# Predicting <sup>232</sup>U Content in Uranium

Anthony J. Peurrung

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Pacific Northwest National Laboratory Richland, Washington 99352

## Summary

The minor isotope  $^{232}$ U may ultimately be used for detection or confirmation of uranium in a variety of applications. The primary advantage of  $^{232}$ U as an indicator of the presence of enriched uranium is the plentiful and penetrating nature of the radiation emitted by its daughter radionuclide  $^{208}$ Tl. A possible drawback to measuring uranium via  $^{232}$ U is the relatively high uncertainty in  $^{232}$ U abundance both within and between material populations. An important step in assessing this problem is to ascertain what determines the  $^{232}$ U concentration within any particular sample of uranium. To this end, we here analyze the production and eventual enrichment of  $^{232}$ U during fuel-cycle operations. The goal of this analysis is to allow approximate prediction of  $^{232}$ U quantities, or at least some interpretation of the results of  $^{232}$ U measurements.

We have found that <sup>232</sup>U is produced via a number of pathways during reactor irradiation of uranium and is subsequently concentrated during the later enrichment of the uranium's <sup>235</sup>U content. While exact calculations are nearly impossible for both the reactor-production and cascade-enrichment parts of the prediction problem, estimates and physical bounds can be provided as listed below and detailed within the body of the report. Even if precise calculations for the irradiation and enrichment were possible, the ultimate <sup>232</sup>U concentration would still depend upon the detailed fuel-cycle history. Assuming that a thermal-diffusion cascade is used to produce highly enriched uranium (HEU), dilution of reactor-processed fuel at the cascade input and the long-term holdup of <sup>232</sup>U within the cascade both affect the <sup>232</sup>U concentration in the product. Similar issues could be expected to apply for the other isotope-separation technologies that are used in other countries.

Results of this analysis are listed below:

- The <sup>232</sup>U concentration depends strongly on the uranium enrichment, with depleted uranium (DU) containing between 1600 and 8000 times less <sup>232</sup>U than HEU does.
- The  ${}^{236}$ U/ ${}^{232}$ U concentration ratio in HEU is likely to be between 10<sup>6</sup> and 2 × 10<sup>7</sup>.
- Plutonium-production reactors yield uranium with between 1 and 10 ppt of  $^{232}$ U.
- Much higher <sup>232</sup>U concentrations can be obtained in some situations.
- Significant variation in the <sup>232</sup>U concentration is inevitable.
- Cascade enrichment increases the <sup>232</sup>U concentration by a factor of at least 200, and possibly as much as 1000.
- The actual <sup>232</sup>U concentration depends upon the dilution at the cascade input.

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## 1.0 Introduction

This brief report is one of a series of reports that discuss issues related to the confirmation of uranium via the detection of <sup>232</sup>U. A report yet to be issued will describe calculations relating to specific uranium confirmation applications such as START III and uranium material interdiction. A report will also be issued describing methods that improve measurement sensitivity by compensating for the variable <sup>232</sup>Th background.

The minor isotope  $^{232}$ U has recently gained interest as the possible key for establishing the presence of uranium for arms control and nuclear smuggling applications. It is becoming clear that  $^{232}$ U is present in sufficient quantities in a large enough fraction of cases to make its use viable. It remains critical, however, to quantify the  $^{232}$ U concentration in the full range of materials that will be encountered in such applications. To this end, a number of measurement campaigns have recorded, or are currently recording, the  $^{232}$ U content of samples from diverse locations.

This brief paper explores the physics by which  $^{232}$ U is formed in reactors and enriched in thermaldiffusion cascades. This work has several objectives. First, the physics must be understood to allow interpretation of notable features in the experimental data. Notable observations so far have included the statistical differences between the uranium produced by different countries, the variation (or lack thereof) of the  $^{232}$ U content within a similar set of measurements, and the occasional occurrence of very high or low  $^{232}$ U concentration relative to the mean. The second objective is to build confidence, which otherwise will be very hard to obtain, that the measurement results accurately describe the entire worldwide uranium inventory. It is standard practice in science to strive for agreement of theory and experiment. Minor isotope quantification is no exception. Only when a comprehensive set of measurements and a theory that explains them are in hand will it be possible to confidently deploy technologies that rely on the presence of  $^{232}$ U.

