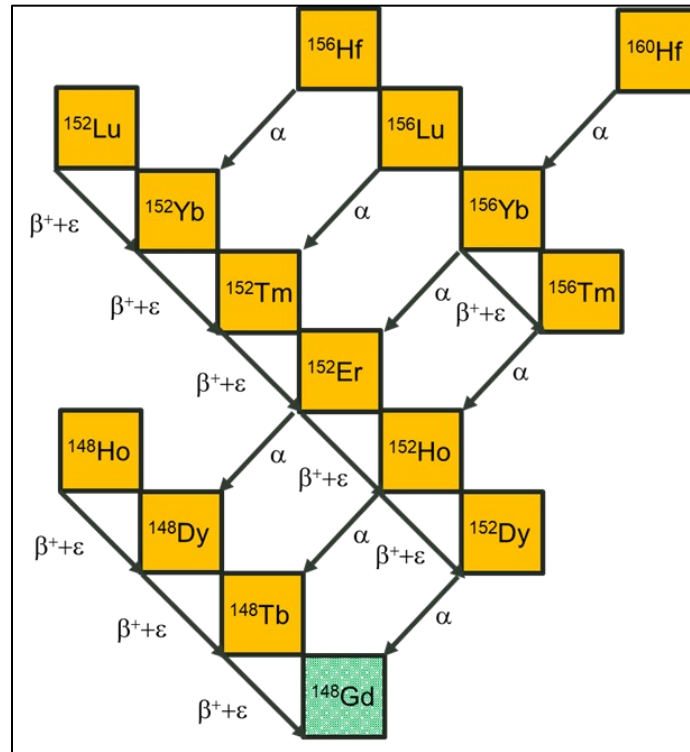
half-life. However, this does not account for depletion of the feedstock by decay and transmutation, and it also neglects the possibility that the produced nuclei are themselves transmuted (burned up) by concomitant nuclear reactions. The correct approach involves Bateman equations with branching and is detailed in Friedlander et al.<sup>2</sup> A typical flux of reactor thermal neutrons of  $10^{14} \text{ cm}^{-2} \text{ s}^{-1}$  is assumed for the calculation, and the cross section for thermal neutron capture by  $^{147}\text{Gd}$ , which is never measured, is taken to be the same as that for capture by  $^{148}\text{Gd}$ , 14 kb.<sup>3</sup> The maximum production of  $^{148}\text{Gd}$  from  $^{147}\text{Gd}$  would occur for  $T_{\text{irr}} \sim 3.5$  days, after which further irradiation of the same  $^{147}\text{Gd}$  target would actually result in less  $^{148}\text{Gd}$ . Mass production of  $^{148}\text{Gd}$  would require replacement of the feedstock with a fresh target to be irradiated and repetition of this cycle. Of course, this is just an exercise to illustrate the general process required to acquire significant amounts of a radioisotope from a feedstock: in this case, there is no stable feedstock of  $^{146}\text{Gd}$  (or for that matter, any stable isotope of Gd with  $A < 150$ ;  $^{148}\text{Gd}$  is proton-rich and neutron-deficient) for neutron-capture production of  $^{147}\text{Gd}$ .

The initial task was to evaluate proton spallation as a potential production process for  $^{148}\text{Gd}$ . This is a process in which high-energy protons (often in the range of hundreds of MeV to GeV kinetic energy) are incident on target nuclei, resulting in the prompt emission of a few to many light particles, for example, neutrons and protons, and the production of a product nucleus that can be of much smaller atomic mass number,  $A$ , than the target nucleus. Encyclopedic discussions of proton reactions at high energies may be found in Filges and Goldenbaum,<sup>4</sup> Hüfner,<sup>5</sup> and David<sup>6</sup>; four reaction regimes are delineated in Hüfner<sup>5</sup>:

- 1) Spallation (“evaporation”): There is one product nucleus per reaction with  $A > 50$ .
- 2) Fission: There are two product nuclei per reaction, which have  $A \sim A_{\text{Target}}/2$  and  $A_{\text{Target}}$  as the atomic mass number of the target nuclei.
- 3) Multifragmentation: There are multiple (more than two) product nuclei per reaction with  $A < 50$  (thus excludes  $^{148}\text{Gd}$ ).
- 4) There are product nuclei like those produced by fission, but the double peaking in the  $A$  distribution are washed out.

The production of radioisotopes due to proton spallation/fission in targets used for spallation-neutron facilities like the Los Alamos Neutron Science Center (LANSCE) is important to understand (and minimize from those facilities’ perspectives). In Kelley,<sup>7</sup> LANSCE,<sup>8</sup> and Kelley et al.<sup>9</sup>  $^{148}\text{Gd}$  is specifically examined as the largest contributor to residual activation of such targets. In those works, measured yields of  $^{148}\text{Gd}$  were obtained from proton irradiations at 600 and 800 MeV on targets of tungsten, tantalum, and gold. Theoretical predictions (see Figure 4 of Kelley<sup>7</sup>) indicate that only approximately 5%–45% (model dependent)

of the  $^{148}\text{Gd}$  present after 800-MeV proton irradiation of a tungsten target is due to direct production, such that much (or most) of this radioisotope production is by accumulation after decays of larger-A nuclides resulting from the spallation reactions. A representation of such an accumulation mass chain is shown in Figure 1.



**Figure 1.** Representation of a chain of radioactive decays leading to  $^{148}\text{Gd}$  (adapted from Figure A.1 of LANSCE<sup>8</sup>).

The cross sections for direct production of each nuclide are difficult to determine experimentally for various reasons; for example, measurements are difficult to obtain for  $^{160}\text{Hf}$  due to its short half-life (13.6 s) and the overall high activities of samples that require delayed handling. For this reason, although the cumulative production of  $^{148}\text{Gd}$  can be directly measured from its  $\alpha$  decays after considerable cooling of a sample, direct production of feeding radioisotopes is often only available from the theory. Kelley et al.<sup>9</sup> concluded that the so-called “Bertini–Dresner” model used by Monte Carlo N-Particle (MCNP) version 2.5.E gave the best agreement between theoretical predictions and measurements. That model predicted that the largest cross section for direct production of radioisotopes (shown in Figure 1) was to  $^{152}\text{Dy}$  (13.9 mb), followed by that to  $^{148}\text{Gd}$  (9.26 mb), whereas the cumulative production to  $^{148}\text{Gd}$  was 20.9 mb from 800-MeV protons on a natural tungsten target. Measurements agreed with a cumulative cross section of 19.4(18) mb for  $^{148}\text{Gd}$ . Figure 2 shows the direct production cross section for

nuclides of various atomic mass numbers predicted by the theory for 800-MeV protons on tungsten.

In this report, the focus is not on the incidental (and unwanted) production of  $^{148}\text{Gd}$  by proton spallation at a facility like LANSCE in normal operations (800-MeV protons on tungsten), but instead it focuses on what conclusions might be drawn regarding the possibility of intentional production of  $^{148}\text{Gd}$ . Hüfner<sup>5</sup> notes that, in the fission region, neutron-rich nuclides are better produced, whereas in the spallation (evaporation) region, neutron-deficient nuclides (like  $^{148}\text{Gd}$ ) are favored, so this region is emphasized in the following discussion.

In Figure 2, a red box draws attention to the evaporation region below the mass number of the target,  $A_{\text{Target}}$ , where the cross section for direct production of nuclides (in particular, neutron-deficient nuclides) is generally maximized. This region has a width of approximately  $\Delta A \approx 20$ , but note that production of nuclides is strongly suppressed for nuclides having the first approximately five mass numbers below  $A_{\text{Target}}$ . For 800-MeV protons, production of  $^{148}\text{Gd}$  would likely be optimized by using a target with  $A_{\text{Target}} \sim 153\text{--}168$ . Similarly, Figure 3b of Kelley<sup>7</sup> shows that the spallation yields are highest for those elements that are no more than several protons less than that of the target; thus, a target with  $Z \sim 65\text{--}70$  would enhance production of  $Z = 64$   $^{148}\text{Gd}$ . Figure 3 shows the portion of the nuclear chart within these  $A$  and  $Z$  ranges, highlighting the stable nuclides (black boxes) that could potentially serve as spallation targets. Natural targets of dysprosium ( $Z = 66$ ) or mono-isotopic terbium (65) or holmium (67) could be used, because all stable isotopes of these elements fall within the given region. Erbium (68) would also be suitable, because only one of its stable isotopes falls just outside of this region. A target of  $^{168}\text{Yb}$  ( $Z = 70$ ) would also be possible, but this is a low-abundance isotope ( $\sim 0.1\%$ ) and would require significant enrichment. Finally, regarding selection of possible target materials, the melting points of approximately 1400 °C (terbium and dysprosium), 1500 °C (holmium and erbium), and 800 °C (ytterbium) compared with 3400 °C for tungsten, since targets for proton spallation are subjected to significant heating.

