

# **Effect of Progeny and Trace Contaminants on Hazard Categorization Assessments**

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## **ABSTRACT**

In 2006, a review of the inventory practices for below Hazard Category (HC) -3 facilities at the Oak Ridge National Laboratory (ORNL) noted the characterization of sealed sources and other research related source materials were, in general, only defined by the predominant isotope associated with the specimen. An extensive effort was undertaken to update the inventory of these facilities to include progeny and trace contaminants that can significantly impact the hazard categorization of the specimen.

The contribution of contaminants to the sum of fraction (SOF) assessments in below HC-3 facilities required an evaluation of the processes through which the radioactive source material was produced. The production processes (reactor, accelerator, natural sources) and the physical separation processes (chemical treatment, mass separator) all contribute to determining the final purity of the radioisotopes.

The role of progeny in the sum of fraction (SOF) assessments in the below HC-3 facilities required the application of basic radioactive decay principals to determine how daughters impact the overall sum of fractions over the life span of radioactive source materials. Based on the results of the progeny and trace contaminants assessments, ORNL below HC-3 facility inventories have been updated.

## **Introduction**

Hazard categorization assessments are by design an inventory based assessment. The potential risks associated with radioactive material operations has been quantified through activity based inventory limits. These radioisotope specific limits, known as Threshold Quantities (TQs), serve as the correlation between facility radioactive material inventory and the potential dose personnel could receive if the radioactive material was released due to a postulated facility event. Accordingly, it is of the utmost importance that the facility source term (i.e. the facility radioactive material inventory) be correctly defined since a hazard categorization assessment is based solely on specific radioisotope activity/mass limits.

Radioactive material, by its very nature, cannot be characterized existing in a pure radioisotope form. The processes through which radioisotopes are produced and purified cannot completely eliminate contaminant radioisotopes. Even if a radioisotope could be obtained in a 100% pure form, the radioactive decay process will result in the in-growth of progeny isotope(s) which could be radioactive. In terms of hazard categorization, the presence of contaminants and progeny require consideration if the dose potential is not negligible.

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In an effort to define the source term associated with the facility radioactive material inventory, facilities will utilize information and resources available to them on an as needed basis. Preferably, assay data is obtained to identify the spectrum (i.e. activity / concentration) of radioisotopes present in a specimen. This assay data can be used to accurately define the facility radioactive material source term.

On occasion, the ability to perform an assay on radioactive material is not feasible. This is typical for sealed sources and other encapsulated materials, where the radioactive material is not directly accessible. For this type of material, the original radioisotopic assay data is required for defining the source term. However, as usually the case, only the “predominant” isotope is known. In the cases the question then becomes, what other radioisotopes are potentially in the source material and what would be the impact to the hazard categorization assessment?

In such cases, a defining of the source term for material only characterized by its predominant isotope involves an investigation of the source material which involves resolution of the following questions; (i) where did it come from / who fabricated the source material, (ii) how was it made / what is the source material it originated from and, (iii) how old is it. The ability to answer these three questions can aid in defining the potential contaminants and progeny that are present in the source material.

In the following sections, a discussion of various predominant isotope source materials is provided along with examples to demonstrate the expected contaminants and progeny that can impact a hazard categorization of the source material. Keep in mind, these are examples that can be used as starting point to define the source term. Holders of source material without complete assay are encouraged to research the source material to confirm the radioisotopic content.

## Contaminants

### Background

#### What are Contaminants?

Contaminants are radioisotopes contained in a specimen that were not removed in the purification of the intended radioisotope(s) (excluding progeny produced subsequent to the purification process).

In many cases, contaminants are radioisotopes of the same element. For example, a  $^{137}\text{Cs}$  specimen purified from fission product material will contain an almost equal mass of  $^{135}\text{Cs}$  and trace quantities of  $^{134}\text{Cs}$ . When  $^{238}\text{Pu}$  is undergoes neutron bombardment, a whole spectrum of plutonium radioisotopes can be produced ( $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$ , etc). Chemical separation processes will only separate the plutonium radioisotopes from other elements, the plutonium spectrum will be unaffected by the separation process.

#### Effect Contaminant Have On Hazard Categorization Assessments

The effect that a contaminant can have on a sum of TQ fractions assessment depends on the following parameters:

- The threshold quantity of the respective radioisotopes present, and
- The actual quantity (mass or activity) of the radioisotopes present.

If a radioisotope “A” contains a contaminant radioisotope “B”, the overall sum of TQ fractions is expressed as:

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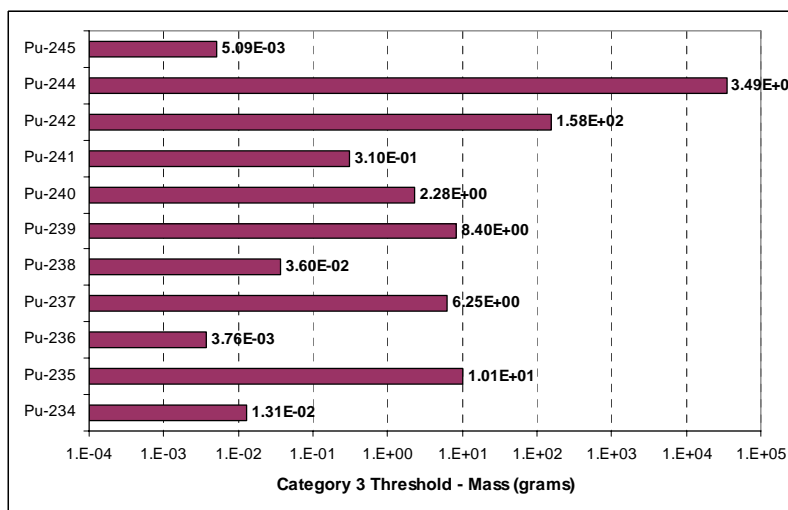
$$\text{SOF} = \sum \frac{M_i}{\text{TQ}_i} = \left( \frac{M_A}{\text{TQ}_A} + \frac{M_B}{\text{TQ}_B} \right) \quad \text{Eqn 1}$$

The threshold quantities “TQ<sub>i</sub>” and “TQ<sub>A</sub>” are the mass TQs. The relative effect that the contaminants, in this case radioisotope “B”, have on the overall sum of fractions is expressed as:

$$f_{\text{SOF}} = \frac{\sum \frac{M_i}{\text{TQ}_i}}{\left( \frac{M_A}{\text{TQ}_A} \right)} = \frac{\left( \frac{M_A}{\text{TQ}_A} + \frac{M_B}{\text{TQ}_B} \right)}{\left( \frac{M_A}{\text{TQ}_A} \right)} \quad \text{Eqn 2}$$

When contaminants are not present (i.e.  $\sum(M_i/\text{TQ}_i) = M_A/\text{TQ}_A$ ), then  $f_{\text{SOF}} = 1.0$ . As the quantity of contaminants increases, then  $f_{\text{SOF}}$  becomes larger than 1.0. As noted in Equation 2, the effect of the contaminant on the value of  $f_{\text{SOF}}$  depends on the contaminant mass and threshold quantity. Contaminants with low TQs will have a greater impact on the SOF and  $f_{\text{SOF}}$  than contaminants with higher TQs, on an equal mass basis.

In Figure 1, the threshold quantities of plutonium radioisotopes from the LANL Fact Sheet for HC-3 Threshold Quantities<sup>1</sup> can be used to illustrate this effect.



**Figure 1. Comparison of HC-3 TQs for plutonium radioisotopes**

As illustrated in Figure 2, a <sup>238</sup>Pu specimen requires a significant amount of <sup>239</sup>Pu contamination to result in a higher SOF. In fact, on a mass basis, the <sup>238</sup>Pu specimen would have to be predominantly <sup>239</sup>Pu. The opposite case however reveals the sensitivity of <sup>238</sup>Pu contamination in <sup>239</sup>Pu specimens. Even at 1 mass % <sup>238</sup>Pu contamination, the SOF factor increases to 3.36. Accordingly, the impact of a contaminant radioisotope with a lower TQ (e.g., <sup>238</sup>Pu) on specimen with a radioisotope of a higher TQ (e.g., <sup>239</sup>Pu) is quite significant.

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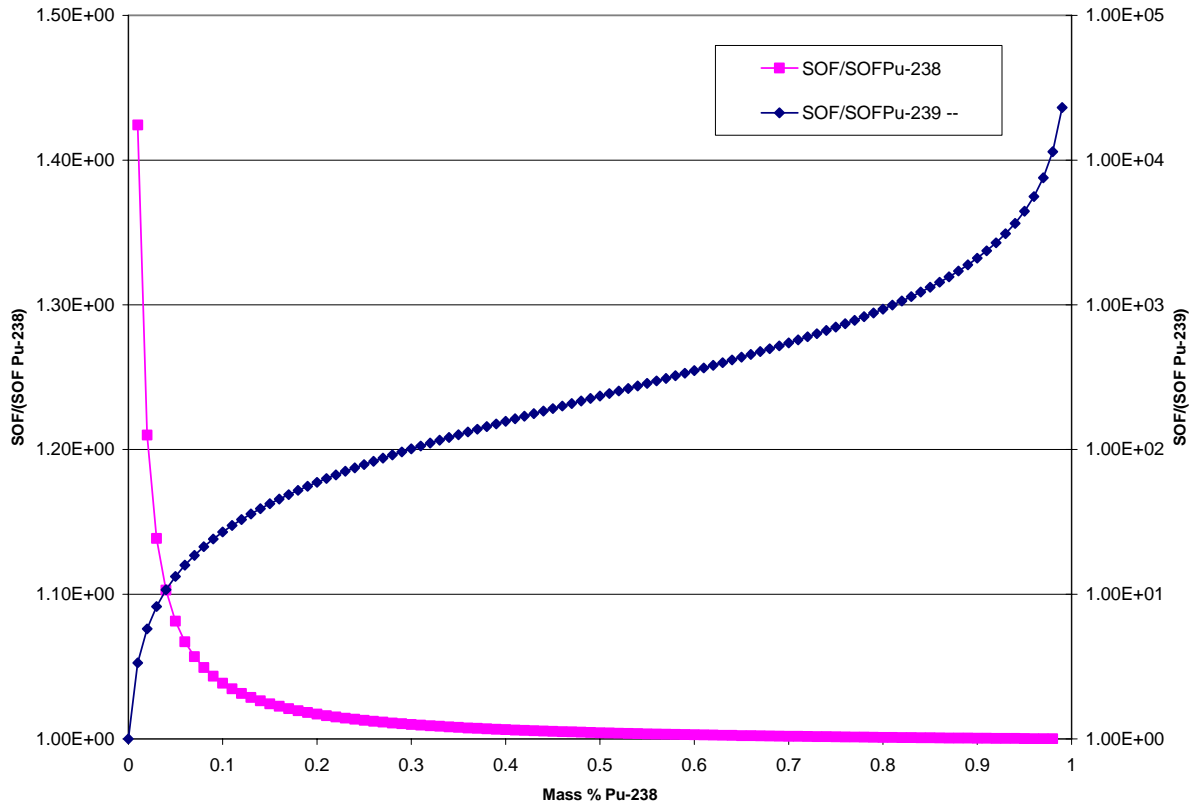


Figure 2. Effect of contaminants on  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  specimens

The example of Figure 2 only looks at the case where only a single contaminant radioisotope affects another radioisotope. As the following will show, there are examples where multiple contaminants can be present in a specimen. Then impact these contaminants can have on a hazard categorization assessment will depend on the relative magnitude of the contaminants TQs and the amount present.

In the following discussion, examples are given where contaminants have been noted to be present in source material. Keep in mind, these are examples that can be used as starting point to define the source term. Holders of source material without complete assay results are encouraged to research the source material to confirm the radioisotopic content.

NOTE: Through out the discussion in this paper, reference is made to “source material”. In this context, source material concerns the radioactive material typically used in the fabrication of sealed sources or as radioactive material of a specific composition. The use of the term “source material” is not meant to imply the designation of material as defined by the NRC in 10 CFR 40 (i.e., natural thorium and natural/depleted uranium).

### Examples of Contaminants In Transuranium Source Materials

#### Uranium

Uranium, a ubiquitous component of Department of Energy (DOE) operations, is typically described in terms of the  $^{235}\text{U}$  and  $^{238}\text{U}$  content. However, almost all specimens that contain  $^{235}\text{U}$  and  $^{238}\text{U}$ , also contain  $^{234}\text{U}$ .

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The presence of  $^{234}\text{U}$  is interesting from the perspective that it is actually a progeny of the  $^{238}\text{U}$  decay series. However, from the perspective of uranium material, it is commonly considered a contaminant because its in-growth from the decay of  $^{238}\text{U}$  is very slow and it cannot be chemically separated from the other uranium isotopes.

The  $^{234}\text{U}$  content in uranium material depends on the type of uranium material (natural, enriched, or depleted) and the source of the uranium material (from uranium ore, from nuclear fuel recycling/recovery processes). Table 1 displays the results associated with expected uranium isotope contamination levels within various grades of unirradiated uranium material. Except for the depleted uranium case, the  $^{234}\text{U}$  radioisotope is dominating the SOF calculation. To make matters worse, as the  $^{235}\text{U}$  enrichment increases, a proportional increase in the  $^{234}\text{U}$  abundance occurs, which exacerbates the effect  $^{234}\text{U}$  has on the overall SOF calculation, effectively overwhelming the SOF contribution of the other uranium isotopes.