## 2.0 **Production During Irradiation**

The numbered list below contains four of the most significant nuclear-reaction chains leading to the formation of  $^{232}$ U from  $^{234}$ U,  $^{235}$ U, and  $^{238}$ U during reactor operations:

(1) 
$$^{235}U(\alpha) ^{231}Th(\beta) ^{231}Pa(n,\gamma) ^{232}Pa(\beta) ^{232}U$$

(2) 
$$^{234}U(\alpha) ^{230}Th(n,\gamma) ^{231}Th(\beta^{-}) ^{231}Pa(n,\gamma) ^{232}Pa(\beta^{-}) ^{232}U$$

(3) 
$$^{235}U(n,\gamma) ^{236}U(n,\gamma) ^{237}U(\beta) ^{237}Np(n,2n) ^{236m}Np(\beta) ^{236}Pu(\alpha) ^{232}U$$

(4) 
$$^{238}$$
U(n,2n)  $^{237}$ U( $\beta$ <sup>-</sup>)  $^{237}$ Np(n,2n)  $^{236m}$ Np( $\beta$ <sup>-</sup>)  $^{236}$ Pu( $\alpha$ )  $^{232}$ U

Since uranium frequently contains some <sup>236</sup>U as a result of an admixture of previously reactorprocessed uranium, one additional reaction chain may be important:

(5) 
$$^{236}U(n,\gamma)^{237}U(\beta)^{237}Np(n,2n)^{236m}Np(\beta)^{236}Pu(\alpha)^{232}U(\alpha)^$$

It is assumed here that chemical impurities such as plutonium, neptunium, protactinium, or thorium arising from previous irradiation are not present when the fuel enters the reactor. If this assumption were false, a number of other pathways might become important. These pathways would have starting points such as <sup>237</sup>Np, <sup>241</sup>Pu, <sup>236</sup>Pu, <sup>231</sup>Pa, and <sup>230</sup>Th. Figure 1 shows reaction pathways (1) through (5) in graphical form.



Figure 1. Reaction Chains Leading to the Production of <sup>232</sup>U

While each of the reaction chains listed above leads inevitably to the production of  $^{232}$ U in a reactor neutron flux, each chain contains one reaction that severely limits the overall production rate. The first two reaction chains begin with the alpha decay of  $^{235}$ U and  $^{234}$ U with half-lives of 250 thousand and 700 million years, respectively. Each of the final three listed reaction chains involves at least one (n,2n) reaction that proceeds only rarely within a fission neutron flux. The reaction,  $^{237}$ Np(n,2n) $^{236m}$ Np, is especially limiting with a reaction threshold of roughly 7 MeV and a maximum cross section of roughly 0.4 barns reached only at roughly 10 MeV. Only a small fraction of the fission-neutron-energy spectrum and an even smaller fraction of the overall reactor-energy spectrum has energy sufficient to drive this reaction.

Several notable parameters upon which the ultimate production of  $^{232}$ U must depend can be inferred from the reaction chains listed above. Because several of these parameters can vary substantially from year to year, country to country, or even reactor to reactor, the  $^{232}$ U production rate is not expected to be steady. A list of several important dependencies is given below:

- *Time Before Irradiation* The initial α-decay in reaction chains (1) and (2) proceeds with or without the presence of neutron irradiation. As a result of their long half-lives, the isotopes <sup>230</sup>Th and <sup>231</sup>Pa accumulate during the entire time between fuel fabric ation and irradiation. Thus, the amount of <sup>232</sup>U produced via these two reactions depends strongly on the pre-irradiation time period.
- *Time Before Reprocessing* Each of the reaction chains (3) through (5) produces the isotope  ${}^{236}$ Pu, which  $\alpha$ -decays to the product  ${}^{232}$ U.
- *Uranium Enrichment* The starting enrichment of the fuel used within the reactor determines the relative propensity for reaction chains (1) through (4) to proceed. For example, higher enrichments should reduce the importance of reaction chain (4) relative to the other three and would especially favor reaction chain (2).
- *Reactor Design* The detailed design of the nuclear reactor has a strong influence on the neutronenergy spectrum and thus determines whether reactions involving (n,2n) reactions are favored. Thus, a reactor with a relatively hard neutron-energy spectrum would favor reaction chains (3) and especially (4).
- *Impurities* The presence of impurities or minor isotopes in the reactor fuel can increase the <sup>232</sup>U production rate through reaction chain (5) or any of a number of other reaction chains.
- Irradiation History The dependence of <sup>232</sup>U production on the details of reactor operation is nonlinear and complex. Calculations similar to those described below indicate that power reactors produce 10<sup>3</sup> to 10<sup>4</sup> times more <sup>232</sup>U per unit of fuel mass than plutonium-production reactors. A number of Russian publications (Matveyev and Tsenter 1985; Zaritskaya et al. 1983) also quantify <sup>232</sup>U production in power reactors as a result of their concern for the radiological hazard of reprocessed power-reactor fuel. These results indicate an expected <sup>232</sup>U concentration relative to <sup>238</sup>U of between 10<sup>-8</sup> and 5.0 × 10<sup>-7</sup>, depending on details of the fuel and irradiation histories.

Perkins and Jenquin<sup>1</sup> have performed calculations using the ORIGIN2 (Croff 1980a; Croff 1980b) computer code to determine the actinide content of Hanford fuel at various times after the cessation of irradiation. These calculations assumed that fuel with natural enrichment has no minor or

<sup>&</sup>lt;sup>1</sup> Fission and Activation Products in Nuclear Reactors Fuels and Nuclear Explosion Debris, by RW Perkins and UP Jenquin, PNNL-11554, Pacific Northwest National Laboratory, Richland, Washington (1997).

non-uranium isotopes at the start of irradiation. This assumption cannot be completely true, but is a useful simplification. The irradiation is assumed to consist of 728 MWd/MTU in 75 days. This reflects the actual production cycle used at Hanford. A number of the relevant actinide contents calculated to exist immediately after the cessation of irradiation are shown in Figure 2. The <sup>232</sup>U content indicated in Figure 2, 1 part in  $10^{12}$ , provides an approximate lower bound for a plutonium-production reactor. The existence of a waiting time before irradiation, a finite time before reprocessing, higher burnup, or fuel with higher enrichment could only increase the concentration of <sup>232</sup>U in the product.

The results in Figure 2 allow several conclusions to be drawn about the importance of reaction chains (1) through (4) in the production of  $^{232}$ U. At the moment in which irradiation ceases,  $^{232}$ U production arises 71% from reaction chains (1) and (2), and 29% from reaction chains (3) and (4). A further examination of the data in the Perkins and Jenquin report indicates that reactions (1) and (2) produce 60% and 40% of the  $^{231}$ Pa, respectively, that leads ultimately to the production of  $^{232}$ U (at the moment when irradiation ceases.) While these percentages are not intended as exact guides, they do serve to indicate that a number of production processes compete in the production of  $^{232}$ U.