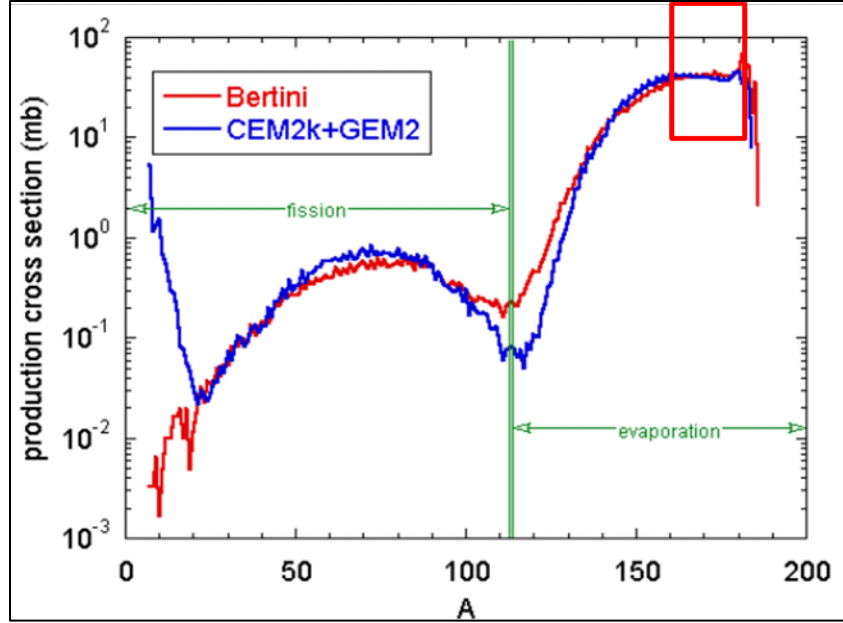


Figure 2. Direct production cross section for nuclides by atomic mass number  $A$  for 800-MeV protons on a natural tungsten target (adapted with permission from Figure 3a of Kelley<sup>7</sup>) from two theoretical models using MCNPX v2.4.0. Our figure shows the regions of mass number where nuclides are preferentially produced by spallation (“evaporation”) and by proton-induced fission of the target nuclei. The red box draws attention to the region within approximately  $\Delta A \approx 20$  below the target’s mass number  $A \sim 184$ .

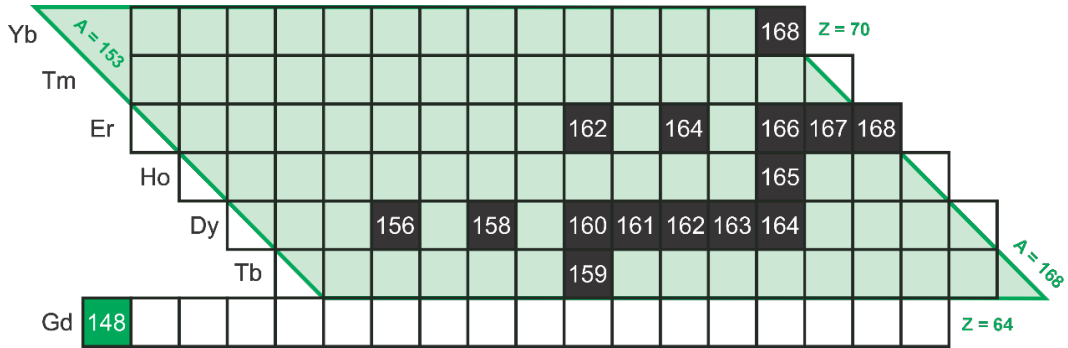


Figure 3. A portion of the nuclear chart (green shading) within the  $Z$  and  $A$  ranges, described in the text, for which a spallation target would be expected to maximize production of  $^{148}\text{Gd}$  (green square). Black squares mark the stable isotopes. The element symbols are indicated along the left side of the graphic, and the mass  $A$  of each relevant isotope is indicated in the square.

In addition to optimizing the target, it should also be possible to optimize  $^{148}\text{Gd}$  production by modifying the proton energy. The work of Kelley et al.<sup>9</sup> compares cumulative production of  $^{148}\text{Gd}$  for both 600- and 800-MeV protons on tungsten, with measurements of 8.31(92) and 19.4(18) mb, respectively. This suggests significant improvement with increasing proton energy, but this may simply reflect that tungsten is relatively far away in atomic number from gadolinium. A full analysis of optimization of proton beam energy and target composition for  $^{148}\text{Gd}$  production could be performed using MCNPX or Geant4 (which also includes the Bertini–Dresner model) but was not attempted.

Hüfner<sup>5</sup> investigates the production of various nuclides due to heavy ions compared with their production due to high-energy proton irradiations. Some enhancement could be obtained for heavy ions (see Figure 3.2 of Hüfner<sup>5</sup> for production of cesium isotopes via different projectiles and energies).

An estimate of the production rate for  $^{148}\text{Gd}$  due to 800-MeV protons on a tungsten target at LANSCE can be obtained using the cumulative cross section of 19.4 mb according to Equation 2 (standard textbook expression, adapted to units typical in nuclear physics experiments):

$$R = 3,760\sigma I_{\text{Beam}}\Theta/A_{\text{Target}} , \quad (2)$$

where  $R$  is the reaction rate in  $\text{s}^{-1}$ ,  $\sigma$  is the reaction cross section in mb,  $I_{\text{Beam}}$  is the “current” of incident particles in particle nanoamperes,\*  $\Theta$  is the thickness of the target in  $\text{mg}/\text{cm}^2$ , and  $A_{\text{Target}}$  is the mass number of the target nuclei. With values of  $\sigma = 19.4$  mb,  $I_{\text{Beam}} = 135 \times 10^3$  (Lisowski and Schoenberg<sup>10</sup>),  $\Theta = 5 \times 10^5$   $\text{mg}/\text{cm}^2$  (~25-cm thickness<sup>11</sup>), and  $A_{\text{Target}} = 184$ ,  $R = 2.7 \times 10^{13}$   $^{148}\text{Gd}$  nuclei produced per second. This corresponds to 1 kg produced of pure  $^{148}\text{Gd}$  in 4141 years, little of which will remain due to its half-life of 71.1 years. As a sanity check, Kelley<sup>7</sup> lists 1.3 Ci as the maximum activity of  $^{148}\text{Gd}$  allowed at LANSCE (for target 1L). Since activity is given by the number of radioactive nuclei divided by the lifetime,  $\tau = T_{1/2}/\ln(2)$ , this corresponds to 40 mg of  $^{148}\text{Gd}$ , which, based on the measured cumulative cross section, would be produced in approximately 2 months of continuous, around-the-clock operation. The similar facility, the Spallation Neutron Source at Oak Ridge National Laboratory (ORNL), changes their proton-spallation targets every 6 months; therefore, considering the possibility of reduced beam currents or less than 24-h continuous operations, the above estimate is reasonable. Although the production of  $^{148}\text{Gd}$  could be optimized as described above, this is not likely to change the production rate by many powers of 10.

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\* One particle nanoampere =  $6.24 \times 10^9$  particles/s.

Production or accumulation of  $^{148}\text{Gd}$  could also occur via several other processes, as depicted in Figure 4. Unfortunately, parents for the decay paths are themselves relatively short-lived, that is,  $^{148}\text{Tb}$  ( $T_{1/2} = 60$  min) for  $\beta^+ + \epsilon$  decay, or  $^{152}\text{Dy}$  ( $T_{1/2} = 2.38$  h) for  $\alpha$  decay, and would present their own production challenges. Figure 4a also shows a possible  $^{154}\text{Gd}(n, 7n)^{148}\text{Gd}$  reaction, but this would require an incident neutron energy of approximately 48 MeV ( $\sim 6 \times 8$  MeV, the neutron binding energy for the gadolinium isotopes) and likely lead to a low cross section. Similarly, as depicted in Figure 4b, deuteron reactions on stable europium isotopes might be feasible. There is also the possibility of production of  $^{148}\text{Gd}$  via the  $^{144}\text{Sm}(\alpha, \gamma)$  reaction, essentially the inverse of  $^{148}\text{Gd}$  decay, which was investigated experimentally over a limited range of incident  $\alpha$  energies in Somorjai et al.<sup>12</sup> Both deuteron and  $\alpha$  reactions were analyzed by an Air Force Institute of Technology graduate student.\*

A final possible production process that has been recently suggested is the  $^{141}\text{Pr}(^{10}\text{B}, 3n)$  reaction. No data is currently available for this reaction, and a measurement of its cross section is planned, perhaps at Australian National University.