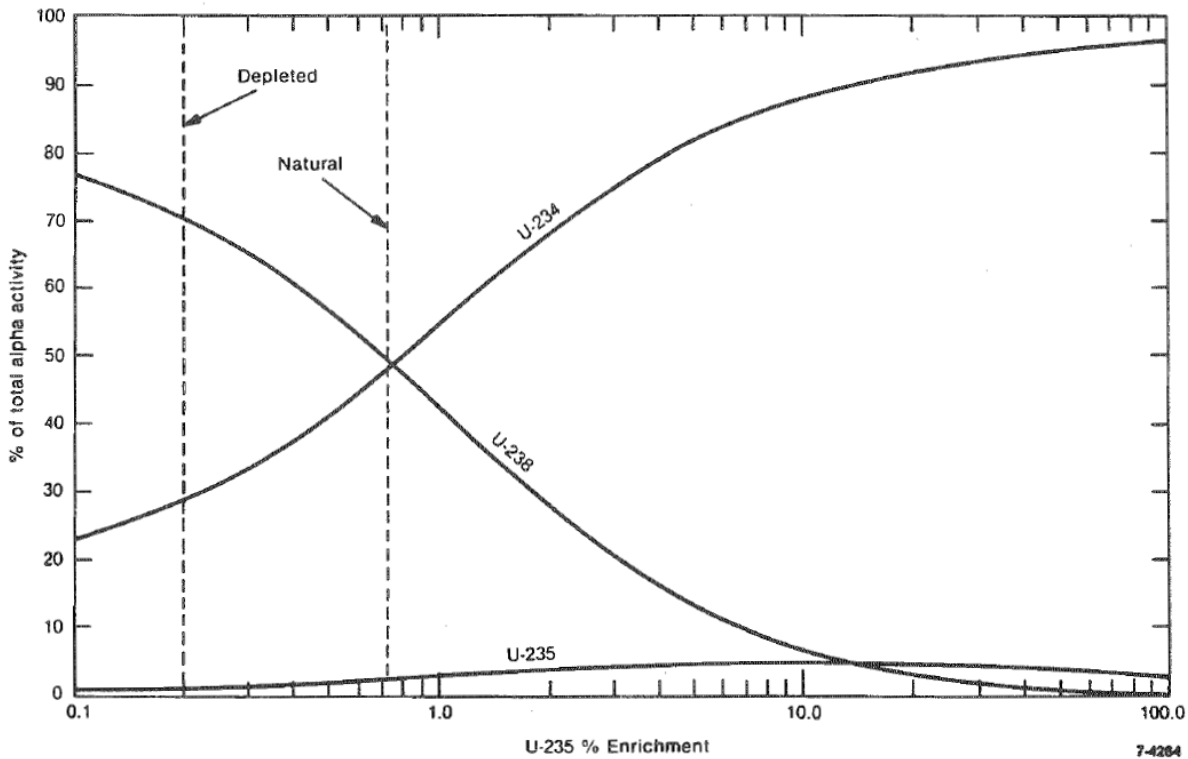
**Table 1. Contaminant Assessment of Unirradiated Natural, Depleted and Enriched Uranium**

Description	Isotope	Activity Abundance <sup>2</sup>	HC-3 TQ (Ci)	SOF / Ci – U
Natural – Uranium <sup>a</sup>  (0.72% mass $^{235}\text{U}$ )	$^{234}\text{U}$	48.83%	4.2	1.16E-01
	$^{235}\text{U}$	2.34%	4.2	5.57E-03
	$^{238}\text{U}$	48.83%	4.2	1.16E-01
	<b>f<sub>SOF</sub></b>			<b>2.05</b>
Depleted – Uranium <sup>b</sup>  (0.25% mass $^{235}\text{U}$ )	$^{234}\text{U}$	8.4%	4.2	2.00E-02
	$^{235}\text{U}$	1.5%	4.2	3.57E-03
	$^{238}\text{U}$	90.1%	4.2	2.15E-01
	<b>f<sub>SOF</sub></b>			<b>1.11</b>
Enriched (5 wt%) – UF <sub>6</sub>  (10x10 <sup>3</sup> μg $^{234}\text{U}$ /g $^{235}\text{U}$ ) <sup>3</sup>	$^{234}\text{U}$	87.98%	4.2	2.09E-01
	$^{235}\text{U}$	3.04%	4.2	7.24E-03
	$^{238}\text{U}$	8.98%	4.2	2.14E-02
	<b>f<sub>SOF</sub></b>			<b>11.1</b>
Enriched (2 wt%) – UF <sub>6</sub>  (11x10 <sup>3</sup> μg $^{234}\text{U}$ /g $^{235}\text{U}$ ) <sup>4</sup>	<b>Mass</b>			
	<b>Isotope</b>	<b>Abundance</b>	<b>HC-3 TQ (Ci)</b>	<b>SOF / Ci – U</b>
	$^{232}\text{U}$	1.00E-10	0.82	1.48E-03
	$^{234}\text{U}$	0.02%	4.2	1.85E-01
	$^{235}\text{U}$	2.00%	4.2	5.83E-03
	$^{236}\text{U}$	2.50E-04	4.2	2.18E-03
$^{238}\text{U}$	97.95%	4.2	4.44E-02	
<b>f<sub>SOF</sub></b>			<b>5.36</b>	

Based on the observations of Table 1, the contribution of  $^{234}\text{U}$  to HC-3 SOF assessment should never be overlooked. Unfortunately, in many cases the actual  $^{234}\text{U}$  content is not specified. When such occasions occur, Figure 3 can be used to estimate the  $^{234}\text{U}$  content based on the known  $^{235}\text{U}$  enrichment (via gaseous diffusion processes).

<sup>a</sup> Data from Table 3.1.1 of NUREG-1717<sup>2</sup>

<sup>b</sup> Ibid



**Figure 3. Percent of total radioactivity by isotope vs, U-235 enrichment from gaseous diffusion<sup>5</sup>**

Uranium recovered from irradiated fuel can have a significant <sup>236</sup>U contamination. Similar to the behavior observed for <sup>234</sup>U, subsequent enrichment processes of <sup>235</sup>U results in a relative increase in the <sup>236</sup>U abundance. Table 2 displays the results associated with expected uranium isotope contamination levels within various grades of irradiated uranium material containing <sup>236</sup>U.

**Table 2. Contaminant Assessment of Chemically Separated Enriched Irradiated Uranium**

Description	Isotope	Activity Abundance	HC-3 TQ (Ci)	SOF / Ci – U	% of Total HC-3 SOF
Enriched Irradiated (5 wt%) – Uranium <sup>6</sup>	<sup>234</sup> U	77.96%	4.2	1.86E-01	78.0%
	<sup>235</sup> U	1.39%	4.2	3.31E-03	1.4%
	<sup>236</sup> U	16.64%	4.2	3.96E-02	16.6%
	<sup>238</sup> U	4.01%	4.2	9.56E-03	4.0%
			<b>f<sub>SOF</sub></b>	<b>24.9</b>	

Similar to the unirradiated case, the irradiated uranium SOF assessment is dominated by the contributed associated with <sup>234</sup>U. However, the <sup>236</sup>U radioisotope comprises 16.6% of the HC-3 SOF calculation. Obviously, consideration must be given to the presence of both <sup>234</sup>U and <sup>236</sup>U in chemically separated enriched irradiated uranium.

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Plutonium

Plutonium radioisotopes do not occur naturally. Plutonium isotopes are produced through man-made processes such as neutron irradiation of uranium materials or the milking of higher atomic number transuranium elements, such as curium. In neutron irradiation production processes, a whole suite of plutonium radioisotopes can be produced, including;  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$ , and  $^{244}\text{Pu}$ .

Table 3 illustrates the effect that contaminants in normal  $^{238}\text{Pu}$  source material have on the HC-3 SOF calculation. The contribution from the contaminants is practically negligible, due to the high activity abundance of  $^{238}\text{Pu}$  and the low HC-3 TQ of  $^{238}\text{Pu}$  relative to the other contaminants present.

**Table 3. Contaminant assessment of  $^{238}\text{Pu}$  source material**

Description	Isotope	Atomic Abundance <sup>7</sup>	Activity Abundance	HC-3 TQ (Ci)	SOF / mCi – Pu	% of Total HC-3 SOF
Normal $^{238}\text{Pu}$	$^{236}\text{Pu}$	1.20E-06	0.42%	2.00E+00	2.12E-06	0.1%
	$^{238}\text{Pu}$	80%	91.77%	6.20E-01	1.48E-03	99.6%
	$^{239}\text{Pu}$	16%	0.07%	5.20E-01	1.27E-06	0.1%
	$^{240}\text{Pu}$	3%	0.04%	5.20E-01	8.01E-07	0.1%
	$^{241}\text{Pu}$	1%	7.70%	3.20E+01	2.41E-06	0.2%
	$^{242}\text{Pu}$	0%	0.00%	6.20E-01	1.73E-09	0.0%
<b>f<sub>SOF</sub></b>					<b>1.00</b>	

Other examples of  $^{238}\text{Pu}$  source material include Radioactive Thermal Generators (RTGs). The source used for the Cassini RTG fuel was predominantly  $^{238}\text{Pu}$ , with an activity abundance of 98.8%<sup>8</sup>. Sources with such high  $^{238}\text{Pu}$  isotopic abundance do not have any contaminants to significantly impact HC-3 SOF calculations.

In the next example, we'll see that just because the predominant isotope has the lowest TQ in comparison to the contaminants, that does not ensure the effect of the contaminants will be minimal. In the late 1950s and into the early 1960s, the Monsanto Research Corporation (MRC), managing contractor of the Mound Laboratory, published a paper detailing the increase in the neutron yield of  $^{239}\text{Pu}/\text{Be}$  sources<sup>9</sup>. Data presented in the paper noted significant levels of  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  contamination. As shown in Table 4, the presence of the contaminants  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  in trace quantities results in a significant contribution to the HC-3 SOF.

**Table 4. Contaminant assessment of  $^{239}\text{Pu}/\text{Be}$  sources from Mound Laboratory**

Description	Isotope	Atomic Abundance <sup>9</sup>	Activity Abundance	HC-3 TQ (Ci)	SOF / mCi – Pu	% of Total HC-3 SOF
Monsanto $^{239}\text{Pu}/\text{Be}$ Neutron Source	$^{238}\text{Pu}$	0.01%	0.21%	6.20E-01	3.42E-06	1.6%
	$^{239}\text{Pu}$	91.6%	7.09%	5.20E-01	1.36E-04	64.9%
	$^{240}\text{Pu}$	7.7%	2.19%	5.20E-01	4.22E-05	20.1%
	$^{241}\text{Pu}$	0.7%	90.50%	3.20E+01	2.83E-05	13.4%
<b>f<sub>SOF</sub></b>					<b>1.54</b>	

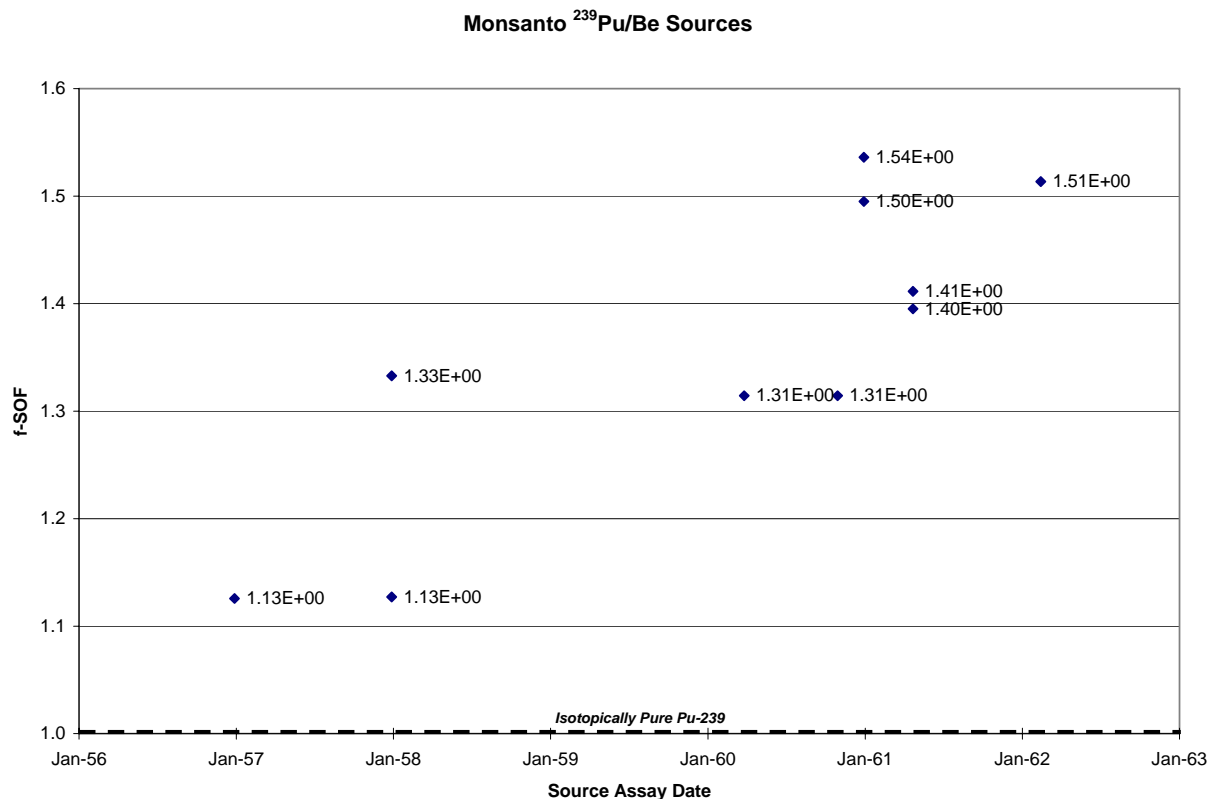
The interesting aspect is that these are referred to as  $^{239}\text{Pu}$  sources instead of  $^{241}\text{Pu}$  sources. Even though  $^{239}\text{Pu}$  is the predominant radioisotope on a mass basis, the  $^{241}\text{Pu}$  radioisotope is the predominant on an activity basis. The justification for referring to these sources as  $^{239}\text{Pu}$  source material is attributed to the fact that  $^{241}\text{Pu}$  is a beta emitter (decays to  $^{241}\text{Am}$ ). Therefore, initially,  $^{239}\text{Pu}$  is the predominant alpha

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radioisotope. And it is the alpha interaction with the beryllium that results in the neutrons emitted from the source.