		<sup>236</sup> Pu	<sup>237</sup> Pu	<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>240</sup> Pu
		$1 \times 10^{-6}$	$3 \times 10^{-7}$	$2 \times 10^{-1}$	$8 \times 10^2$	$5 \times 10^{1}$
			<sup>236</sup> Np	<sup>237</sup> Np	<sup>238</sup> Np	<sup>239</sup> Np
			$1 \times 10^{-7}$	$6 \times 10^{\circ}$	$1 \times 10^{-2}$	$4 \times 10^{1}$
<sup>232</sup> U	<sup>233</sup> U	<sup>234</sup> U	<sup>235</sup> U	<sup>236</sup> U	<sup>237</sup> U	<sup>238</sup> U
$1 \times 10^{-6}$	$4 \times 10^{-5}$	$6 \times 10^{1}$	$7 \times 10^3$	$1 \times 10^2$	$1 \times 10^{\circ}$	$1 \times 10^{6}$
<sup>231</sup> Pa	<sup>232</sup> Pa	<sup>233</sup> Pa				
$2 \times 10^{-6}$	$3 \times 10^{-9}$	$1 \times 10^{-7}$				
<sup>230</sup> Th	<sup>231</sup> Th	<sup>232</sup> Th		<sup>234</sup> Th		
$3 \times 10^{-5}$	$5 \times 10^{-8}$	$4 \times 10^{-7}$		$1 \times 10^{-5}$		

**Figure 2.** Abundance of  $^{232}$ U in Units of mg/kg of  $^{238}$ U. These abundances exist immediately after a 75-day irradiation resulting in a fuel burnup of 728 MWd/MTU.

The results in Figure 2 also allow a rough estimate of how the <sup>232</sup>U production depends on several of the parameters discussed above. Since the data shown in Figure 2 assume that no <sup>231</sup>Pa and no <sup>230</sup>Th are present at the start of irradiation, these results only reflect the case where zero time elapses between fuel fabrication and irradiation. Since the assumed irradiation was only 75 days, a 2-year delay could clearly increase the <sup>232</sup>U production via reaction chains 1 and 2 by more than a factor of 10. Additionally, note that the abundance of <sup>236</sup>Pu and <sup>232</sup>U are equal at the cessation of irradiation. Since <sup>236</sup>Pu decays to <sup>232</sup>U with a 2.87-year half-life, the overall production of <sup>232</sup>U will be roughly doubled should 3 or more years elapse between irradiation and fuel reprocessing. Clearly, the use of fuel that has <sup>234</sup>U and <sup>235</sup>U enriched above natural abundances will lead to the formation of proportionally more <sup>232</sup>U via reaction chains (1) and (2).

In summary, the production of plutonium in reactors using natural uranium cannot lead to the production of less than 1 ppt of  $^{232}$ U in the product uranium. Additional production resulting from preirradiation delays, post-irradiation delays, higher burn-up, higher enrichment fuel, or hard neutronenergy-spectrum reactor design may easily increase the  $^{232}$ U production rate by an order of magnitude or more. The typical value is likely to be roughly 3 ppt, with considerable scatter dependent on variations in the fuel cycle over the course of a production campaign. It is important to also note that dramatically higher <sup>232</sup>U contents are possible for substantially different reactor designs or for situations in which fuel is repeatedly recycled and/or contains significant impurities. To illustrate this point, the <sup>232</sup>U contents of power-reactor fuel are likely to be 3 or 4 orders of magnitude larger than the <sup>232</sup>U content of uranium used in plutonium-production reactors.

## 3.0 Enrichment Within the Cascade

The <sup>232</sup>U content that is to be expected for highly enriched uranium (HEU) and depleted uranium (DU) depends not only on the <sup>232</sup>U production within reactors, but also upon the behavior of <sup>232</sup>U within isotope-separation hardware used to make HEU. Because it is the lightest long-lived uranium isotope, <sup>232</sup>U can be expected to undergo transport primarily to the high-enrichment output of whatever technology is used. As with the previous section on reactor production of <sup>232</sup>U, this section attempts to outline the issues involved in predicting <sup>232</sup>U enrichment and provide sensible bounds and estimates to allow some understanding of the experimental measurements being performed by several laboratories within the DOE complex. Although it is generally assumed within this section that a thermal-diffusion cascade is used for uranium enrichment, much of the physics should be similar for alternate technologies.