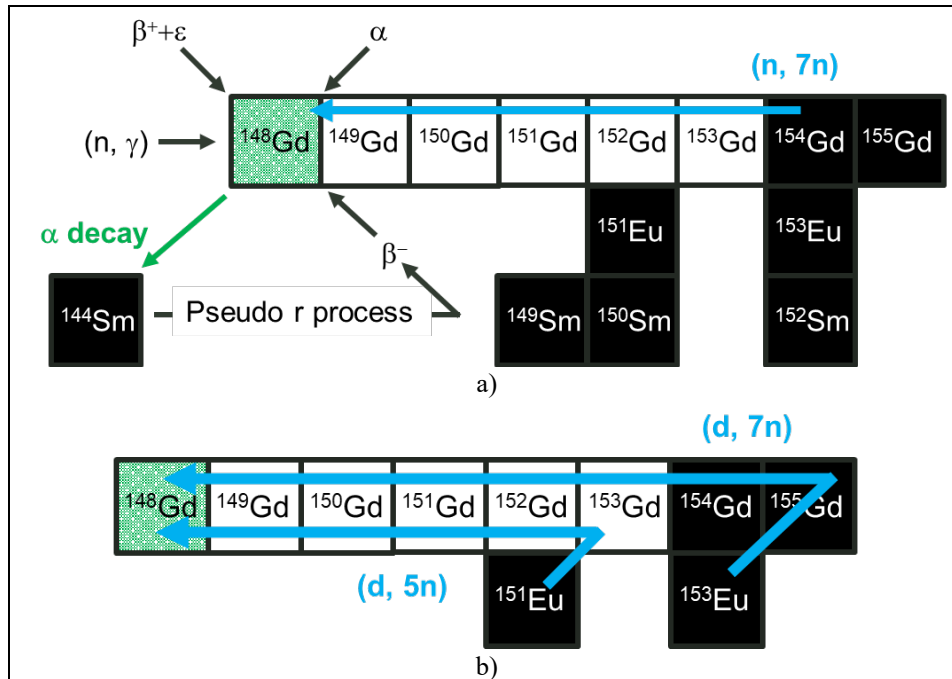


Figure 4. Section of the Chart of the Nuclides<sup>13</sup> showing several hypothetical decay or reaction pathways to  $^{148}\text{Gd}$ . Nuclides in black are stable. Although the “pseudo r process” pathway might exist for some nuclides, it cannot produce  $^{148}\text{Gd}$  because there is no known  $\beta^-$  decay from  $^{148}\text{Eu}$ .

\* Analysis performed by Second Lieutenant Dustin Rodgers (U.S. Air Force) as part of his master’s degree work but not reported in his thesis.

## 2.2 $^{192\text{m}}\text{Ir}$ and $^{242\text{m}}\text{Am}$

The nuclides  $^{192}\text{Ir}$  and  $^{242}\text{Am}$  have nuclear isomers (metastable excited states) that are distinguished by their long half-lives, suitable for energy storage, and the much shorter half-lives for their ground states, providing the potential for much greater power output. Table 3 provides relevant known decay properties. A key to their use would be the ability to transform such nuclei from their isomeric states to shorter-lived radioactive states. Research into the processes for such a transformation, called isomer depletion or switching, has been discussed elsewhere<sup>14,15</sup> and was not part of this project. The reduced activity (number of decays/second) of these radioisotopes when in their isomers would also lead to reduced shielding requirements during ground operations. In this section, we compare the properties of these two long-lived isomers with those of  $^{170}\text{Tm}$  to assess the suitability of the former as alternative options\* for power sources.

**Table 3. Nuclear parameters for isomer- and ground-state decays of  $^{192}\text{Ir}$  and  $^{242}\text{Am}$  (adapted with permission from Carroll,<sup>14</sup> values from ENSDF<sup>1</sup>).**

| Nuclide           | Isomer            |             |          |                              | Ground state |         |                                     |                   |
|-------------------|-------------------|-------------|----------|------------------------------|--------------|---------|-------------------------------------|-------------------|
|                   | $T_{1/2}$ , years | $E_m$ , keV | $J^\pi$  | Decay modes                  | $T_{1/2}$    | $J^\pi$ | Decay modes                         | Decay energy, keV |
| $^{192}\text{Ir}$ | 241               | 168.14      | $(11^-)$ | IT: 100%                     | 73.829 days  | $4^+$   | $\beta^-$ : 95%<br>$\epsilon$ = 5%  | 1455              |
| $^{242}\text{Am}$ | 141               | 48.60       | $5^-$    | IT: 99.5%<br>$\alpha$ = 0.5% | 16.01 h      | $1^-$   | $\beta^-$ : 83%<br>$\epsilon$ = 17% | 664               |

Note: Isomer energies ( $E_m$ ) are given, as well as angular momenta ( $J$ ), parities ( $\pi$ ), and isomer and ground-state decays. Decay modes are as follows: IT = isomer transition (electromagnetic decay toward the ground state);  $\beta^-$  and  $\beta^+$  = electron and positron decay;  $\epsilon$  = atomic electron capture by the nucleus; and  $\alpha$  = alpha decay. Some gamma and conversion-electron emission, and X-rays, from the daughter nuclei are expected after radioactive decays of the parents.

Switching has not yet been demonstrated experimentally for  $^{242\text{m}}\text{Am}$ , but at least one candidate depletion path exists. If this isomer can be switched, after a fast release of energy in the form of  $\gamma$ -rays and conversion electrons, the ground state of  $^{242}\text{Am}$  would be populated, which subsequently decays by  $\beta^-$  emission (with  $\beta$  energies  $E_\beta$  up to 0.664 MeV) to  $^{242}\text{Cm}$  with a 16-h half-life. The  $^{242}\text{Cm}$  ground state then  $\alpha$  decays (energy  $Q_\alpha$  = 6.215 MeV) to  $^{238}\text{Pu}$  with  $T_{1/2}$  = 163 days. No candidate depletion path is currently known for  $^{192\text{m}}\text{Ir}$ , but, if switching is possible, the nucleus would be transformed to a  $T_{1/2}$  = 73.8-day ground state, which would then go on to decay by  $\beta^-$  emission with  $E_\beta$  up to 1.46 MeV.

The ability to switch a nuclide from an isomeric energy-storing state to an energy-releasing, higher-power state has limited practical utility unless sufficient bulk

\* One can consider these to be examples of Next-Generation Radioisotopes, ARL Nuclide Science Team nomenclature.

quantities of the isomer could be produced for these purposes. Therefore, we performed a literature search of known production mechanisms for  $^{242\text{m}}\text{Am}$  and  $^{192\text{m}}\text{Ir}$  and compared them with  $^{170}\text{Tm}$ .

### 2.3 $^{242\text{m}}\text{Am}$ Production

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The prospect of using  $^{242\text{m}}\text{Am}$  as a nuclear fuel for propulsion is not new. Ronen and Leibson<sup>16</sup> proposed the concept decades ago, with further exploration done in subsequent work (e.g., Ronen and Shwageraus<sup>17</sup>). In contrast to the current consideration of  $^{242\text{m}}\text{Am}$  isomers being switched to the respective  $^{242}\text{Am}$  ground states whose decays would serve as a power source, Ronen and Shwageraus's<sup>17</sup> interest was in the form of harnessing neutron-induced fission of the isomer itself as the fuel. A nuclear engine design based on fission of  $^{242\text{m}}\text{Am}$  was described in Ronen et al.<sup>18</sup> Despite the difference in these approaches, a common concern for both is the availability of sufficient quantities of  $^{242\text{m}}\text{Am}$ .

Americium isotopes are known byproducts of the transmutation cycle in light-water reactors (LWRs). Figure 5 illustrates the complex network of decays and neutron captures that occur in such an environment, with the arrow widths representing percentages, calculated in Sasahara et al.<sup>19</sup> with ORIGEN2,<sup>20</sup> that break down the relative amounts by which each nuclide transmutes by following a given path. The production of  $^{242}\text{Am}$  in LWRs is through the  $(n, \gamma)$  capture reaction on  $^{241}\text{Am}$ ,\* indicated towards the center of Figure 5, going predominantly to the ground state but with a smaller fraction also populating the  $^{242\text{m}}\text{Am}$  isomer. In Shinohara et al.'s<sup>21</sup> experiment, the cross sections for thermal neutron capture,  $^{241}\text{Am}(n, \gamma)$ , were measured to be  $\sigma = 85.7(63)$  and  $768(58)$  b to the isomer and to the ground state, respectively. For epithermal neutrons, the corresponding resonance integrals were reported to be  $I_0 = 114(7)$  and  $1694(146)$  b, respectively.<sup>21</sup> These values differ by a factor of approximately 1.5 from those in a previous evaluation,<sup>3</sup> with the resonance integral to the isomer being lower in Shinohara et al.<sup>21</sup> than in Mughabghab<sup>3</sup> and the other three values being larger; this difference was attributed by Shinohara et al.<sup>21</sup> as being partly due to their having a highly purified  $^{241}\text{Am}$  target for their measurements. Although the absolute scales are different, the ratio of the cross section for the  $(n, \gamma)$  reaction to the ground state to the total  $(n, \gamma)$  cross section to  $^{242}\text{Am}$  (isomer plus ground state), whether from thermal or epithermal neutrons, is equivalent in both works.