Additional review of the data from paper notes that the degree of contamination of  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  varied over time. Initially the  $f_{\text{SOF}}$  value for the  $^{239}\text{Pu}/\text{Be}$  sources were only 1.13 in January 1957. But as the years progressed, the degree of  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  increased significantly, finally peaking at a  $f_{\text{SOF}}$  value 1.54 four years later in 1961. These results are illustrated in Figure 4. Although the paper did not postulate why the contamination increased over time, it reveals the fact that contamination levels are not necessarily static. The degree of contamination should be expected to vary due to factors that affect the final production such as: composition of the material prior to irradiation, duration of neutron irradiation and neutron irradiation flux history.



**Figure 4.  $f_{\text{SOF}}$  Values for Monsanto  $^{239}\text{Pu}/\text{Be}$  Neutron Sources<sup>9</sup>**

Source material described as  $^{240}\text{Pu}$  has a typical radiopurity (a.k.a. activity abundance) of 75% to 95%<sup>10</sup>. For the lower radiopurity material, the isotopic content of the radioisotopes  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  should be verified since these radioisotopes have equal or lower HC-3 TQ activity values.

The DOE Office of Nuclear Science Isotopes Program offers source material described as  $^{241}\text{Pu}$  with a radiopurity 80% to 93%<sup>10</sup>. The source material described as originating from the neutron irradiation of  $^{238}\text{U}/^{238}\text{Pu}$  and subsequently undergoing chemical and electromagnetic separation processing. Since the HC-3 TQ of  $^{241}\text{Pu}$  is much larger (32 Ci) than the HC-3 TQs for  $^{238}\text{Pu}$  (0.62 Ci),  $^{239}\text{Pu}$  (0.52 Ci),  $^{240}\text{Pu}$  (0.52 Ci) and  $^{242}\text{Pu}$  (0.62 Ci), then these contaminants can have a significant impact on the HC-3 SOF assessment. (Also, the decay of  $^{241}\text{Pu}$  into  $^{241}\text{Am}$  impacts the SOF calculation as noted later the discussion on the effects of progeny on SOF assessments). Table 5 presents the hypothetical situation where the 80% radiopurity  $^{241}\text{Pu}$  source material has  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  as the primary contaminants. The

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presence of these contaminants would account for almost 94% of the total HC-3 SOF, which would be 16.4 times higher than the SOF contribution of just the  $^{241}\text{Pu}$  radioisotope.

**Table 5. Assessment of  $^{241}\text{Pu}$  source material with  $^{239}\text{Pu}$  and/or  $^{240}\text{Pu}$  as the contaminants**

Description	Isotope	Activity Abundance	HC-3 TQ (Ci)	SOF / mCi – Pu	% of Total HC-3 SOF
$^{241}\text{Pu}$ w/ $^{239}\text{Pu}$ & $^{240}\text{Pu}$	$^{239}\text{Pu} + ^{240}\text{Pu}$	20.0%	5.20E-01	3.85E-04	93.9%
	$^{241}\text{Pu}$	80.0%	3.20E+01	2.50E-05	6.1%
<b><math>f_{\text{SOF}}</math></b>				<b>16.4</b>	

If the contaminants were of a higher HC-3 TQ (i.e.  $^{238}\text{Pu}$  or  $^{242}\text{Pu}$ ), then the effect would still result in the contaminants dominating the HC-3 SOF assessment.

**Table 6. Assessment of  $^{241}\text{Pu}$  source material with  $^{238}\text{Pu}$  and/or  $^{242}\text{Pu}$  as the contaminants**

Description	Isotope	Activity Abundance	HC-3 TQ (Ci)	SOF / mCi – Pu	% of Total HC-3 SOF
$^{241}\text{Pu}$ w/ $^{238}\text{Pu}$ & $^{242}\text{Pu}$	$^{238}\text{Pu} + ^{242}\text{Pu}$	20.0%	6.20E-01	3.23E-04	92.8%
	$^{241}\text{Pu}$	80.0%	3.20E+01	2.50E-05	7.2%
<b><math>f_{\text{SOF}}</math></b>				<b>13.9</b>	

If the isotopic content of  $^{241}\text{Pu}$  source material is not defined, then a bounding HC-3 SOF assessment can be made by assuming the composition in Table 5.

The last plutonium isotope to be discussed is  $^{242}\text{Pu}$ . The typical radiopurity of  $^{242}\text{Pu}$  source material from the DOE Office of Nuclear Science Isotopes Program is > 99%<sup>10</sup>. Given the relatively low HC-3 TQ for  $^{242}\text{Pu}$ , the potential plutonium contaminants present would not significantly affect the HC-3 SOF assessment of this source material.

In 1995, a paper published by the Los Alamos National Laboratory (LANL) provided an excellent treatment of the effect plutonium mixtures have on HC-2 assessments<sup>11</sup>. The paper provided examples of various plutonium source materials and demonstrated how the HC-2 TQ and relative hazard of the source materials changed over time due to radioactive decay. In the LANL paper, the  $f_{\text{SOF}}$  values were calculated in terms of the HC-2 TQs for the various mixtures.

The data presented in Table 7 takes the information from the LANL plutonium mixture assessment and determines the impact on HC-3 SOF assessments. In general, the HC-3 SOF increases as the  $^{238}\text{Pu}$  content increases. A better correlation is noted between the total mass percentage of ( $^{238}\text{Pu} + ^{241}\text{Pu}$ ). As the ( $^{238}\text{Pu} + ^{241}\text{Pu}$ ) content increases, so does the overall HC-3 SOF. This is attributed to  $^{238}\text{Pu} + ^{241}\text{Pu}$  have the lowest HC-3 TQs in relation to the other plutonium radioisotopes.

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**Table 7. HC-3 SOF Assessment of various plutonium mixtures<sup>11</sup>**

Material	Wt % Pu Radioisotope						<sup>238</sup> Pu + <sup>241</sup> Pu	HC-3 SOF/g-Pu
	<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu	<sup>244</sup> Pu		
MT 51	0.01%	<b>96.77%</b>	3.13%	0.08%	0.02%	0.00%	0.08%	1.33E-01
MT 52	0.01%	<b>93.78%</b>	6.00%	0.20%	0.02%	0.00%	0.21%	1.47E-01
MT 95%	0.45%	0.56%	2.47%	0.91%	<b>95.58%</b>	0.03%	1.36%	1.72E-01
MT 54	0.05%	<b>87.42%</b>	11.50%	0.81%	0.22%	0.00%	0.86%	1.93E-01
MT 55	0.06%	<b>82.88%</b>	14.73%	1.03%	0.30%	0.00%	1.09%	2.13E-01
MT 56	0.06%	<b>81.90%</b>	16.51%	1.18%	0.36%	0.00%	1.24%	2.25E-01
MT 53	0.03%	<b>91.08%</b>	8.45%	3.66%	0.07%	0.00%	3.69%	2.72E-01
MT 42 90%	0.72%	1.26%	6.40%	1.86%	<b>89.77%</b>	0.00%	2.58%	2.95E-01
MT 57	0.43%	<b>74.63%</b>	20.70%	2.55%	1.69%	0.00%	2.98%	3.82E-01
MT 42 84%	1.02%	1.37%	10.32%	3.13%	<b>84.14%</b>	0.02%	4.15%	4.37E-01
MT 83 83%	<b>83.89%</b>	13.80%	1.90%	0.32%	0.09%	0.00%	84.21%	2.33E+01
MT 83 89%	<b>89.26%</b>	10.07%	0.63%	0.02%	0.02%	0.00%	89.28%	2.48E+01

Americium

Americium is available both commercially and through internal DOE sources, such as the DOE Office of Nuclear Science Isotopes Program. The level of contamination associated with the americium material will be dependent on the production techniques employed and the degree of purity required.

Commercially, <sup>241</sup>Am is available with an isotopic purity of 95%<sup>13</sup>. The typical contaminants found in <sup>241</sup>Am source material is <sup>242m</sup>Am and <sup>243</sup>Am. The typical <sup>242m</sup>Am content is 0.4 wt% of the americium content from light water reactors<sup>12</sup>. Table 8 summarizes the results associated with <sup>241</sup>Am source material that originates from light water reactors.

**Table 8. Contaminant assessment of <sup>241</sup>Am sources**

Description	Isotope	Mass Abundance	Activity Abundance	HC-3 TQ (Ci)	SOF / mCi - Am	% of Total HC-3 SOF
Commercially Available	<sup>241</sup> Am	95.0% <sup>13</sup>	98.547%	5.20E-01	1.90E-03	98.5%
	<sup>242m</sup> Am	0.4% <sup>12</sup>	1.176%	5.20E-01	2.26E-05	1.2%
	<sup>243</sup> Am	4.6%	0.277%	5.20E-01	5.32E-06	0.3%
<b>f<sub>SOF</sub></b>					<b>1.01</b>	

The presence of contaminants <sup>242m</sup>Am and <sup>243</sup>Am only result in a 1% increase in the overall HC-3 SOF assessment of the material. Source <sup>241</sup>Am material from the DOE Office of Nuclear Science Isotopes Program has a rated radiopurity of > 99 %. This radiopurity level is higher than the commercially available <sup>241</sup>Am material assessed in Table 8. As such, the expected level of contamination in <sup>241</sup>Am sources do not adversely affect HC-3 SOF assessments.

A word of caution regarding <sup>241</sup>Am from other production sources. The <sup>242m</sup>Am content in americium material can be much higher than 0.4 wt% when produced in a fast flux reactor (JOP 2004). Americium material that originates from fast flux reactors should be researched for <sup>242m</sup>Am isotopic content to ensure proper calculation of the HC-3 SOF.

The DOE Office of Nuclear Science Isotopes Program offers <sup>243</sup>Am sources with a radiopurity of > 99.9%. The method of production for these sources is through neutron irradiation of <sup>242</sup>Pu. This results

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in very low levels of  $^{241}\text{Am}$  and  $^{242\text{m}}\text{Am}$  contamination. With such a high radiopurity, the potential contaminants do not significantly affect the HC-3 SOF assessment of  $^{243}\text{Am}$  source material.

### Curium

The DOE Office of Nuclear Science Isotopes Program offers two curium radioisotopes;  $^{244}\text{Cm}$  and  $^{248}\text{Cm}$ . Although the radiopurity of  $^{244}\text{Cm}$  is not specifically noted, other indicate that a mass abundance of 91.77%  $^{244}\text{Cm}$  can be achieved in curium reclaimed from reactor wastes<sup>14</sup>. Table 9 summarizes the results associated with  $^{244}\text{Cm}$  source material that is recovered from reactor wastes.

**Table 9. Contaminant assessment of  $^{244}\text{Cm}$  sources**

Description	Isotope	Mass Abundance <sup>14</sup>	Activity Abundance	HC-3 TQ (Ci)	SOF / mCi – Cm	% of Total HC-3 SOF
$^{244}\text{Cm}$ source material derived from reactor wastes	$^{243}\text{Cm}$	0.25%	0.173%	8.20E-01	2.11E-06	0.2%
	$^{244}\text{Cm}$	91.77%	99.807%	1.04E+00	9.60E-04	99.7%
	$^{245}\text{Cm}$	7.2%	0.017%	5.20E-01	3.18E-07	0.0%
	$^{246}\text{Cm}$	0.8%	0.003%	5.20E-01	6.51E-08	0.0%
<b>f<sub>SOF</sub></b>					<b>1.00</b>	

For  $^{248}\text{Cm}$  source material, the DOE Office of Nuclear Science Isotopes Program notes the specification radiopurity is 97% when derived from the decay of  $^{252}\text{Cf}$  source material. As will be noted in the californium section,  $^{252}\text{Cf}$  source material is not isotopically pure. A curium purification study published in 2001, reported on the purification of  $^{248}\text{Cm}$  from a 13- $\frac{1}{2}$  year old  $^{252}\text{Cf}$  source. Table 9 summarizes the results associated with the  $^{248}\text{Cm}$  source material that recovered. The activity abundance of the recovered  $^{248}\text{Cm}$  was only 50.7%. Given that the DOE Isotopes Program can provide  $^{248}\text{Cm}$  source material with a radiopurity of 97%, then it would be conservatively bounded by the analysis presented in Table 10.

**Table 9. Contaminant assessment of  $^{248}\text{Cm}$  sources**

Description	Isotope	Mass Abundance <sup>15</sup>	Activity Abundance	HC-3 TQ (Ci)	SOF / mCi – Cm	% of Total HC-3 SOF
$^{248}\text{Cm}$ source material derived from $^{252}\text{Cf}$ source material	$^{245}\text{Cm}$	0.11%	0.824%	5.20E-01	1.58E-05	0.5%
	$^{246}\text{Cm}$	6.24%	82.113%	5.20E-01	1.58E-03	48.8%
	$^{247}\text{Cm}$	0.04%	0.000%	6.20E-01	2.53E-09	0.0%
	$^{248}\text{Cm}$	93.61%	17.063%	1.04E-01	1.64E-03	50.7%
<b>f<sub>SOF</sub></b>					<b>1.97</b>	

### Californium

The DOE Office of Nuclear Science Isotopes Program offers two californium radioisotopes;  $^{249}\text{Cf}$  and  $^{252}\text{Cf}$ . These radioisotopes are produced through the sequential neutron captures in transuranium isotopes and purified.