A cautionary note about the following estimates is best stated at the outset. Much like the preceding calculations on reactor production of  $^{232}$ U, calculations of  $^{232}$ U enrichment cannot possibly be exact without a detailed knowledge of the cascade's operation, which is unavailable at the present time. For example, the long-term holdup of  $^{232}$ U in the cascade causes the  $^{232}$ U concentration in the enriched product to depend upon the cascade's history over at least the previous weeks or months. Once a cascade is "contaminated" with reactor-processed uranium,  $^{232}$ U is likely to appear in the enriched product for some time subsequently. Additionally, the  $^{232}$ U concentration in the enriched product depends upon the detailed mode in which the cascade is operated. Even for a cascade whose design has been fixed by construction, there are a number of free parameters that can be changed, including: product enrichment, feed enrichment, tails enrichment, side-stream removal rate, and side-stream enrichment. Additionally, enrichment from natural uranium to highly enriched uranium may be accomplished using two entirely different cascades. As will be illustrated by some of the calculations below, the product  $^{232}$ U concentration does depend strongly on these operational choices.

A number of different assumptions can be used to provide estimates and bounds for the  $^{232}$ U enrichment. Each of these methods is listed and explained briefly below:

- 1. *Uranium-235 lower bound* Clearly, the <sup>232</sup>U enrichment cannot be less than the factor by which the <sup>235</sup>U concentration is increased by the cascade. Assuming natural uranium feed and a product <sup>235</sup>U content of E, this method leads to a firm lower bound for the <sup>232</sup>U concentration of 140E. For HEU with 90% <sup>235</sup>U, this method predicts that the <sup>232</sup>U enrichment should exceed 125.
- 2. Uranium-235 separation-factor estimate The "power" of an enrichment cascade is formally expressed by a ratio known as the "separation factor." For a binary mixture of species A and B with input and output concentration of  $A_I$ ,  $B_I$ ,  $A_O$ , and  $B_O$ , the separation factor, S, is defined by the relation,  $E = (A_O/A_I)(B_I/B_O)$  (Perry and Green 1997, ch. 16, p.14). Again assuming natural uranium feed and a product <sup>235</sup>U content of E, the separation factor becomes S = 140(E)/(1-E). For HEU with 90% <sup>235</sup>U, the separation factor becomes 1250. Since the separation factor for <sup>232</sup>U should at least equal (more below) the separation factor for <sup>235</sup>U, we might choose to conclude that 1250 is a lower bound for the <sup>232</sup>U separation factor. Although the separation factor represents only what is possible at equilibrium and not what will be achieved in an operating cascade. The separation factor might serve, however, as an estimate of what the <sup>232</sup>U enrichment might be under some conditions.
- 3. Extrapolation based on <sup>236</sup>U and <sup>234</sup>U Another way of estimating the <sup>232</sup>U enrichment achieved within the cascade is to extrapolate on the basis of the two other dilute uranium components, <sup>234</sup>U and <sup>236</sup>U. A series of papers by Blumkin and Von Halle (1972; 1976) indicates that the enrichments are roughly 50 and 250 for <sup>234</sup>U and <sup>236</sup>U, respectively. A simple geometric extrapolation would therefore lead to an estimate of roughly 1250 for the enrichment of <sup>232</sup>U in the same cascade, in support of the estimate above. The primary flaw in this estimate lies in the extrapolation itself, which should only

be taken as crude. Note that this extrapolation is only valid for the particular assumptions made in the Blumkin and Von Halle paper. It might best be concluded that extrapolation supports a lower bound on the <sup>232</sup>U enrichment of 250 and supports the possibility that enrichments could exceed 1000 in some cases.