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\* For decades,  $^{241}\text{Am}$  has been harvested from reactors, processed, and used in microgram quantities in common household smoke detectors.

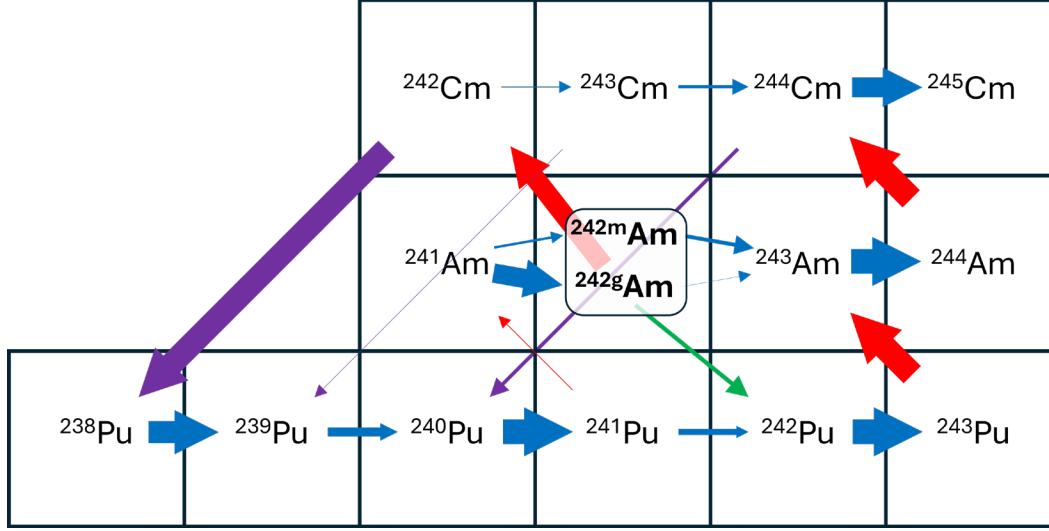


Figure 5. Transmutation of nuclides in LWRs. Arrows represent reaction or decay mechanisms that transform the nucleus into another: blue arrows are for the  $(n, \gamma)$  reaction; red, green, and purple arrows are for  $\beta^-$ , electron-capture, and  $\alpha$  decays, respectively. Arrow widths are proportional to the percentage of the transmutations for a given nuclide by the corresponding decay or reaction mode (adapted with permission from Figure 12 of Sasahara et al.<sup>19</sup>). For example, the arrow width for the  $^{240}\text{Pu}(n, \gamma)^{241}\text{Pu}$  reaction represents about 100% of the  $^{240}\text{Pu}$  transmutations. Fission branches, not indicated in the figure, make up the rest of the yields such that the branches out of each nuclide sum to 100%. The ground state and isomer of  $^{242}\text{Am}$  are populated through the  $^{241}\text{Am}(n, \gamma)$  capture reactions.

Genreith et al.<sup>22</sup> measured the  $(n, \gamma)$  cross section using cold ( $<10$  meV) neutrons. They did not quote the value for neutron capture leading to the  $^{242}\text{Am}$  isomer, but this can be determined from the difference between their reported thermal-equivalent cross sections for production of the ground state and of the total  $^{242}\text{Am}$  to be approximately 62 b (with a likely  $\sim 5\%$  uncertainty).

Competing with this production of  $^{242\text{m}}\text{Am}$  is its subsequent destruction (burn up) through other reactions with thermal neutrons, in this case by induced fission. The cross section for this  $^{242\text{m}}\text{Am}(n, f)$  reaction has been reported to be the largest value known for this process in any nuclide, that is, 6.3 kb.<sup>23</sup> With this cross section being more than 30 times larger than the combined thermal and epithermal  $^{241}\text{Am}(n, \gamma)^{242\text{m}}\text{Am}$  production cross section (200 b), this presents a challenge for bulk production of  $^{242\text{m}}\text{Am}$  through neutron irradiation: after more than approximately 3.2% of a  $^{241}\text{Am}$  sample has been transmuted into  $^{242\text{m}}\text{Am}$ , there is a higher likelihood that a subsequent neutron incident upon the same target will induce fission of an existing  $^{242\text{m}}\text{Am}$  nuclide rather than transmute a  $^{241}\text{Am}$  nuclide into a new one.

Other reactions in which cross sections for  $^{242}\text{Am}$  production have been measured are in deep-inelastic collisions between  $^{129,132}\text{Xe}$  and  $^{248}\text{Cm}$ <sup>24</sup> and between  $^{56}\text{Fe}$  and  $^{209}\text{Bi}$ .<sup>25</sup> The total cross section to  $^{242}\text{Am}$  was reported as  $\sigma \sim 0.3$  mb for the former

and  $<20 \mu\text{b}$  for the latter, with no distinction made for population of the isomer versus the ground state.

A series of reactions was evaluated by Rzadkiewicz et al.<sup>26</sup> for their cross sections for  $^{242}\text{Am}$  production with GEMINI++, a Monte-Carlo fusion-evaporation code. A spin cutoff was applied to preferentially populate the isomer ( $5^-$  state) over the ground state ( $1^-$ ). The reaction  $^{242}\text{Pu}(d, 2n)$  was found to have a maximum cross section of approximately 300 mb; for  $^{238}\text{U}(^7\text{Li}, 3n)$ , the maximum was down to approximately 30 mb; and the cross section for  $^{241}\text{Pu}(d, n)$  was significantly lower. These predictions have not been experimentally verified, but in all considered cases, the yields would be far less than for neutron-induced reactions.

One critical point that must be considered when producing bulk quantities of  $^{242\text{m}}\text{Am}$  is criticality. Dias et al.<sup>27</sup> found that a bare metal sphere of  $^{242\text{m}}\text{Am}$  has a critical mass of approximately 9 to 14 kg, depending on which nuclear-data libraries were used for the calculations. This is comparable to the critical masses for  $^{238,239}\text{Pu}$ , which Blanchard et al.<sup>28</sup> list as approximately 10 kg as well. If surrounded by water or steel reflectors, the  $^{242\text{m}}\text{Am}$  critical mass reduces to approximately 3 to 5 kg; if used in oxide form,  $\text{AmO}_2$ , the critical masses are determined to be approximately 10% to 20% larger.<sup>27</sup> Ronen and Shwageraus<sup>17</sup> performed a criticality calculation using a different geometry: a cylinder of dispersed  $^{242\text{m}}\text{Am}$  fuel, 1 m in diameter and 5 m high, surrounded by a 3-m-thick, heavy-water reflector. This configuration yielded a much smaller critical mass (0.41 kg) than reported in Dias et al.<sup>27</sup> Clearly, bulk production of  $^{242\text{m}}\text{Am}$  must weigh the impact of the geometry, overall mass, and reflectors for a sample that is to be used as an isomer power source.

## 2.4 $^{192\text{m}}\text{Ir}$ Production

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The cross section for  $^{192}\text{Ir}$  production has been measured for a variety of reaction mechanisms. In general, however, these cross sections are reported for the total  $^{192}\text{Ir}$  yield, with no distinction for population of the long-lived isomer. In contrast to  $^{242}\text{Am}$ , where the spin difference between the ground state ( $J = 1$ ) and isomer ( $J = 5$ ) is relatively small, in  $^{192}\text{Ir}$  the isomer has spin  $J = 11$ . This moderately high spin is not as likely to be populated in reactions that do not necessarily bring sizable angular momentum into the system, such as neutron-induced reactions. The isomer yield, consequently, may be considerably lower than the quoted measurements; the latter values should be treated as upper limits unless noted otherwise. Several examples from the literature are provided below.

Gantumur et al.<sup>29</sup> measured the cross section for  $^{192}\text{Ir}$  production with protons incident upon a  $^{\text{nat}}\text{Pt}$  target. They obtained a value of approximately 1.4 mb at a proton energy of 30 MeV. The cross section increases with energy; 30 MeV was

the highest energy measured, although the cross section probably does not increase that dramatically beyond this energy.

The creation of  $^{192}\text{Ir}$  through fragmentation of an 85-MeV/nucleon  $^{198}\text{Pt}$  beam (16.8 GeV) was investigated by Haak et al.<sup>30</sup> A total cross section of approximately 70 mb was measured. The isomer fraction was not specified.