The radiopurity of  $^{249}\text{Cf}$  source material is specified to be > 98%. Since no other californium radioisotope has HC-3 TQ lower than  $^{249}\text{Cf}$  (0.52 Ci), then the potential impacts of contaminants are not expected to significantly affect the HC-3 SOF assessment of this material.

The isotopic abundance of  $^{252}\text{Cf}$  source material is specified to be 80% to 85%. Table 11 summarizes the results associated with the  $^{252}\text{Cf}$  source capsule produced at a DOE facility in 1982.

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**Table 11. Contaminant assessment of <sup>252</sup>Cf sources**

Description	Isotope	Mass Abundance <sup>15</sup>	Activity Abundance	HC-3 TQ (Ci)	SOF / mCi - Cm	% of Total HC-3 SOF
<sup>252</sup> Cf source material	<sup>249</sup> Cf	3.76%	0.034%	5.20E-01	6.52E-07	0.2%
	<sup>250</sup> Cf	10.44%	2.499%	1.04E+00	2.40E-05	7.3%
	<sup>251</sup> Cf	3.29%	0.012%	5.20E-01	2.21E-07	0.1%
	<sup>252</sup> Cf	82.50%	97.456%	3.20E+00	3.05E-04	92.4%
<b>f<sub>SOF</sub></b>					<b>1.08</b>	

Neptunium

The only neptunium source material typically encountered in <sup>237</sup>Np sources. The only potential neptunium contaminants would be <sup>235</sup>Np and <sup>236</sup>Np since all other neptunium isotopes have half-lives that are too short (< 5 days) to be considered present.

The radiochemical purity of <sup>237</sup>Np sources is specified to be >97% from the ORNL Transuranium Element Program (TRU)<sup>16</sup>. The only significant contaminant is the progeny <sup>233</sup>Pa. The impact of <sup>233</sup>Pa is addressed in the progeny section of this discussion.

Thorium

Natural thorium is predominantly comprised of <sup>232</sup>Th. The primary contaminant is <sup>228</sup>Th, which is a progeny of <sup>232</sup>Th. The impact of <sup>228</sup>Th is addressed in the progeny section of this discussion.

**Examples of Contaminants In Other Source Materials**

Cesium

Cesium source material is predominantly derived from fission product sources. The source material is characterized as <sup>137</sup>Cs source material and is available through commercial vendors and internally from DOE resources. Typical contaminants associated with <sup>137</sup>Cs source material are <sup>134</sup>Cs and <sup>135</sup>Cs.

The fission yield of cesium material dependent on several factors including; (i) source material associated with the fission process (e.g., <sup>235</sup>U, Pu radioisotopes), (ii) the neutron irradiation energy levels (e.g. thermal), (iii) neutron flux, (iv) duration of fission process (for total cesium yield).

Table 12 reports the fission yield of <sup>134</sup>Cs, <sup>135</sup>Cs and <sup>137</sup>Cs from the thermal neutron induced fission of <sup>235</sup>U source material (typical of cesium source material from pressurized light water reactors).

**Table 12. Yield of Cesium Source Material From <sup>235</sup>U Thermal Fission<sup>17</sup>**

Isotope	Half-life (yr) <sup>18</sup>	Fission Yield Data		
		Meek & Rider (M&R)	ENDF/B-VI (B-6)	JEF-2 (J-2)
<sup>133</sup> Cs	Stable	6.69E-02	6.70E-02	6.62E-02
<sup>134</sup> Cs	2.06E+00	1.33E-07	7.71E-08	2.03E-07
<sup>135</sup> Cs	3E+06	6.54E-02	6.54E-02	6.58E-02
<sup>137</sup> Cs	3.02E+01	6.22E-02	6.19E-02	6.24E-02

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Using the Meek & Rider fission yield data<sup>17</sup>, Table 13 summarizes the impact the contaminants have cesium source material. The presence of the contaminants do not affect the HC-3 SOF assessment of the <sup>137</sup>Cs source material.

**Table 13. Contaminant assessment of <sup>137</sup>Cs sources**

Description	Isotope	Isotopic Abundance	Activity Abundance	HC-3 TQ (Ci)	SOF / Ci – Cs	% of Total HC-3 SOF
<sup>137</sup> Cs source material	<sup>133</sup> Cs	34.40%	--	--	--	--
	<sup>134</sup> Cs	0.00%	0.003%	4.20E+01	7.38E-07	0.0%
	<sup>135</sup> Cs	33.62%	0.001%	4.20E+02	3.26E-08	0.0%
	<sup>137</sup> Cs	31.98%	99.996%	6.00E+01	1.67E-02	100.0%
<b>f<sub>SOF</sub></b>					<b>1.00</b>	

### Europium

Similar to cesium source material, europium source material is can be derived from fission product sources. While <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>155</sup>Eu are produced primarily as fission products, <sup>152</sup>Eu can also be produced by neutron activation of nuclear reactor control rods. The fission yield of <sup>155</sup>Eu is about 0.03% while the yield of the other two isotopes is much lower<sup>19</sup>. Commercially available <sup>155</sup>Eu source material has a radiopurity specification of 99%<sup>20</sup>. Since <sup>155</sup>Eu decays into <sup>155</sup>Gd, a stable isotope, then the remaining 1% radiopurity is attributed to contaminants. The HC-3 TQ for <sup>155</sup>Eu (980 Ci) is larger than the HC-3 TQs for <sup>152</sup>Eu (200 Ci) and <sup>154</sup>Eu (200 Ci). Assuming the contaminants are <sup>152</sup>Eu and/or <sup>154</sup>Eu (i.e., the europium source material is assume to originate from fission product material), Table 14 shows that the presence of these contaminants only has a moderate impact on the HC-3 SOF assessment of the source material

**Table 14. Contaminant assessment of commercially available <sup>155</sup>Eu source material**

Description	Isotope	Activity Abundance <sup>20</sup>	HC-3 TQ (Ci)	SOF / Ci – Eu	% of Total HC-3 SOF
<sup>155</sup> Eu source material	<sup>152</sup> Eu + <sup>154</sup> Eu	1%	200	5.00E-05	4.5%
	<sup>155</sup> Eu	99%	980	1.05E-03	95.5%
<b>f<sub>SOF</sub></b>				<b>1.05</b>	

Specialty sources categorized as <sup>152</sup>Eu and <sup>154</sup>Eu tend to originate from other processes (i.e. neutron irradiation of <sup>151</sup>Eu) or are subjected to addition purification to remove other europium isotopes (mass separator operations), achieving radiopurities in excess of 99%. Contaminants in these source materials are too low to significantly affect an HC-3 SOF assessment of these europium source materials<sup>21</sup>.

### Cobalt

Cobalt source material can be produced by the neutron activation of metals such as iron or nickel or through particle accelerator processes. Only <sup>57</sup>Co (t<sub>1/2</sub> = 270 days) and <sup>60</sup>Co (t<sub>1/2</sub> = 5.3 years) have half-lives long enough to warrant concern since the remaining cobalt radioisotopes have half-lives that are less than 80 days.

Source material designated as <sup>57</sup>Co can be produced in a cyclotron, via the proton irradiation of natural nickel. The irradiation process primarily produces <sup>57</sup>Ni and <sup>57</sup>Co along with other nickel and cobalt trace

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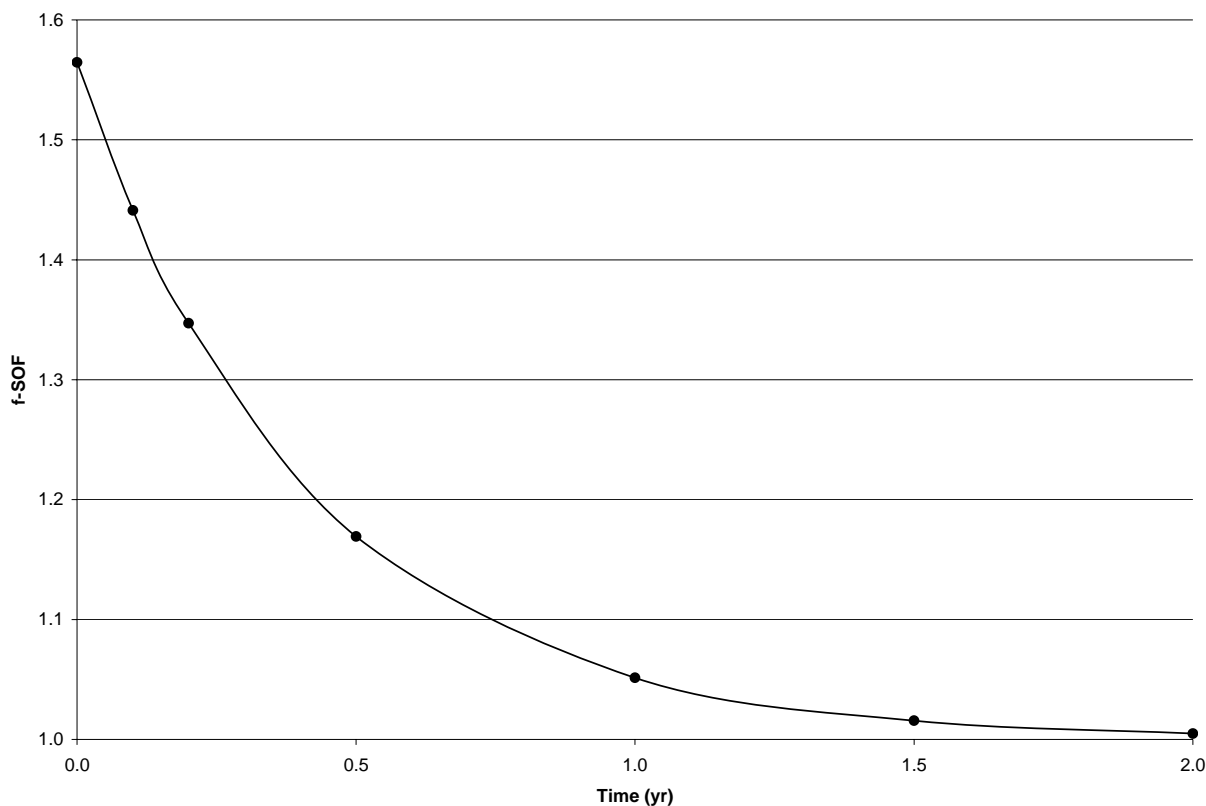
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isotopes<sup>22</sup>. After chemical separation from the nickel, the <sup>57</sup>Co source material has trace amounts of <sup>55</sup>Co, <sup>56</sup>Co, and <sup>58</sup>Co. Table 15 summarizes the impact the contaminants have <sup>57</sup>Co source material.

**Table 15. Contaminant assessment of cyclotron produced <sup>57</sup>Co source material**

Description	Isotope	Activity Abundance <sup>22</sup>	HC-3 TQ (Ci)	SOF / Ci – Co	% of Total HC-3 SOF
<sup>57</sup> Co source material	<sup>55</sup> Co	0.1%	9.80E+02	6.14E-07	0.2%
	<sup>56</sup> Co	1.3%	2.20E+02	5.99E-05	23.9%
	<sup>57</sup> Co	95.9%	6.00E+03	1.60E-04	63.9%
	<sup>58</sup> Co	2.7%	9.00E+02	2.98E-05	11.9%
				<b>f<sub>SOF</sub></b>	<b>1.56</b>

The interesting aspect of <sup>57</sup>Co source material is that the contaminants that have a significant impact on the HC-3 SOF of the source material are relatively short-live radioisotopes. As illustrated in Figure 5, after two years of decay, the contaminants no longer significantly affect the HC-3 SOF assessment of the <sup>57</sup>Co source material.



**Figure 5. Time dependent f<sub>SOF</sub> behavior of <sup>57</sup>Co source material**

Source material designated as <sup>60</sup>Co is offered by the DOE Office of Nuclear Science Isotopes Program with a specification radiopurity of >99%. Since <sup>60</sup>Co (HC-3 TQ = 280 Ci) has a lower HC-3 TQ than <sup>57</sup>Co (HC-3 TQ = 6000 Ci), then the impact of <sup>57</sup>Co contamination in a <sup>60</sup>Co source would be negligible.

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### Progeny

The inter-relationship of progeny in the hazard categorization is a topic that is not explicitly discussed in DOE-STD-1027<sup>23</sup>. Since DOE-STD-1027 does not provide any discussion relative to the role of progeny in hazard categorization assessment, then references and other supporting documentation must be utilized in determining the role of progeny.

#### Basis for inclusion of progeny in hazard category 3 assessments

##### Hazard Category 3 Dose Limit

In 1992, DOE established Hazard Category 3 dose thresholds based on the value that is accepted by the EPA for the protection of workers for planned reentry into a facility after an incident, which is 10 rem<sup>23</sup>. Guidance later published by DOE also reaffirmed dose limit for reentry after an incident to be limited to 10 rem for the recovery of deceased victims, and for the protection of health and property<sup>24</sup>. The establishment of a dose limit implies that radionuclides that contribute to the dose personnel can receive upon reentry would be a candidate for the development of a threshold value, including progeny. However, that is not explicitly detailed in the discussion of the EPA and the DOE reentry limit. In order to determine the relevance of progeny, a review of the derivation of the HC-3 TQs is required.

##### Development of Hazard Category 3 Threshold Quantities

The selection of Hazard Category 3 TQs in DOE-STD-1027<sup>23</sup> and the companion LANL document<sup>1</sup> was based on the EPA model used in the derivation of the radioisotope Reportable Quantity (RQ) values<sup>25</sup>. The EPA defines an RQ to be an amount of radioactive material that if released would result in a dose of 0.5 rem to a person located 30 meters away. Therefore, to use the EPA model, the DOE HC-3 TQs were derived by multiplying the EPA RQs by 20, resulting in an activity based limit corresponding to 10 rem of dose exposure.