- 4. Mass conservation for a simple cascade Both this and the next enrichment estimation method invoke the principle of mass conservation under the assumption that *all* of the <sup>232</sup>U that enters the cascade winds up in the high-enrichment product. A simple cascade is assumed to use natural uranium feed and yield an enriched product with <sup>235</sup>U content E and a depleted product with <sup>235</sup>U content D. Under the assumption stated above, mass conservation implies that the <sup>232</sup>U enrichment is (E-D)/(N-D), where N=0.0072 is the <sup>235</sup>U content in the natural (feed) uranium. For a depleted <sup>235</sup>U content of 0.25% and an HEU content of 90%, the <sup>232</sup>U enrichment cannot exceed 191. In fact, 190 is probably a reasonable estimate for the <sup>232</sup>U enrichment for this simple cascade because the <sup>232</sup>U is overwhelmingly likely to exit the cascade with the high-enrichment product.
- 5. Mass conservation in a complex cascade As an example, we here calculate the <sup>232</sup>U enrichment expected under the assumption that a side stream removes roughly 10% of the input uranium (by mass) at an enrichment of 4%. These assumptions match those used in an example in the later Blumkin and Von Halle paper. Such side streams and other complexities are not uncommon in cascade operation. Mass conservation for this case implies that the <sup>232</sup>U enrichment is (E-0.04)/(N-.9D-0.004), or roughly 1100 for a depleted <sup>235</sup>U content of 0.25% and an HEU content of 90%. While this calculation yields an upper bound valid only for the particular assumptions used, it should serve as a plausible estimate for these assumptions and a guide to indicate how the <sup>232</sup>U enrichment can sensitively depend upon the details of cascade operation.

The above set of estimates supports the conclusion that the <sup>232</sup>U content in the cascade output is at least 250, and possibly as much as 1000 times larger than that of the natural uranium input to the cascade. Note that one cannot combine a <sup>232</sup>U enrichment factor such as 500 with the expected typical post-reprocessing <sup>232</sup>U concentration of 3 ppt to conclude that the <sup>232</sup>U concentration in HEU should be roughly 1.5 ppb. This <sup>232</sup>U concentration would be correct only for the case where all of the uranium input to the cascade consists of reactor-processed uranium. The actual <sup>232</sup>U concentrations may well be less should reactor processed uranium comprise only a fraction of the uranium that is input to the cascade.

## 4.0 Selected Applications

#### 4.1 Other Enrichments:

For START III applications, it is important to estimate the <sup>232</sup>U content of uranium with <sup>235</sup>U contents greater than natural, but less than the 90% used in the numerical examples above. Figure 3 shows the results obtained by using methods 1, 2, 4, and 5 above to estimate the <sup>232</sup>U enrichment with the assumption that the depleted uranium tails stream contains 0.25% <sup>235</sup>U. This figure gives a sense of both how the <sup>232</sup>U enrichment depends upon the cascade operational parameters and how the <sup>232</sup>U enrichment is likely to depend upon the <sup>235</sup>U content of the enriched product. The meaning of the curves is again summarized below:

- 1. Firm physical lower bound.
- 2. Minimum <sup>232</sup>U enrichment at cascade equilibrium (which may never be established).
- 4. Upper bound and good estimate for simple cascade.
- 5. Upper bound and reasonable estimate for complex cascade.



**Figure 3.** Four Estimates of the <sup>232</sup>U Enrichment as a Function of the <sup>235</sup>U Product Content Assuming a Depleted Tails Stream Containing 0.25% <sup>235</sup>U

#### 4.2 Depleted Uranium

Estimation of the <sup>232</sup>U content of depleted uranium is also important for START III and similar applications because it is desirable not to misidentify depleted uranium as uranium with a higher enrichment. The fate of <sup>232</sup>U within the "tails" part of the cascade is especially easy to estimate since both <sup>235</sup>U and <sup>232</sup>U are dilute within this part of the cascade and because the function of this part of the cascade is to remove, or "strip," all non-<sup>238</sup>U components. The depletion factor for <sup>232</sup>U can be estimated as  $(F/D)^2$ , where F is the <sup>235</sup>U content in the feed uranium and D is, as usual, the <sup>235</sup>U content of the depleted product. Under the assumptions of 0.72% and 0.25% <sup>235</sup>U contents for the feed and depleted product, the depletion factor for <sup>232</sup>U becomes 8.3. Incidentally, the same result is obtained using estimate method 3 above. Extrapolation of the known depletions of <sup>236</sup>U and <sup>234</sup>U to the unknown

depletion for  $^{232}$ U leads to the conclusion that DU should contain roughly eight times less  $^{232}$ U than that of the cascade input.