Kalamara et al.<sup>31</sup> measured the  $^{193}\text{Ir}(n, 2n)$  reaction at a neutron energy of 11.3 MeV obtaining a cross section of approximately 2 b. For comparison, we input this reaction into the Monte Carlo fusion-evaporation code PACE4<sup>32</sup> and similarly found 2 b at this energy for the total cross section to  $^{192}\text{Ir}$ , with no distinction between isomer and ground-state yields. Requiring a compound-nucleus spin of at least  $J = 9$ , to favor feeding of the  $11^-$  isomer  $^{192m}\text{Ir}$ , reduces this cross section to approximately 22% of the total, thus leading to the crude estimate of an approximately 0.4-b cross section for isomer production via  $(n, 2n)$  at this energy. Increasing the neutron energy to 14.1 MeV in the PACE4 calculation leads to an estimated cross section to  $^{192m}\text{Ir}$  of approximately 0.7 b.

Other neutron-induced reactions, identified via the Evaluated Nuclear Data File (ENDF) reactions database,<sup>33</sup> include  $^{192}\text{Pt}(n, p)$ , with  $\sigma \sim 10$  mb at a neutron energy of 30 MeV, and the  $^{195}\text{Au}(n, \alpha)$  reaction, with a cross section of, at best, a few megabarns. The largest measured cross section leading to  $^{192}\text{Ir}$  is, by far, for the thermal neutron capture reaction,  $^{191}\text{Ir}(n, \gamma)$ . A total cross section of 954(10) b was reported for this reaction.<sup>34</sup>

This literature search did not reveal any studies in which the cross section for population of the  $^{192m}\text{Ir}$  isomer was explicitly reported. To gauge the potential rate of production with  $(n, \gamma)$  reactions, the experimental details of the work that identified the isomer for the first time<sup>35</sup> were used in an estimate. For that measurement, the researchers produced the isomer by putting a  $\sim 50$ -mg sample of natural Ir in the Brookhaven reactor for 1 month and then allowing it to cool. (The initial cooling period was 2 years, but the measurement of  $^{192m}\text{Ir}$  ultimately came 7–8 years after irradiation.) The activity at that time was 2820 Bq  $\approx$  76 nCi. Using the relationship  $A(t) = \left(\frac{\ln 2}{T_{1/2}}\right) N(t)$  between activity as a function of time, the half-life  $T_{1/2} = 241$  years  $= 7.60 \times 10^9$  s, and the number of nuclei present  $N(t)$ , the number of isomeric  $^{192}\text{Ir}$  nuclei produced after 1 month of irradiation and surviving an approximately 7-year cool-down period is  $N(\sim 7 \text{ years}) = 3.1 \times 10^{13}$ . Working backward by  $t = 7$  years to the end of the irradiation period, before cooling, using the equation  $N(t) = N(0)e^{-t \ln 2 / T_{1/2}}$ , the number of  $^{192m}\text{Ir}$  isomers produced after 1 month in the reactor was  $N(0) = 3.2 \times 10^{13}$ . At 192 g/mol, this amounts to  $1.0 \times 10^{-8}$  g of  $^{192m}\text{Ir}$  produced from an initial approximately 0.05 g of  $^{\text{nat}}\text{Ir}$  after 1 month

of irradiation in the reactor. If a use case (power profile) for  $^{192\text{m}}\text{Ir}$  as an isomer power source required 1 kg of the isomer, a factor of  $10^{11}$  times more mass would be needed, which would come from an increase in the initial sample size, the irradiation time, and/or the neutron flux. Assuming the flux to be the same (this value was not specified in Scharff-Goldhaber and McKeown<sup>35</sup>) and a  $^{\text{nat}}\text{Ir}$  sample size of 10 kg ( $\sim 2 \times 10^5$  times larger than in Scharff-Goldhaber and McKeown<sup>35</sup>), this still requires  $5 \times 10^5$  times the duration of irradiation, or  $4.2 \times 10^4$  years, which clearly is impractical. This highlights the major issue with  $^{192\text{m}}\text{Ir}$  production in bulk quantities.

## 2.5 Comparisons for $^{170}\text{Tm}$ Production

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How do the cross sections for  $^{242\text{m}}\text{Am}$  or  $^{192\text{m}}\text{Ir}$  production compare with that of  $^{170}\text{Tm}$ ? In this section, we collect some of the findings from the literature for a variety of reactions leading to  $^{170}\text{Tm}$  (ground state in this case, not an isomer).

First, we examine reactions induced by light charged particles: for the  $^{170}\text{Er}(p, n)^{170}\text{Tm}$  reaction, a cross section of  $\sigma \sim 0.1$  b for protons in the 8- to 20-MeV range was measured by Dellepiane et al.<sup>36</sup> Saito et al.<sup>37</sup> bombarded a natural Er target with  $\alpha$  particles, producing  $^{170}\text{Tm}$  through the  $^{167}\text{Er}(\alpha, p)$ ,  $^{168}\text{Er}(\alpha, d)$ , and  $^{170}\text{Er}(\alpha, \text{tn})$  reactions; the combined cross section for these processes was approximately 15 mb for  $E(\alpha)$  up to 50 MeV. For deuteron-induced reactions, Khandaker et al.<sup>38</sup> studied the  $^{\text{nat}}\text{Er}(d, 2n)$  reaction over a range of deuteron energies, finding the maximum yield of approximately 0.1 b for  $E(d) \sim 12$  MeV. (The research reports the reaction as the nonspecific  $^{\text{nat}}\text{Er}(d, x)^{170}\text{Tm}$ , where  $x$  is meant to represent any outgoing particles; however, the only possible reaction other than  $(d, 2n)$  leading to  $^{170}\text{Tm}$  would be  $(d, \gamma)$ , i.e., fusion of the  $d$  and  $^{168}\text{Er}$  with no massive particles subsequently emitted, which would not peak as high in energy as shown in Figure 7 of Khandaker et al.<sup>38</sup>) Saito et al.<sup>39</sup> looked at deuterons incident upon a  $^{169}\text{Tm}$  target, the  $^{169}\text{Tm}(d, p)^{170}\text{Tm}$  reaction, and found the cross-section peaks with a maximum of approximately 0.17 b at  $E(d) \sim 10$  MeV.

Turning to heavier ions, the  $^{169}\text{Tm}(^9\text{Be}, ^8\text{Be})^{170}\text{Tm}$  1n-transfer reaction was studied by Fang et al.<sup>40</sup> The cross section increases with energy from 32 to 38 MeV, then starts to level off, “saturating” at a maximum of approximately 0.15 b. In the latter region, complete fusion begins to dominate over the 1n transfer; thus, it does not significantly contribute to any additional  $^{170}\text{Tm}$  production.

Like the case for  $^{242\text{m}}\text{Am}$  and  $^{192\text{m}}\text{Ir}$ , the neutron-capture reaction is the winner for  $^{170}\text{Tm}$  in terms of the overall cross section. In the neutron-energy region from eV to MeV, Ren et al.<sup>41</sup> measured the cross sections as plotted in Figure 6 (left panel). The cross section peaks at  $\sigma \sim 20$  kb for a resonance at  $E(n) \sim 4$  eV, whereas in the

tens of keV region it is more like 1 b. Extending down into the thermal-neutron domain, Takiue et al.<sup>42</sup> report  $\sigma = 105(2)$  b for thermal neutron capture; this result is generally supported by other works by Zimmerman et al.<sup>43</sup> and by Sims and Juhnke.<sup>44</sup> The thermal neutron cross section to produce  $^{170}\text{Tm}$  is comparable to that for production of  $^{242\text{m}}\text{Am}$ .

### 3. Effective Dose Rate and Emitted Power Scaling: $\beta^-$ Decay

As shown in Table 1, radioactive decay of  $^{170}\text{Tm}$  can occur by different modes following given average probabilities; for example, 81.9% of the time  $\beta^-$  decay occurs with a  $Q_\beta = 968.1$  keV. The  $Q_\beta$  represents the total energy released in that particular decay, but an electron is almost never emitted from the nucleus carrying that total energy. The  $\beta^-$  decay is a three-body process in which the total energy is shared among the daughter nucleus, an electron, and an antineutrino. Thus, to correctly simulate  $^{170}\text{Tm}$   $\beta^-$  decay, it is necessary to model the  $\beta^-$  energy distribution (depicted in Figure 6) instead of treating them as monoenergetic particles. For example, one particular  $^{170}\text{Tm}$  decay event in the simulation might be randomly selected to have a  $\beta$  electron emitted with an energy of 400 keV with the remaining  $Q_\beta - 400$  keV = 568.1 keV being “lost” to the nuclear recoil or the essentially undetectable antineutrino. This particular event would not be accompanied by emission of a gamma ray by the daughter  $^{170}\text{Yb}$  nucleus because this decay with  $Q_\beta = 968.1$  keV would correspond to a ground-state-to-ground-state occurrence (shown in the graphic on page 19). Another simulated  $^{170}\text{Tm}$  decay event might also be a  $\beta^-$  emission but from the  $Q_\beta = 883.8$  keV branch, leading to the first excited state of the daughter and thus accompanied by essentially simultaneous emission of a gamma ray at 84.3 keV or a conversion electron following the established probabilities (see ENSDF<sup>1</sup>). In all, full simulation of  $^{170}\text{Tm}$  decays must occur by sampling all decay possibilities according to the known mean probabilities and decay branches (shown in Table 1 and Figure 7). Early models for this project used direct electron emission with the energies determined by a normalized probability distribution function (like that shown in Figure 6) built to match the  $\beta^-$  decay energies from  $^{170}\text{Tm}$ . In addition, Figure 6 shows that Bremsstrahlung photons produced from electrons of a given energy also fall within a spectrum of values whose maximum value (the spectrum’s “endpoint”) is the electron energy; Bremsstrahlung production occurs intrinsically during a Geant4 simulation of electron transport. Subsequently, more detailed models used the Geant4 radioactive decay libraries to inject  $^{170}\text{Tm}$  nuclei into a volume, which would then undergo radioactive decay following the known probabilities for all modes.