The model that the EPA used to develop the RQs assessed four different exposure pathways (inhalation, water ingestion, food ingestion and direct exposure). The pathway that required the least activity to yield the dose limit was designated as the limiting pathway and used to establish the final RQs published in 40 CFR 302.4 Appendix B<sup>26</sup>.

##### Role of Progeny In EPA Pathway Models

The EPA dose models for the inhalation, water ingestion and food ingestion exposure pathways utilized the Annual Limits of Ingestion (ALIs) published by the International Commission for Radiation Protection in ICRP-30<sup>27</sup>. The ALIs are defined by the ICRP to be represent “*the activity of a radionuclide which taken alone would irradiate a person.....to the limit set by the ICRP for each year of occupational exposure*”. The values for the ALIs were estimated for exposure, both by ingestion and inhalation, to specific radionuclides.

It is here in the derivation of the ALIs, which are used in the EPA model, where the ICRP notes that “*Any daughter radionuclide produced in the body after intake of the specific radionuclide are taken into account*”. The ICRP further clarified the role of progeny relevant to the committed dose equivalent:

*“...if the radionuclide has radioactive daughters, an allowance is made for the committed dose equivalent contributed by the build-up of daughters produced in the body from the parent. In general there is little evidence to indicate whether these daughters will remain associated with, and behave as, their parent, or whether, upon being produced, they will assume their own*

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*metabolic behavior. When experimental evidence is available, e.g. concerning the behaviour of noble gases radon and thoron when produced from their parents  $^{226}\text{Ra}$  and  $^{224}\text{Ra}$  in the body, it is given in the metabolic data. In all other cases it is assumed that the daughters and all subsequent progeny produced in the body (i.e. including the respiratory and gastrointestinal systems), stay with and behave metabolically like the inhaled or ingested parent radionuclide.*

Based on the ICRP statements, it can be concluded that progeny produced in the body is already accounted for in derivation of the ALI value. Accordingly, any progeny produced outside of the body would not be accounted for in the derivation of the ALIs. Progeny produced prior to ingestion/inhalation must accordingly be assessed as their own specific radionuclide parent.

The above discussion on the use of the ICRP ALIs focuses on three of the four EPA dose exposure pathways: inhalation, ingestion (water) and ingestion (food). Each one of these three pathways uses the ICRP ALI in the EPA model. The fourth pathway, direct exposure, does not use the ICRP ALI in the pathway dose model. This can be attributed to the fact that the ALIs were specifically noted by the ICRP to only apply to ingestion and inhalation pathways. For the direct exposure pathway, the EPA model for calculating dose is based on the dose associated with the spectrum of gamma rays that emanate from the decay of a specific radionuclide. The EPA direct exposure model based on the gamma dose received from the single isotope. Progeny are not included in the model. Therefore, if progeny are present that can contribute to the direct dose, it is implied that they would have to also be assessed for their dose impact.

### Examples of Assessment Including Progeny

The EPA lends credibility to this conclusion by noting in the Technical Background Document a methodology for determining the RQ of a mixture of radionuclides. Known as the mixture rule, it is a sum of RQ fractions assessment, similar in application to the hazard categorization sum of TQ fraction methodology. The principal behind the mixing rule is that assumption that risk of a biological effect is linearly related to dose equivalent<sup>27</sup>. Using the mixture rule, the EPA calculated the mixture RQ for four specific radioisotope mixtures: (1)  $^{226}\text{Ra}$  in secular equilibrium with its progeny, (2) natural uranium (i.e. uranium chemically separated from its progeny), (3) natural uranium in secular equilibrium with its progeny (i.e. natural uranium ore), and (4) natural thorium and its progeny in secular equilibrium. In three of the four RQ mixture examples, the progeny are assessed along with the parent radioisotope in determining the mixture RQ. The RQ for these four mixtures were subsequently codified into EPA regulations, in 40 CFR 302.4 Appendix B<sup>26</sup> and the mixture rule in general is codified in 40 CFR 306<sup>28</sup>.

### Summarizing the Findings – Progeny Present In Inventory Must Be Included

Given (i) the basis for the ICRP ALIs which only accounts for progeny produced within the body (not produced prior to ingestion), (ii) their use in the EPA dose exposure pathway models, (iii) the EPA direct exposure pathway dose model which is based on single radioisotope effects (progeny are not account for separately), (iv) the methodology associated with the codification of the EPA mixture rule and (v) the use of the EPA RQs (derived from the EPA pathway assessments) for deriving the DOE HC-3 TQs, results in the requirement that progeny present in the facility inventory must be assessed for impact on the overall facility hazard categorization.

### **Progeny That Significantly Affect Hazard Categorization**

As demonstrated above, progeny present in the facility radioactive material inventory must be included in hazard categorization sum of fraction assessments. On the surface, this is a straightforward statement which requires no clarification. However, in practice, defining the progeny component of the facility inventory present a significant challenge.

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### Determining Progeny Content

A straightforward way to determine radioisotope content, including progeny, involves the assaying of the specimen in question. The results from the assay examination can provide the radioisotope content and concentration/activity, which can be directly used in a sum of fractions assessment.

However in some cases, it's not possible or even practical to conduct an assay assessment. Accordingly, the ability to estimate the progeny content in a specimen relies on the ability to accurately profile the parent radionuclide(s) present, including:

- Original activity of the parent radioisotope(s).
- Date(s) associated with the determination of the parent(s) original activity.
- Serial decay chain information associated with the parent radionuclide(s) (e.g. specific progeny radionuclides, progeny half-life).

Armed with the above information, the Bateman equations can be used to estimate the in-growth of progeny in a specimen, where<sup>29</sup>:

$$N_n(t) = C_1 e^{-k_1 t} + C_2 e^{-k_2 t} + \dots + C_n e^{-k_n t}$$

with

$$C_1 = \frac{k_1 k_2 \dots k_{n-1}}{(k_2 - k_1)(k_3 - k_1) \dots (k_n - k_1)} N_{A_0},$$

$$C_2 = \frac{k_1 k_2 \dots k_{n-1}}{(k_1 - k_2)(k_3 - k_2) \dots (k_n - k_2)} N_{A_0},$$

$$C_n = \frac{k_1 k_2 \dots k_{n-1}}{(k_1 - k_n)(k_2 - k_n) \dots (k_{n-1} - k_n)} N_{A_0}$$

Where:

- $N_n(t)$  = Number of atoms of radioisotope "n" at time = t.
- $k_n$  = Decay constant for radioisotope "n" (where n = 1, 2, 3, 4, ..., n).
- $N_{A_0}$  = Number of atoms of the parent radioisotope initially present (@ t = 0).

The activity of isotope "n" is expressed as follows<sup>1</sup>:

$$A_n = \frac{N_n \cdot MW_n \cdot SA_n}{N_g}$$

where;

$$A_n = \text{Activity of isotope "n" (Ci)}$$

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$N_n$	= Total number of atoms of isotope “n”
$N_g$	= Avogadro's No. ( $6.02E+23$ atoms/mole)
$MW_n$	= Molecular weight of isotope “n” (g/mol)
$SA_n$	= Specific Activity of isotope “n” (Ci/g)

### Assessing Which Progeny Significantly Impact Sum of Fractions Assessments

The ability to estimate the progeny content using the Bateman Equations leads to a computationally intensive effort that can consume personnel and computational resources. The effect that progeny has on the overall sum of fractions is relative to not just the progeny content, but the HC-3 TQs of each progeny radioisotope. Progeny with HC-3 TQs higher than the parent isotope tend to not significantly impact the sum of fractions assessment, even when present in significant quantities. Conversely, progeny with HC-3 TQs the same as or lower than the parent isotope can significantly affect the sum of fractions assessment, even when present in trace amounts.

To complicate the issue, a decay chain containing multiple progeny can have both effects in play: (i) progeny with HC-3 TQs higher than the parent, (ii) progeny with HC-3 TQs the same as or lower than the parent isotope. Throw in the component time, which governs the calculated in-growth of progeny, and add in the fact that number of radioisotopes with radioactive progeny numbers in the hundred; then the management of the whole hazard categorization sum of fractions process quickly becomes a slow and tedious process without further clarification.

In order to provide a level of significance to the sum of fractions calculation process, a screening process was employed to limit the assessment of progeny as follows:

- A database, comprised of the 757 radioisotopes with assigned HC-3 TQs, was established. Radioisotopes with a half-life less than 0.25 yr were screened from further consideration. This screening was justified on the basis that these short-lived radioisotopes are not typically present below HC-3 facilities. Use of these isotopes in below HC-3 facilities is evaluated on a case-by-case basis to determine the effect of the resultant progeny.
- The radioisotopes with half-lives in excess of 0.25 yr that directly decayed into a stable isotope were also screened from the database. Since the progeny is a stable radioisotope, then it does not have an assigned HC-3 TQ. As such, the sum of fractions is solely based on the parent.

The results of this screening resulted in 79 radioisotopes that have half-lives longer than 0.25 yr and yield radioactive progeny. Isotopic decay information was assembled and a solution to the Bateman equations was calculated in successive time increments spanning at least 100 years. The calculations were performed using a Microsoft Excel® spreadsheet that contained the input information and calculated the time dependent activities of the parent and progeny<sup>c</sup>.

The calculated SOF of the mixture was compared to the SOF of the parent radionuclide for each time increment. A review of the results of each calculation notes that only 36 of the radioisotopes had  $f_{SOF}$  values that exceeded 1.10 within a 100 year period. Accordingly, the remaining 42 radioisotopes had  $f_{SOF}$  values that were less than 1.10 throughout the 100 year period. These 42 radioisotopes were screened out for further consideration. Justification for this screening is based on the operational limit the Oak Ridge National Laboratory imposes on its below HC-3 facilities (a 0.9 HC-3 SOF limit). This 10% margin

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<sup>c</sup> The Bateman equations can only be used to determine the activity of progeny in successive irreversible serial decay chains without branching. As such, the Bateman equations are only meant to be used in the special case where no branching occurs. The general solution to the serial decay problem that could involve branching is a complex calculation which requires resources beyond the use of spreadsheets. In general, when branching was encountered in defining the decay chain, the predominant branch was used since it yields the higher activity progeny.

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between the HC-3 threshold and the operating limit accounts provides a buffer to account for unanalyzed progeny and other uncertainties associated with the facility radioactive material inventory. The criterion of 100 years is based on the expectation that specimens within the various facilities are not older 100 years old. Radioactive materials at ORNL and other DOE facilities are based on processing and purification activities that should only date back as far as the Manhattan project, circa the mid 1940's. That involves a time frame of no more than 70-75 years for the oldest materials. Although it is possible to have radioactive materials older than 75 years old (pre-Manhattan Project), none of the materials have been identified in the facility inventories at ORNL.

The remaining 37 radioisotopes have progeny that results in  $f_{\text{SOF}}$  values exceeding 1.10 within the 100 year assessment period. These radioisotopes can be generalized in two classes: (1) Radioisotopes with progeny in secular equilibrium, and (2) Radioisotopes with progeny not in secular equilibrium (i.e. either transient equilibrium or no definable equilibrium).

Table 16 displays the radioisotopes that have progeny that are in secular equilibrium. These progeny build-in to an equilibrium level, resulting in the  $f_{\text{SOF}}$  that asymptotically approaches a value in excess of 1.1. The progeny that contributor significantly to the SOF assessment are displayed in bold (i.e., the progeny contributes more than 1% of the overall SOF). These radioisotopes can be bounded by assuming the maximum  $f_{\text{SOF}}$  condition exists, which means the activity of the progeny are the same as the activity of the parent. In these cases, only the activity of the parent needs to be calculated over time, with the progeny conservatively assumed to be at secular equilibrium at every instant. If inventory margins are encroached upon, then a more formal assessment can be made to determine the actual activity of the progeny if secular equilibrium conditions have yet to be attained.