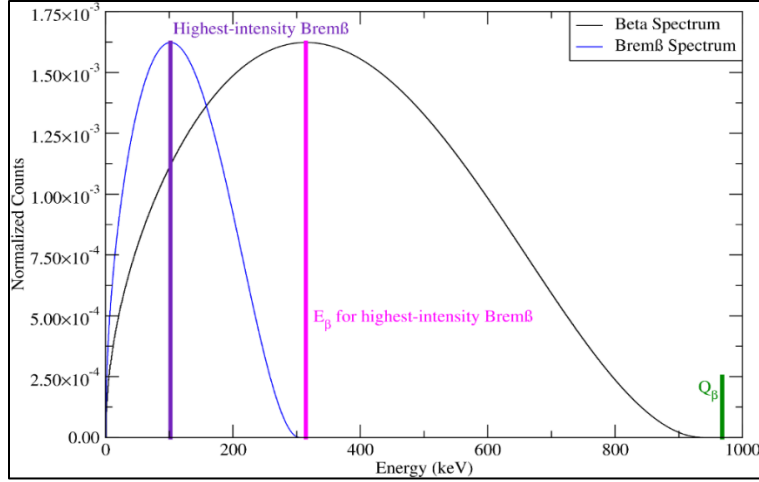


Figure 6. Schematic of  $\beta$  and Bremsstrahlung spectra showing the relationship between the end-point energies and spectral maxima. The Bremsstrahlung spectrum shown is that generated only by the electrons at the energy corresponding to the highest intensity of electrons within the  $\beta$  spectrum.

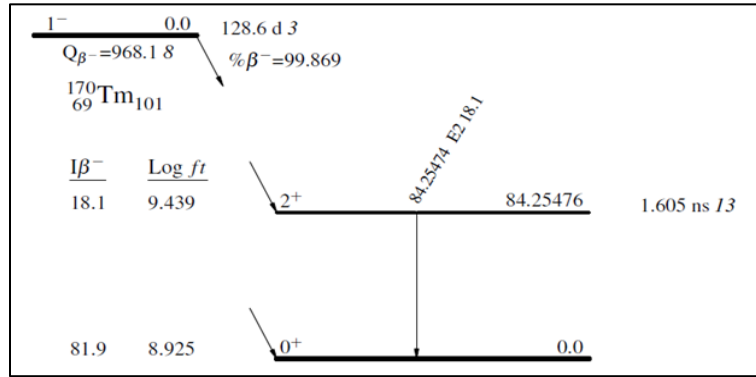
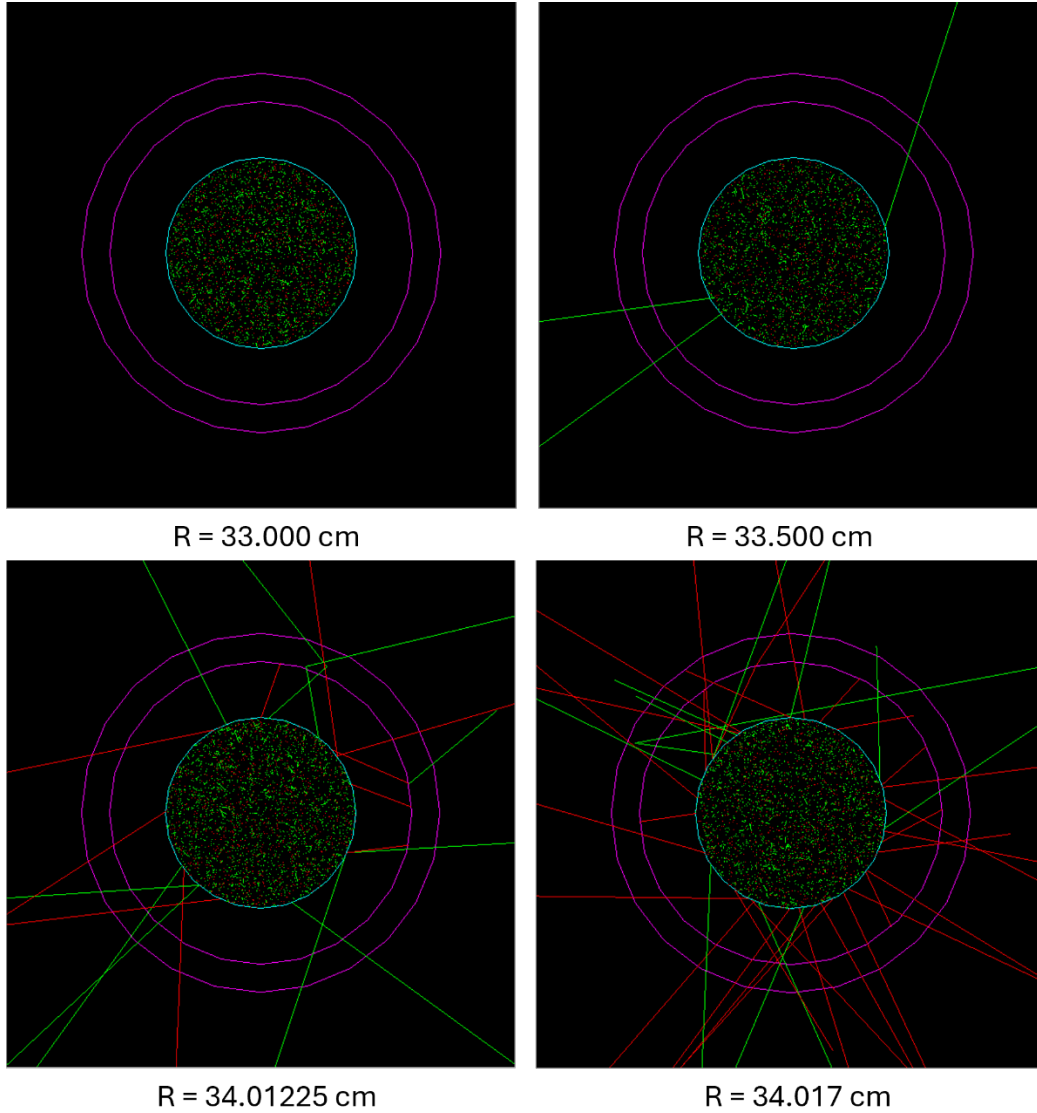


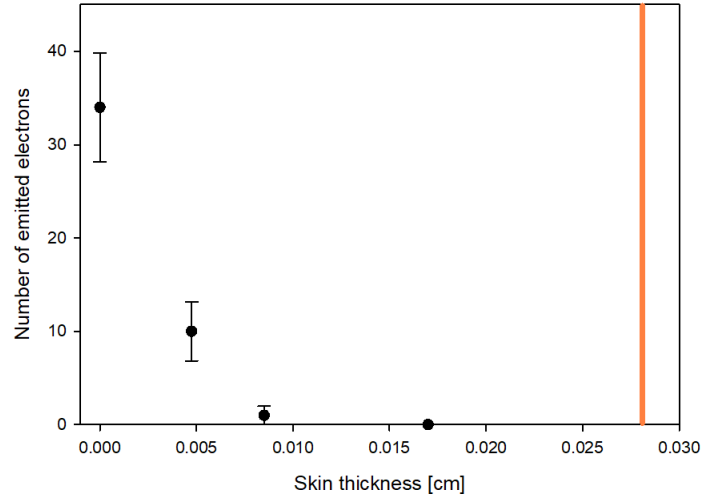
Figure 7. Diagram from ENSDF<sup>1</sup> showing the  $\beta^-$  decays branches of  $^{170}\text{Tm}$  to  $^{170}\text{Yb}$ .

For  $\beta^-$  emitters like  $^{170}\text{Tm}$ , one can expect significantly different scaling behavior of the emitted decay power compared with the effective dose rate at locations outside the volume. The  $\beta^-$  particles will have a limited range within matter, depending on their energies, such that they will only be able to escape from a relatively narrow surface region of the volume. Of course, Bremsstrahlung created by those electrons and any gamma rays accompanying the  $\beta^-$  decays can penetrate matter more effectively. Nevertheless, photons generated deep within the radioisotope material will experience attenuation. One can expect that the fraction of the total emitted radiation that escapes a volume will be reduced as the size of the volume is increased, in a similar way that the surface area increases more slowly than the size of the enclosed volume. Conversely, the fraction of the total emitted radiation that is retained within the volume to generate heat will increase as the volume becomes larger.

The impact of these scaling behaviors was investigated quantitatively via Geant4 simulation using a simplified radioactive material geometry as a solid cylinder of uniform density  $\text{Tm}_2\text{O}_3$  with 10%  $^{170}\text{Tm}$  content. Figure 8 illustrates that the simulations agree with the expectation that only a thin surface layer (skin) is responsible for electron and photon emission from the volume's interior into the outside space. One would also expect that the skin for electron emission would be much thinner than for photons due to the known stopping power and range for charged particles versus the attenuation behavior of photons. Figure 9 shows the general expected trend, albeit with poor statistical accuracy, for the reduction of electrons that escape from the volume based on an increased emission depth from within the volume. The Continuous Stopping Distance Approximation (CSDA) range represents the stopping of electrons originally emitted in a collimated beam directed perpendicular to a semi-infinite slab of stopping material. For the simulation, the direction of electron emission is rarely radially outward, so the path lengths of electron trajectories directed toward escape are typically much larger than the actual skin thickness. Thus, the effective skin thickness for stopping electron escapes is much less than the CSDA range.

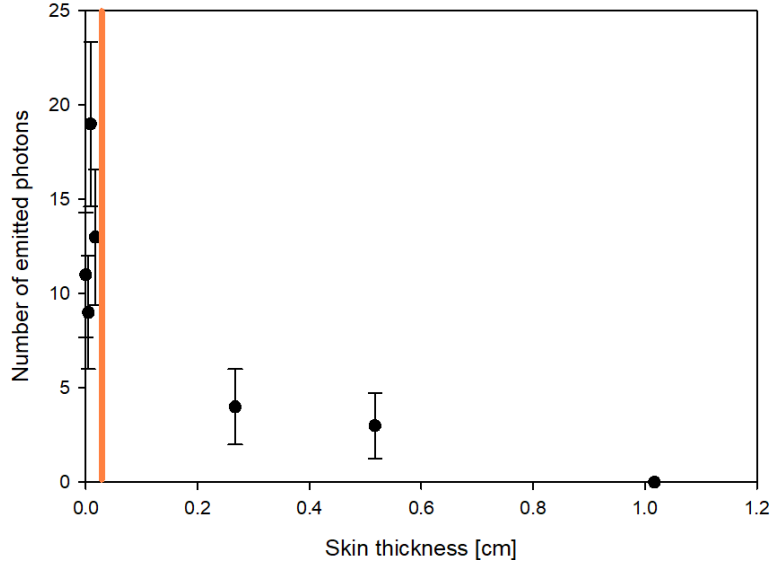


**Figure 8.** Simulation results illustrating the limited range of surface thickness (“skin”) from which radiation can escape from within the radioactive volume. Only 10,000 histories were simulated to allow easy identification of trajectories (green = emitted electrons, red = photons). The outer radius of the volume was fixed at 34.017 cm, whereas the radius of the volume from which isotropic 500-keV electron emission occurred was changed as shown above, where the radii of the emitting volumes are listed. The length of the emitting volume was sufficiently less than that of the total material volume so that all radiation-escape trajectories shown are from the cylindrical sides of the material volume and not the ends. The remainder of the simulation space is a vacuum.



**Figure 9.** Plot of the numbers of electron tracks escaping the cylindrical surface of the radioactive volume from Figure 8 as a function of the skin thickness (the thickness of  $\text{Tm}_2\text{O}_3$  material between the outer cylindrical surface of the emitting volume and that of the total radioactive volume). Error bars reflect one standard deviation based on counting statistics. The orange line presents the CSDA range of 0.028 cm for 500-keV electrons.<sup>45</sup>

Figure 10 shows the number of escaping photons as a function of skin thickness, with a measured attenuation that falls with a half-value thickness of approximately 0.2 cm. Although the initial electrons are monoenergetic (500 keV), the photons are part of a continuous Bremsstrahlung spectrum with 500-keV endpoint. The (extremely few) photons near 500 keV would experience an attenuation coefficient of  $0.12 \text{ cm}^{-1}$  corresponding to a half-value thickness of 0.66 cm, whereas photons with 167-keV energies (near the peak of the Bremsstrahlung spectrum) would experience a half-value thickness of 0.088 cm. Thus, the effective half-value thickness from the spectrum of photons should lie between 0.088 and 0.66 cm, which the measured 0.2 cm satisfies. Although Figures 9 and 10 provide numerical values, those results were from very limited (10,000-history) simulations for purely illustrative purposes. According to Figure 10, over the range of depths from which electrons can escape (to the left of the CSDA range shown in Figures 9 and 10 by the orange lines), no statistically different change was observed in the number of photon (Bremsstrahlung) tracks escaping the radioactive volume. This is expected because this distance is small compared with the half-value thickness for photons.

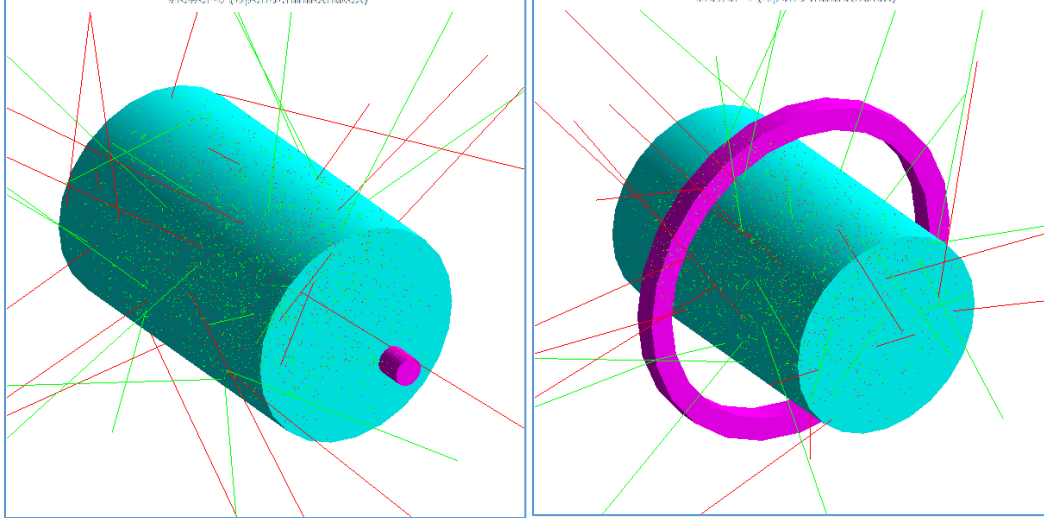


**Figure 10.** Plot of the numbers of photon tracks escaping the cylindrical surface of the radioactive volume (from Figure 8) as a function of the skin thickness (the thickness of  $\text{Tm}_2\text{O}_3$  material between the outer cylindrical surface of the emitting volume and that of the total cylindrical volume). Error bars reflect one standard deviation based on counting statistics. The CSDA range for 500-keV electrons is shown by the orange line for reference.

The actual effect of self-shielding of the radioactive volume was evaluated using simulations with isotropic emission of monoenergetic 500-keV electrons within emitting volumes that matched the total right circular cylinders) in each simulation. The total volumes were incremented while keeping the aspect ratio of  $L/R$  constant, with the smallest volume  $V$  containing  $R = 27$  cm and  $L = 80$  cm. The simulations for quantitative results were all conducted with  $10^9$  (a convenient value that is close to the Geant4 signed-integer limit) electron-emission histories. For the same density of emitting  $^{170}\text{Tm}$  nuclei, the number of emissions should increase with increased volume. To accommodate this, the output results from each simulated volume were scaled upward according to the ratio of the simulated volume to  $V$ . A scoring volume containing water at  $1\text{-g cm}^{-3}$ , as a surrogate human phantom, was used for recording the energy deposited by electrons and photons that escaped from the radioactive cylinder. The distance from the outer surface of the radioactive cylinder to the near surface of the scoring volume was kept constant as was the thickness of the scoring volume.