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**Table 16. Radioisotopes with progeny in secular equilibrium**

<b>Isotope</b>	<b>Decay Series</b>	<b>f<sub>SOF</sub> @ 100 yr</b>	<b>Significant Progeny Contributing to f<sub>SOF</sub> (% of SOF @ 100 yr in-growth)</b>
<sup>186m</sup> Re	--	1.11E+00	<b><sup>186</sup>Re (10.1%)</b>
<sup>245</sup> Cm	<sup>233</sup> U Series (N+1)	1.14E+00	<sup>245</sup> Pu (1.4%), <b><sup>241</sup>Am (10.5%)</b>
<sup>232</sup> Th	<sup>232</sup> Th Series (N)	1.16E+00	<sup>228</sup> Ra (0.7%), <b><sup>228</sup>Th (8.6%), <sup>220</sup>Rn (4.3%)</b>
<sup>210</sup> Pb	<sup>238</sup> U Series (N+2)	1.19E+00	<sup>210</sup> Bi (0.1%), <b><sup>210</sup>Po (5.1%)</b>
<sup>235</sup> U	<sup>235</sup> U Series (N+3)	1.19E+00	<sup>231</sup> Pa (3.7%), <b><sup>227</sup>Ac (12.4%)</b>
<sup>172</sup> Hf	--	1.20E+00	<b><sup>172</sup>Lu (16.5%)</b>
<sup>121m</sup> Te	--	1.27E+00	<b><sup>121</sup>Te (21.1%)</b>
<sup>233</sup> U	<sup>233</sup> U Series (N+1)	1.42E+00	<b><sup>229</sup>Th (29.7%), <sup>225</sup>Ac (0.1%)</b>
<sup>228</sup> Th	<sup>232</sup> Th Series (N)	1.56E+00	<sup>224</sup> Ra (0.3%), <b><sup>220</sup>Rn (32.2%), <sup>212</sup>Pb (0.2%), <sup>208</sup>Tl (3.2%)</b>
<sup>126</sup> Sn	--	1.61E+00	<b><sup>126</sup>Sb (37.8%)</b>
<sup>93</sup> Mo	--	2.00E+00	<b><sup>93m</sup>Nb (8.8%)</b>
<sup>232</sup> U	<sup>232</sup> Th Series (N)	2.34E+00	<b><sup>228</sup>Th (36.0%), <sup>224</sup>Ra (0.2%), <sup>220</sup>Rn (19.0%), <sup>212</sup>Pb (0.1%), <sup>208</sup>Tl (1.9%)</b>
<sup>184m</sup> Re	--	2.75E+00	<b><sup>184</sup>Re (53.0%)</b>
<sup>125</sup> Sb	--	2.77E+00	<b><sup>125m</sup>Te (63.9%)</b>
<sup>192m</sup> Ir	--	3.13E+00	<b><sup>192</sup>Ir (68%)</b>
<sup>32</sup> Si	--	5.33E+00	<b><sup>32</sup>P (81.3%)</b>
<sup>231</sup> Pa	<sup>235</sup> U Series (N+3)	5.58E+00	<b><sup>227</sup>Ac (81.9%), <sup>227</sup>Th (0.1%), <sup>223</sup>Ra(0.1%)</b>
<sup>242m</sup> Am	<sup>238</sup> U Series (N+2)	1.48E+01	<sup>242</sup> Cm (0.9%), <b><sup>238</sup>Pu (31.4%)</b>
<sup>228</sup> Ra	<sup>232</sup> Th Series (N)	2.90E+01	<b><sup>228</sup>Th (61.9%), <sup>224</sup>Ra (0.3%), <sup>220</sup>Rn (31.0%), <sup>212</sup>Pb (0.2%), <sup>208</sup>Tl (3.1%)</b>
<sup>226</sup> Ra	<sup>238</sup> U Series (N+2)	4.06E+01	<sup>222</sup> Rn (3.0%), <b><sup>210</sup>Pb (79.4%), <sup>210</sup>Bi (0.1%), <sup>210</sup>Po (15.0%)</b>

Table 17 and Table 18 displays the radioisotopes that have progeny that are in not in secular equilibrium. The in-growth of progeny does not necessarily decay at the same rate as the parent isotopes. This behavior results in the f<sub>SOF</sub> value growing slowing at first and then rapidly becoming very large as the parent isotope decays away. Similar as before, the progeny that contributed significantly to the SOF assessment are displayed in bold (i.e., progeny that contributes more than 1% of the overall SOF).

Table 17 represents the parent radioisotopes that have half-lives of at least 10 years. These radioactive parents are expected to be significantly decayed, but not necessarily gone at the end of the 100 years assessment period. In each case, only a single progeny was responsible for the increase in the f<sub>SOF</sub> value. Accordingly, only these progeny need to be tracked in the facility inventory.

Table 18 represents the parent radioisotopes that have half-lives less than 10 years. These radioactive parents are expected to be all but consumed by its radioactive decay process at the end of the 100 years assessment period. In each case, indicated progeny comprise at least 85% of the overall SOF. According, tracking of the noted progeny is necessary to ensure the radioactive materials are continued to be assessed in hazard categorization assessment, even as the parent decays away.

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**Table 17. Radioisotopes with progeny in not in secular equilibrium ( $T_{1/2} \geq 10$  yr)**

<b>Isotope</b>	<b><math>T_{1/2}</math> (yr)</b>	<b>Decay Series</b>	<b>Time (yr) @ <math>f_{SOF}=1.10</math></b>	<b>Significant Progeny Contributing to <math>SOF_{mix}</math> (% of SOF @ 100 yr in-growth)</b>
<sup>250</sup> Cf	1.31E+01	<sup>238</sup> U Series (N+2)	5.5E+01	<sup>246</sup> Cm (52.1%)
<sup>244</sup> Cm	1.81E+01	<sup>232</sup> Th Series (N)	7.7E+01	<sup>240</sup> Pu (19.7%)
<sup>241</sup> Pu	1.44E+01	<sup>233</sup> U Series (N+1)	1.0E+00	<sup>241</sup> Am (99.5%)

**Table 18. Radioisotopes with progeny in not in secular equilibrium ( $T_{1/2} < 10$  yr)**

<b>Isotope</b>	<b><math>T_{1/2}</math> (yr)</b>	<b>Decay Series</b>	<b>Time (yr) @ <math>f_{SOF}=1.10</math></b>	<b>Significant Progeny Contributing to <math>SOF_{mix}</math> (% of SOF @ 100 yr in-growth)</b>
<sup>252</sup> Cf	2.64E+00	<sup>232</sup> Th Series (N)	4.0E-03	<sup>248</sup> Cm (0.1%), <sup>244</sup> Pu (98.7%), <sup>240</sup> Pu (1.2%)
<sup>257</sup> Fm	2.75E-01	<sup>233</sup> U Series (N+1)	7.0E-02	<sup>249</sup> Cf (99.1%), <sup>245</sup> Cm (0.9%)
<sup>102m</sup> Rh	5.67E-01	--	1.7E-01	<sup>102</sup> Rh (100%)
<sup>254</sup> Es	7.55E-01	<sup>238</sup> U Series (N+2)	1.7E-01	<sup>250</sup> Cf (49.4%), <sup>246</sup> Cm (50.6%)
<sup>242</sup> Cm	4.46E-01	<sup>238</sup> U Series (N+2)	2.0E-01	<sup>238</sup> Pu (100%)
<sup>248</sup> Cf	9.14E-01	<sup>232</sup> Th Series (N)	2.6E-01	<sup>244</sup> Cm (81.1%), <sup>240</sup> Pu (18.9%)
<sup>174m</sup> Lu	3.89E-01	--	2.9E-01	<sup>174</sup> Lu (100%)
<sup>249</sup> Bk	8.77E-01	<sup>233</sup> U Series (N+1)	9.0E-01	<sup>249</sup> Cf (99.1%), <sup>245</sup> Cm (0.9%)
<sup>145</sup> Sm	9.32E-01	--	9.5E-01	<sup>145</sup> Pm (100%)
<sup>236</sup> Pu	2.85E+00	<sup>232</sup> Th Series (N)	2.2E+00	<sup>232</sup> U (43.2%), <sup>228</sup> Th (36.4%), <sup>224</sup> Ra (0.2%), <sup>220</sup> Rn (18.2%), <sup>212</sup> Pb (0.1%), <sup>208</sup> Tl (1.8%)
<sup>235</sup> Np	1.09E+00	<sup>235</sup> U Series (N+3)	1.3E+01	<sup>235</sup> U (84.1%), <sup>231</sup> Pa (3.7%), <sup>227</sup> Ac (12.1%)
<sup>151</sup> Gd	3.29E-01	--	1.6E+01	<sup>147</sup> Sm (100%)
<sup>147</sup> Pm	2.62E+00	--	5.5E+01	<sup>147</sup> Sm (100%)
<sup>146</sup> Pm	5.53E+00	--	6.8E+01	<sup>146</sup> Sm (85.5%)

As noted in Table 16, Table 17 and Table 18, the radioactive decay series is identified to show the relationship amongst the various radioisotope source materials. Each of the four decay series is represented in these three tables. This indicates that each decay series has its progeny that if allowed to accumulate, can significantly impact the HC-3 SOF.

Four specific mixtures and the impact of progeny

Four specific radioactive mixtures are evaluated by the EPA and their mixture RQs are published in the EPA Technical Background Document as well as Appendix B of 40 CFR 302.4<sup>26</sup>. An equivalent analysis for three of the following four radioactive mixtures is performed here to demonstrate the impact the progeny have on the HC-3 SOF assessment of the source material.

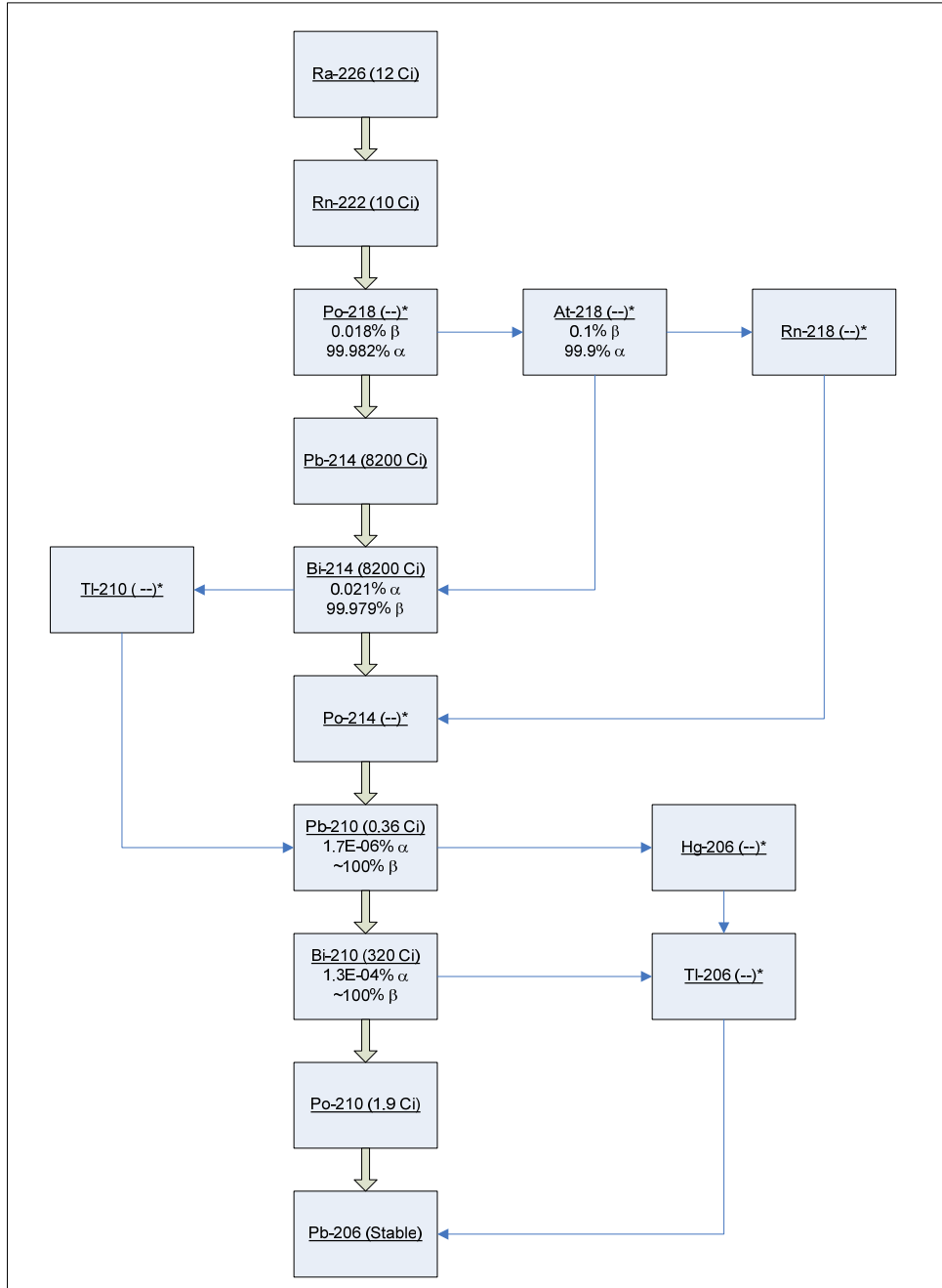
- <sup>226</sup>Ra in secular equilibrium with its progeny
- Natural uranium in secular equilibrium with its progeny (i.e. Uranium ore)
- Natural thorium, in secular equilibrium with its progeny.

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The fourth radioactive mixture discussed by the EPA is chemically separated uranium. That case was previously discussed in the Contaminants section.

*Radium-226 in secular equilibrium with its progeny*

The decay of  $^{226}\text{Ra}$  is illustrated in Figure 6<sup>18</sup>. The values in parenthesis represent the TQ for individual radioisotopes. An "--" in the TQ field indicates a HC-3 TQ value is not defined for that particular radioisotope.



**Figure 6.  $^{226}\text{Ra}$  Decay Series**

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As illustrated in Figure 6, there are nine major progeny (including  $^{206}\text{Pb}$ , a stable isotope) and five minor progeny, produced via small branching ratios.

The half-life of  $^{226}\text{Ra}$  (16,000 y) is relatively long with respect to its progeny. In fact, the longest lived progeny is  $^{210}\text{Pb}$  (22.3 y). Accordingly, the progeny of  $^{226}\text{Ra}$  will eventually be in secular equilibrium in slightly more than 100 years.

In Table 19, the activity of each radioisotope and the TQ fraction for each radioisotope is calculated to determine the  $f_{\text{SOF}}$  value of the source material.

**Table 19. Determination of  $f_{\text{SOF}}$  for  $^{226}\text{Ra}$  in secular equilibrium with its progeny.**

Isotope	HC-3 TQ (Ci)	Act (Ci)	HC-3 SOF	% of HC-3 SOF
$^{226}\text{Ra}$	12	1.00E+00	8.33E-02	2.4%
$^{222}\text{Rn}$	10	1.00E+00	1.00E-01	2.9%
$^{218}\text{Po}$	--	1.00E+00	0.00E+00	--
$^{214}\text{Pb}$	8200	1.00E+00	1.22E-04	--
$^{218}\text{At}$	--	1.80E-04	0.00E+00	--
$^{218}\text{Rn}$	--	1.80E-07	0.00E+00	--
$^{214}\text{Bi}$	8200	1.00E+00	1.22E-04	--
$^{214}\text{Po}$	--	1.00E+00	0.00E+00	--
$^{210}\text{Tl}$	--	2.10E-04	0.00E+00	--
$^{210}\text{Pb}$	0.36	1.00E+00	2.78E+00	79.6%
$^{210}\text{Bi}$	320	1.00E+00	3.12E-03	0.1%
$^{206}\text{Hg}$	--	1.70E-08	0.00E+00	--
$^{210}\text{Po}$	1.9	1.00E+00	5.26E-01	15.1%
$^{206}\text{Tl}$	--	1.32E-06	0.00E+00	--
<b>TOTALS</b>		<b>9.00E+00</b>	<b>3.49E+00</b>	
		<b><math>f_{\text{SOF}}</math></b>	<b>4.19E+01</b>	

The result of Table 19 confirms the data in Table 16, which shows after 100 years of in-growth, a  $^{226}\text{Ra}$  source is close to being in complete secular equilibrium with its progeny. Essentially, this limits the  $^{226}\text{Ra}$  to an effective HC-3 TQ to 0.286 Ci, which is only 2.4% of the published HC-3 TQ. On a total curie basis, the HC-3 TQ for a  $^{226}\text{Ra}$  source in secular equilibrium with its progeny is 2.58 Ci (total source activity).

### Natural uranium in secular equilibrium with its progeny

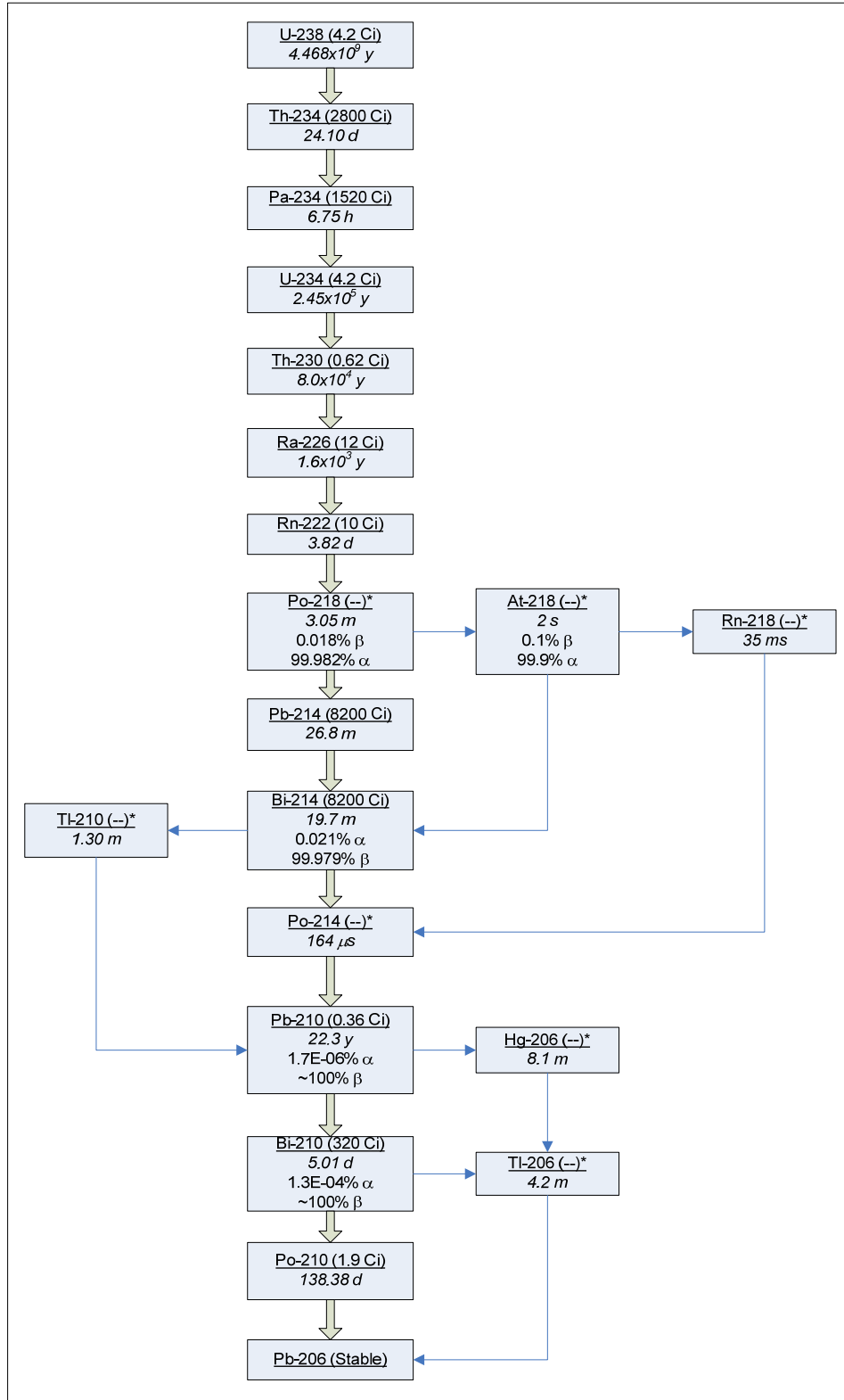
Natural uranium is comprised of three uranium radioisotopes: U-234, U-235 and U-238. The activity abundance of these radioisotopes is specified in Table 20<sup>2</sup>.

**Table 20. Activity abundance of natural uranium radioisotopes**

Isotope	Activity Abundance
U-234	48.83%
U-235	2.34%
U-238	48.83%

The decay of U-238 is illustrated in Figure 7 and the decay of U-235 is illustrated in Figure 8<sup>18</sup>. A separate illustration for U-234 decay is not provided since it is a progeny of the U-238 decay series.

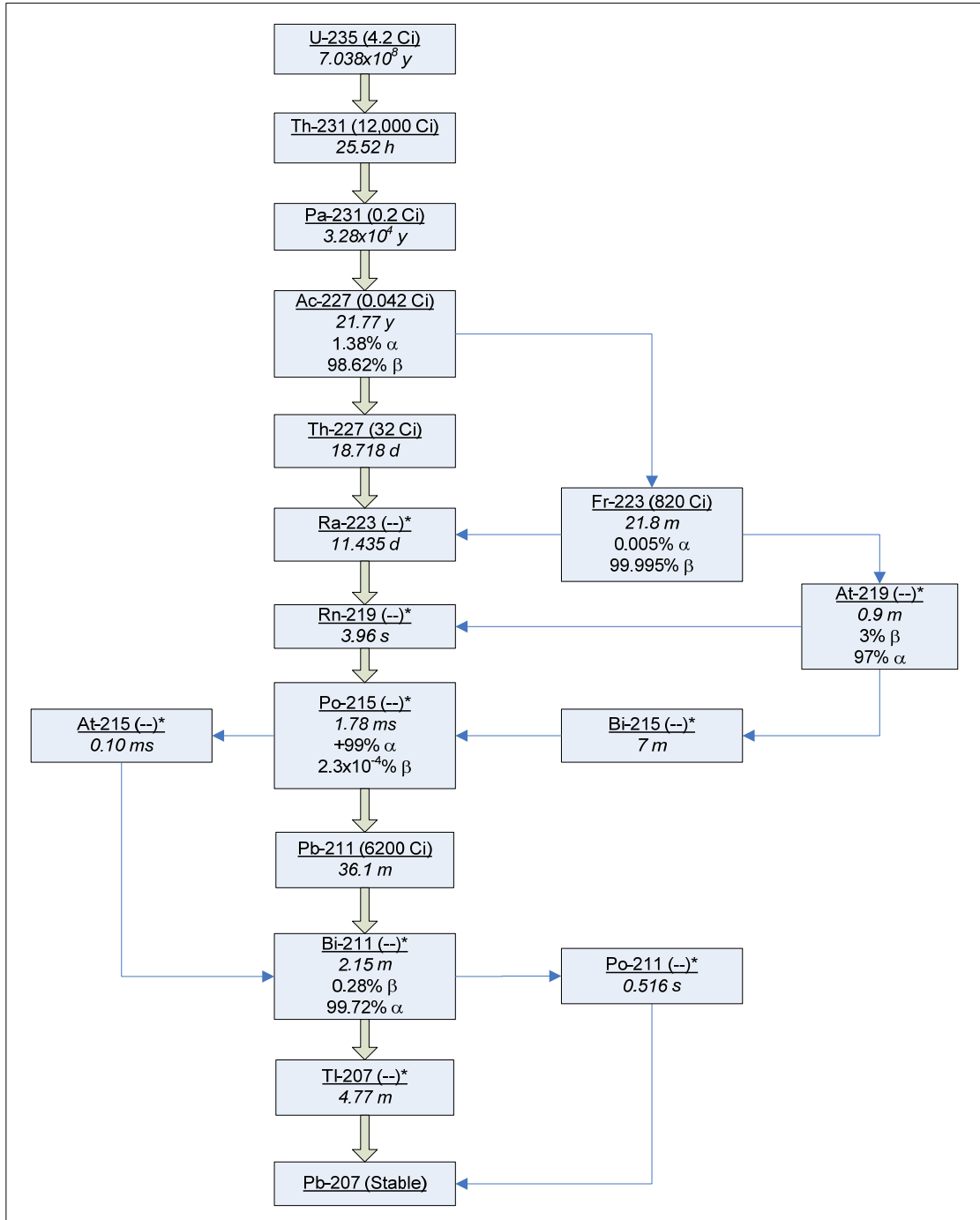
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**Figure 7.  $^{238}\text{U}$  Decay Series**

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**Figure 8.  $^{235}\text{U}$  Decay Series**

As illustrated in Figure 8,  $^{235}\text{U}$  has eleven major progeny (including  $^{207}\text{Pb}$ , a stable isotope) and five minor progeny, produced via minor branching. Similarly, as illustrated in Figure 7,  $^{238}\text{U}$  has fourteen major progeny (including  $^{206}\text{Pb}$ , a stable isotope) and five minor progeny, produced via minor branching decay schemes (same as the  $^{226}\text{Ra}$  decay chain).

Natural uranium ore which has not been chemically treated will have progeny present in secular equilibrium with the uranium isotopes.

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In Table 21, the activity of each radioisotope and the TQ fraction for each radioisotope is calculated to determine the  $f_{\text{SOF}}$  value of the source material.

**Table 21. Determination of  $f_{\text{SOF}}$  for natural uranium in secular equilibrium with its progeny.**

Isotope	HC-3 TQ (Ci)	Act (Ci)	HC-3 SOF	% of HC-3 SOF
<sup>238</sup> U	4.2	--	4.88E-01	3.4%
<sup>234</sup> Th	2800	1.00E+00	4.88E-01	--
<sup>234</sup> Pa	1520	1.00E+00	4.88E-01	--
<sup>234</sup> U	4.2	1.00E+00	4.88E-01	3.4%
<sup>230</sup> Th	0.62	1.00E+00	4.88E-01	23.1%
<sup>226</sup> Ra	12	1.00E+00	4.88E-01	1.2%
<sup>222</sup> Rn	10	1.00E+00	4.88E-01	1.4%
<sup>218</sup> Po	--	1.00E+00	4.88E-01	--
<sup>214</sup> Pb	8200	1.00E+00	4.88E-01	--
<sup>218</sup> At	--	1.80E-04	8.79E-05	--
<sup>218</sup> Rn	--	1.80E-07	8.79E-08	--
<sup>214</sup> Bi	8200	1.00E+00	4.88E-01	--
<sup>214</sup> Po	--	1.00E+00	4.88E-01	--
<sup>210</sup> Tl	--	2.10E-04	1.03E-04	--
<sup>210</sup> Pb	0.36	1.00E+00	4.88E-01	39.8%
<sup>210</sup> Bi	320	1.00E+00	4.88E-01	--
<sup>206</sup> Hg	--	1.70E-08	8.30E-09	--
<sup>210</sup> Po	1.9	1.00E+00	4.88E-01	7.5%
<sup>206</sup> Tl	--	1.32E-06	6.43E-07	--
<b><sup>238</sup>U TOTALS</b>		<b>6.84E+00</b>	<b>2.73E+00</b>	<b>80.0%</b>
<sup>235</sup> U	4.2	2.34E-02	5.57E-03	0.2%
<sup>231</sup> Th	12,000	2.34E-02	1.95E-06	--
<sup>231</sup> Pa	0.2	2.34E-02	1.17E-01	3.4%
<sup>227</sup> Ac	0.042	2.34E-02	5.57E-01	16.4%
<sup>227</sup> Th	32	2.31E-02	7.21E-04	--
<sup>223</sup> Fr	8200	3.23E-04	3.94E-08	--
<sup>223</sup> Ra	62	2.34E-02	3.77E-04	--
<sup>219</sup> At	--	1.61E-08	0.00E+00	--
<sup>219</sup> Rn	--	2.34E-02	0.00E+00	--
<sup>215</sup> Bi	--	1.57E-08	0.00E+00	--
<sup>215</sup> Po	--	2.34E-02	0.00E+00	--
<sup>215</sup> At	--	5.38E-08	0.00E+00	--
<sup>211</sup> Pb	6200	2.34E-02	3.77E-06	--
<sup>211</sup> Bi	--	2.34E-02	0.00E+00	--
<sup>211</sup> Po	--	6.55E-05	0.00E+00	--
<sup>207</sup> Tl	--	2.33E-02	0.00E+00	--
<b><sup>235</sup>U TOTALS</b>		<b>2.57E-01</b>	<b>6.81E-01</b>	<b>20.0%</b>
<b>Nat U TOTALS</b>		<b>7.09E+00</b>	<b>3.41E+00</b>	<b>100.0%</b>
		<b><math>f_{\text{SOF}}</math></b>	<b>2.93E+01</b>	

The results of Table 21 would be indicative natural uranium ore prior to chemical processing (which would result in the removal of the progeny). On a total curie basis, the HC-3 TQ for natural uranium in secular equilibrium with its progeny is 2.08 Ci (total source activity).