Figure 11 illustrates the geometries used to investigate the axial and radial behaviors of the radiation dose, for a radioactive volume four times larger than the standard volume,  $V$ . The radioactive volume is depicted by the teal cylinders (Figure 11). The dose-scoring volume is depicted by magenta: for axial scoring, the face of the cylindrical scoring volume was maintained at 20 cm from the near face of the radioactive volume, whereas for radial scoring a ring volume was used and

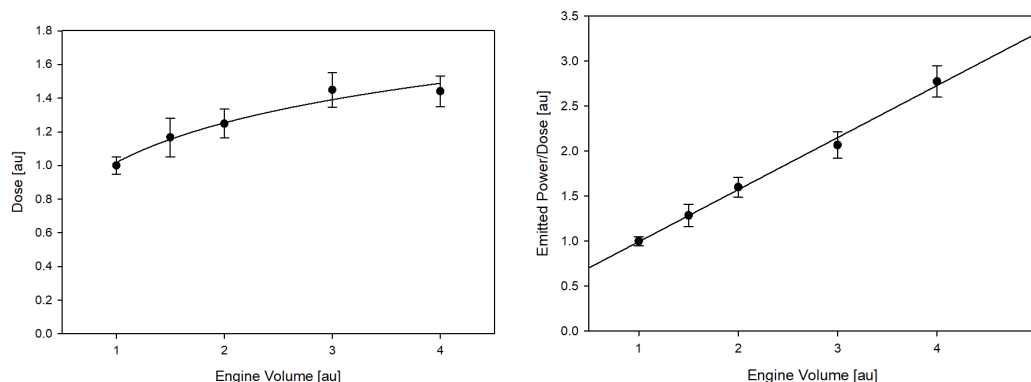
the gap between the inner surface of the scoring was maintained at 20 cm from the outer surface of the radioactive volume (Figure 11). The ring was placed around the midpoint of the radioactive volume. For Figure 11, simulations were run with 10,000 electron-emission histories.



**Figure 11.** Geometry for a 4V radioactive volume (teal) with (left) axial and (right) radial scoring volumes (magenta). Electron trajectories are shown in red while photon trajectories from X-rays and Bremsstrahlung are shown in green. The remainder of the simulation volume is a vacuum.

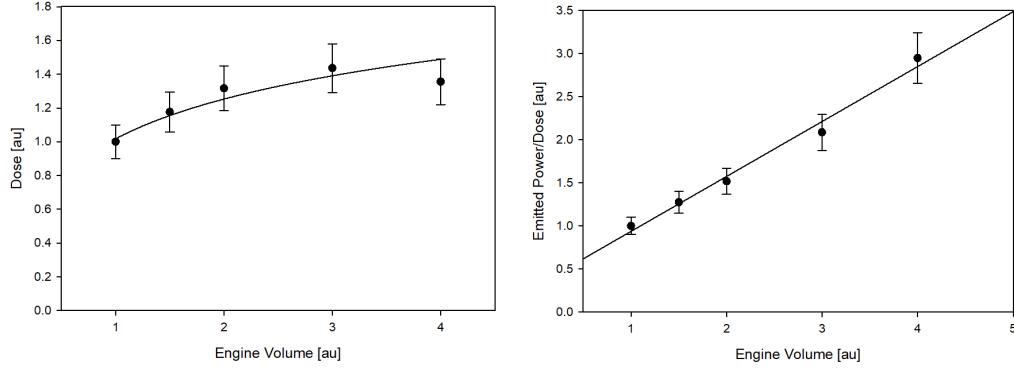
Figure 12 shows the results obtained for an axial scoring volume kept at a constant spacing from the end of the radioactive volume, for volumes of  $V$ : 1.5V, 2.0V, 3.0V, and 4.0V. Error bars were determined from the standard deviation between multiple identical (except for random number seeds) simulations for each volume; the largest error was just under 10%. The left plot in Figure 12 shows the dose absorbed by the scoring volume as a function of radioactive volume and exhibits a trend toward saturation. This behavior should be expected because electrons and photons emitted from the radioactive volume should only be able to escape from that volume from within a relatively short depth inside the volume (quite small for the 500-keV electrons, larger for photons depending on their energies). Thus, as the total radioactive size is enlarged while maintaining the  $L/R$  aspect ratio, the important emitting depth does not increase (that depth is much smaller than  $L = 80$  cm for the smallest volume), which means that the important emitting depth is always present. Although the important emitting depth does not change in the simulations for different volumes, the area of the end face does increase as the radioactive volume is enlarged. Thus, the total dose delivered to the axial scoring volume increases, but it does so at a reduced rate as the radioactive volume is enlarged. This indicates that when considering the radiation dose to positions outside a cylindrical radioactive volume, there is an economy of scale in that a

larger radioactive volume is beneficial in increasing the total emitted power (proportional to the radioactive volume) while modestly increasing the exterior dose rate. This is illustrated further in the right plot in Figure 12, which shows the ratio of emitted power to the external axial dose. If the dose were to scale with volume in the same way as the power does, the plot on the right would be a horizontal line; the positive slope indicates larger gains in the power compared with the increase in dose as a function of volume.



**Figure 12.** (Left) Radiation dose absorbed in the axial scoring volume as a function of radioactive volume. (Right) Power in radioactive emission per dose absorbed in the axial scoring volume as a function of radioactive volume. Arbitrary units are used for radiation dose, emitted power, and radioactive volume to focus on the systematic trends.

Figure 13 shows the results obtained for a radial scoring volume kept at a constant spacing between its inside curved surface and the outer curved surface of the radioactive volume, for volumes of V, 1.5V, 2.0V, 3.0V, and 4.0V. Error bars were at the 10% level based on the variation in the axial simulations. Since the constant radial spacing was maintained, the absorbed dose in the scoring volume was scaled to account for the increased size of the scoring volume. The same trend is seen for the axial simulations, where economy of scale indicates that a larger radioactive volume is best for improving increased emitted power while only modestly increasing the radiation dose to the exterior in a radial direction. In practice, this means that the thickness of shielding required for greater emitting volumes (with ever-greater power outputs) will not increase as quickly as the increase in volume. However, the total amount of shielding may still increase to enclose a larger radioactive volume.



**Figure 13.** (Left) Radiation dose absorbed in the radial scoring volume as a function of radioactive volume. (Right) Power in radioactive emission per dose absorbed in the radial scoring volume as a function of radioactive volume. Arbitrary units are used for radiation dose, emitted power, and source volume to focus on the systematic trends.

## 4. Conclusions

Production of the radioisotopes  $^{148}\text{Gd}$ ,  $^{191\text{m}}\text{Ir}$ , and  $^{242\text{m}}\text{Am}$  were considered and were based on currently available reaction cross sections. Of these three radioisotopes, production of sufficient quantities of  $^{242\text{m}}\text{Am}$  is most feasible and achievable using neutron capture. However, the burn up of this radioisotope, once produced, by subsequent neutron capture would require specific steps such as frequent replacement of samples to be included in a full-scale production design. Such a design was not part of this effort. Direct production cross sections by neutron capture are comparable for  $^{170}\text{Tm}$  and  $^{242\text{m}}\text{Am}$ .

A simplified simulation geometry was used to investigate the scaling of the radiation dose at exterior positions when the radioactive volume size with a constant density of  $^{170}\text{Tm}$  emitters was varied. Because radiation only escaped from a relatively thin shell of the radioactive volume (thinner for electrons than for Bremsstrahlung photons), the radiation dose delivered to either radial or axial positions at a constant distance from the nearest surface of the cylindrical radioactive volume tended toward a saturation-like behavior; continuing to increase the radioactive volume's size led to only modest increases in the external radiation dose. Meanwhile, the total power output from the radioactive decay simply increased proportionately to the volume of the radioactive material. The conclusion is that the larger the radioactive volume can be made, the more benefit in that the available decay (and thermal) power will increase without proportionately increasing the radiation dose to an external location that would otherwise require proportionately more shielding thickness.

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## List of Symbols, Abbreviations, and Acronyms

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|                  |                                                   |
|------------------|---------------------------------------------------|
| AFRL             | Air Force Research Laboratory                     |
| ARL              | Army Research Laboratory                          |
| CSDA             | Continuous Stopping Distance Approximation        |
| DEVCOM           | U.S. Army Combat Capabilities Development Command |
| ENDF             | Evaluated Nuclear Data File                       |
| ENSDF            | Evaluated Nuclear Structure Data File             |
| LANSCCE          | Los Alamos Neutron Science Center                 |
| LWR              | Light-Water Reactor                               |
| MCNP             | Monte Carlo N-Particle                            |
| OECIF            | Operational Energy Capability Improvement Fund    |
| ORNL             | Oak Ridge National Laboratory                     |
| T <sub>irr</sub> | Irradiation Period                                |

1 DEFENSE TECHNICAL  
(PDF) INFORMATION CTR  
DTIC OCA

1 DEVCOM ARL  
(PDF) FCDD RLB CB  
TECH LIB