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### Natural thorium in secular equilibrium with its progeny

Natural thorium is comprised of two thorium radioisotopes:  $^{232}\text{Th}$  and  $^{228}\text{Th}$ . The decay of  $^{232}\text{Th}$  is illustrated in Figure 9<sup>18</sup>. A separate illustration for the decay of  $^{228}\text{Th}$  is not provided since it is a progeny of the  $^{232}\text{Th}$  decay series.

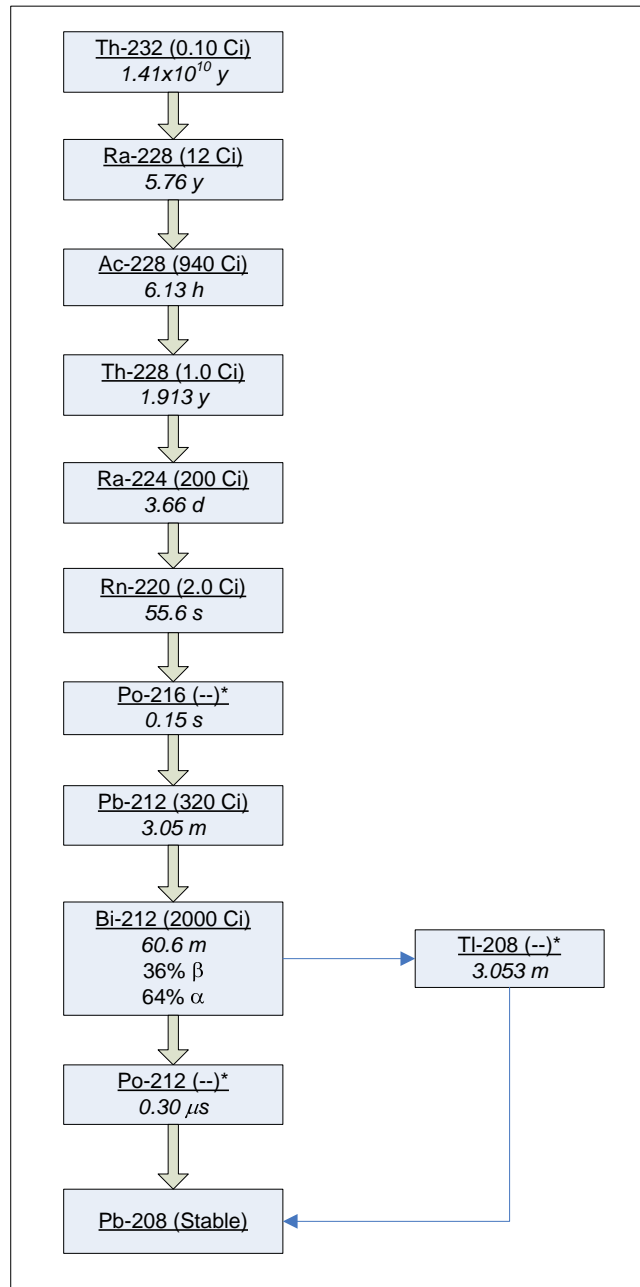


Figure 9.  $^{232}\text{Th}$  Decay Series

As illustrated in Figure 9,  $^{232}\text{Th}$  has ten major progeny (including Pb-208, a stable isotope) and one minor progeny, produced via minor branching.

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Natural thorium ore which has not been chemically treated will have progeny present in secular equilibrium with <sup>232</sup>Th. The time for progeny to build back into secular equilibrium is not long given the short half-lives of the progeny.

In Table 22, the activity of each radioisotope and the TQ fraction for each radioisotope is calculated to determine the  $f_{\text{SOF}}$  value of the source material.

**Table 22. Determination of  $f_{\text{SOF}}$  for natural thorium in secular equilibrium with its progeny.**

Isotope	HC-3 TQ (Ci)	Act (Ci)	HC-3 SOF	% of HC-3 SOF
<sup>232</sup> Th	0.1	1.00E-01	1.00E+00	86.1%
<sup>228</sup> Ra	12	1.00E-01	8.33E-03	0.7%
<sup>228</sup> Ac	940	1.00E-01	1.06E-04	--
<sup>228</sup> Th	1	1.00E-01	1.00E-01	8.6%
<sup>224</sup> Ra	200	1.00E-01	5.00E-04	--
<sup>220</sup> Rn	2	1.00E-01	5.00E-02	4.3%
<sup>216</sup> Po	--	1.00E-01	0.00E+00	--
<sup>212</sup> Pb	320	1.00E-01	3.13E-04	--
<sup>216</sup> At	--	1.00E-01	0.00E+00	--
<sup>212</sup> Bi	2000	1.00E-01	5.00E-05	--
<sup>212</sup> Po	--	6.41E-02	0.00E+00	--
<sup>208</sup> Tl	20	3.59E-02	1.80E-03	0.2%
<b>TOTALS</b>		<b>1.10E+00</b>	<b>1.16E+00</b>	
		<b><math>f_{\text{SOF}}</math></b>	<b>1.16</b>	

The results of Table 22 would be indicative of natural thorium ore prior to chemical processing (which would result in the removal of the progeny). Even after chemical separation, both <sup>232</sup>Th and <sup>228</sup>Th will still be present, resulting  $f_{\text{SOF}} = 1.10$ . On a total curie basis, the HC-3 TQ for natural thorium in secular equilibrium with its progeny is 0.947 Ci (total source activity).

### Impact of progeny and contaminants on fissionable material inventory assessments.

As is the case with the SOF calculations, the fissionable equivalent mass (FEM) and fissile gram equivalent (FGE) calculations can also be impacted by the presence of progeny and contaminants. The fissionable material limits in DOE-STD-1027 define a Hazard Category 2 threshold. The specific radioisotopes that are of concern to nuclear criticality safety assessments are shown in Table 23.

**Table 23. <sup>235</sup>U Fissionable Equivalent Mass Factors<sup>30,31</sup>**

Nuclide	FEM Factor	Nuclide	FEM Factor
<sup>233</sup> U	1.40	<sup>241</sup> Am	0.044
<sup>235</sup> U	1.00	<sup>242m</sup> Am	54
<sup>236</sup> Np	140	<sup>243</sup> Am	0.028
<sup>237</sup> Np	0.035	<sup>243</sup> Cm	7.8
<sup>238</sup> Pu	0.23	<sup>244</sup> Cm	0.23
<sup>239</sup> Pu	1.56	<sup>245</sup> Cm	23
<sup>240</sup> Pu	0.047	<sup>247</sup> Cm	0.78
<sup>241</sup> Pu	3.5	<sup>249</sup> Cf	70
<sup>242</sup> Pu	0.018	<sup>251</sup> Cf	140

The <sup>235</sup>U Fissionable Equivalent Mass (FEM) factor is a normalization that permits the radioisotope of interest to be described in terms of mass of <sup>235</sup>U of equivalent criticality safety concern. The impact in the

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total fissionable mass assessment depends on the mass and FEM factor of the specific contaminant or progeny of concern.

As previously discussed,  $^{241}\text{Am}$  source material can have up to 0.4 wt%  $^{242\text{m}}\text{Am}$  and 4.6%  $^{243}\text{Am}$ <sup>12,13</sup>. By inspection of the FEM factors in Table 23, it is readily apparent that  $^{242\text{m}}\text{Am}$  will have a significant impact on the total FEM of  $^{241}\text{Am}$  source material. Table 24 shows that the presence of  $^{242\text{m}}\text{Am}$  increases the  $^{235}\text{U}$  FEM assessment of  $^{241}\text{Am}$  source material by 6.2 times.

**Table 24. FEM Analysis of  $^{241}\text{Am}$  Source Material**

Nuclide	Mass (g)	FEM Factor	FEM (g)
$^{241}\text{Am}$	0.95	0.044	4.18E-02
$^{242\text{m}}\text{Am}$	0.004	54	2.16E-01
$^{243}\text{Am}$	0.046	0.028	1.29E-03
<b>TOTAL FEM</b>			2.59E-01
<b>f<sub>FEM</sub></b>			6.20E+00

Also as previously noted, source material designated as  $^{252}\text{Cf}$  has californium contaminants and associated curium progeny. In Table 25, a  $^{252}\text{Cf}$  source with 13-<sup>1</sup>/<sub>2</sub> years of progeny in-growth is used as an example to demonstrate the effect of the trace isotopes on the FEM assessment of the source material. Although there is not a FEM factor for  $^{252}\text{Cf}$ , the presence of  $^{249}\text{Cf}$ ,  $^{251}\text{Cf}$  as well as  $^{245}\text{Cm}$  and  $^{247}\text{Cm}$  contribute significantly to the calculated FEM. In fact, the specimen total FEM is 300 times more than the mass of remaining  $^{252}\text{Cf}$  mass due to the presence of the californium and curium trace isotopes.

**Table 25. FEM Analysis of a  $^{252}\text{Cf}$  Source**

Nuclide	Mass <sup>15</sup> (μg)	FEM Factor	FEM (μg)
$^{249}\text{Cf}$	6.69E+02	70	4.68E+04
$^{250}\text{Cf}$	9.26E+02	--	--
$^{251}\text{Cf}$	5.92E+02	140	8.29E+04
$^{252}\text{Cf}$	4.33E+02	--	--
$^{245}\text{Cm}$	1.70E+01	23	3.91E+02
$^{246}\text{Cm}$	9.49E+02	--	--
$^{247}\text{Cm}$	6.00E+00	0.78	4.68E+00
$^{248}\text{Cm}$	1.42E+04	--	--
<b>TOTAL MASS (μg)</b>	1.78E+04	<b>TOTAL FEM (μg)</b>	1.30E+05

Similarly, curium source material should be assessed for trace isotopes to ensure a complete accounting of the fissionable isotopes present. In Table 26,  $^{244}\text{Cm}$  source material is used to demonstrate the effect of the trace isotopes on the FEM assessment of the source material. The presence of  $^{245}\text{Cm}$  and  $^{243}\text{Cm}$  results in a  $^{235}\text{U}$  FEM that is 8.94 times more than contribution of just the  $^{244}\text{Cm}$  material.

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**Table 26. FEM Analysis of <sup>244</sup>Cm Source Material**

Nuclide	Mass <sup>15</sup> (μg)	FEM Factor	FEM (μg)
<sup>243</sup> Cm	2.50E-01	7.8	1.95E+00
<sup>244</sup> Cm	9.18E+01	0.23	2.11E+01
<sup>245</sup> Cm	7.20E+00	23	1.66E+02
<sup>246</sup> Cm	8.00E-01	--	--
<b>TOTAL MASS (μg)</b>	<b>1.00E+02</b>	<b>TOTAL FEM (μg)</b>	<b>1.89E+02</b>
		<b>f<sub>FEM</sub></b>	<b>8.94E+00</b>

In the case of plutonium, there are different grades which yield differing quantities of contaminant isotopes. Similar to the <sup>235</sup>U FEM concept, a <sup>239</sup>Pu fissionable gram equivalent (FGE) relates the criticality potential of various fissionable isotopes in terms of a mass of <sup>239</sup>Pu of an equivalent criticality safety concern.

Table 27 provides the examples of plutonium material of differing isotopic composition. In example 1, the plutonium source material is predominantly <sup>238</sup>Pu. However, <sup>239</sup>Pu has a higher FGE factor than <sup>238</sup>Pu. Although <sup>239</sup>Pu is only comprises 9.1 wt% of example 1, its presence almost doubles the total FGE.

In example 2, <sup>239</sup>Pu is the predominant isotope. The only other isotope with a higher FGE factor is <sup>241</sup>Pu. In this example, there simply isn't enough <sup>241</sup>Pu to significantly affect the total FGE. In example 3, the <sup>239</sup>Pu and <sup>241</sup>Pu isotopes dominant the FGE calculation.

As these examples demonstrate the isotopic content of plutonium source materials should be verified to ensure the FGE (or FEM) assessment is accurate.

**Table 27. Determination of FGE various grades of plutonium material.**

		<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu	Total FGE
Example #1 (90% <sup>238</sup> Pu)	Wt%	90.0%	9.1%	0.6%	0.03%	0.01%	
	<sup>239</sup> Pu FGE Factor	1.13E-01	1.00E+00	2.25E-02	2.25E+00	7.50E-03	
	<sup>239</sup> Pu FGE per g <sup>239</sup> Pu	1.12E+00	1.00E+00	1.48E-03	7.42E-03	8.24E-06	
Example #2 (93.6% <sup>239</sup> Pu)	Wt%	0.1%	93.6%	6.0%	0.40%	0.05%	
	<sup>239</sup> Pu FGE Factor	1.13E-01	1.00E+00	2.25E-02	2.25E+00	7.50E-03	
	<sup>239</sup> Pu FGE per g <sup>239</sup> Pu	6.04E-05	1.00E+00	1.44E-03	9.62E-03	4.01E-06	
Example #3 (58.1% <sup>239</sup> Pu)	Wt%	1.5%	58.1%	24.1%	11.40%	4.90%	
	<sup>239</sup> Pu FGE Factor	1.13E-01	1.00E+00	2.25E-02	2.25E+00	7.50E-03	
	<sup>239</sup> Pu FGE per g <sup>239</sup> Pu	2.92E-03	1.00E+00	9.33E-03	4.41E-01	6.33E-04	

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