Combining the results above, we can infer that the ratio of the <sup>232</sup>U contents in highly enriched uranium and depleted uranium is between roughly 1600 and 8000, depending on the detailed mode of cascade operation. It would therefore take one metric ton of DU to contain as much <sup>232</sup>U as a single kilogram of HEU. It appears safe to conclude that the presence of DU should not affect measurements using <sup>232</sup>U as an indicator of the presence of HEU, except inasmuch as the DU may shield the 2614 keV gamma-ray flux.

## 4.3 Relating <sup>232</sup>U and <sup>236</sup>U

Provided that a quantitative relationship between <sup>232</sup>U and <sup>236</sup>U can be developed, the question of <sup>232</sup>U abundance may be answered indirectly by using the relatively large quantity of available data on the <sup>236</sup>U content of uranium. Notably, the <sup>236</sup>U content of weapons-grade uranium may have been tightly controlled to enhance reliability. Any such control would have also led to some control on the <sup>232</sup>U and <sup>236</sup>U content of weapons-grade uranium. Good reasons exist to trust in a relationship between the <sup>232</sup>U and <sup>236</sup>U concentrations. Neither of these isotopes is natural; both are produced during the irradiation of uranium within a nuclear reactor. Consequently, any dilution that takes place at the input to an isotope-separation technology will affect both <sup>232</sup>U and <sup>236</sup>U equally and not affect the ratio between the two.

The relationship derived below, while approximate, is quantitative and should hold as long as the assumptions used are valid. We start by assuming that plutonium production yields uranium containing 3 ppt of <sup>232</sup>U. As described in section 2, this value is uncertain by roughly a factor of 2, but almost certainly not more than a factor of 3. This same plutonium production yields uranium with a <sup>236</sup>U content of approximately 100 ppm. This value is expected to be much more constant than the <sup>232</sup>U concentration since the dominant reaction that produces it is the simple neutron capture reaction on <sup>235</sup>U, <sup>235</sup>U(n, $\gamma$ )<sup>236</sup>U. Thus, the 236-to-232 ratio in any material fed to the enrichment cascade should be 3 × 10<sup>7</sup> within a factor of roughly 2. Subsequent enrichment affects both <sup>236</sup>U and <sup>232</sup>U, but is expected to enrich the <sup>236</sup>U concentration by only a factor of roughly 50. Thus, the 236-to-232 ratio is decreased during enrichment by a factor that is likely to be between 4 and 20, depending upon the details of cascade operation as discussed in the previous section. Table 1 summarizes the range of ratios between the <sup>236</sup>U and <sup>232</sup>U contents of HEU.

	High Reactor <sup>232</sup> U	Low Reactor <sup>232</sup> U
High <sup>232</sup> U Enrichment	$\sim 1.0 \times 10^{6}$	$\sim 3.0 \times 10^{6}$
Low <sup>232</sup> U Enrichment	$\sim 4.0 \times 10^{6}$	$\sim 1.7 \times 10^{7}$

 Table 1. Ratio of <sup>236</sup>U Content to <sup>232</sup>U Content for Uranium Containing 90% <sup>235</sup>U

Both the <sup>232</sup>U and <sup>236</sup>U contents of a particular sample of oralloy are provided in the reference by Moody (1994). This analysis indicates a 236-to-232 ratio of  $1.8 \times 10^7$ . More data are clearly needed to confirm this relationship before it can be widely used to predict the <sup>232</sup>U content of uranium.

While the ratios shown in the above table are intended for HEU, these ratios are not terribly sensitive to uranium enrichment. Note that the range of ratios for reactor-processed uranium,  $1.5-6.0 \times 10^7$ , is only somewhat higher than the range valid for HEU. The appropriate ratios for uranium of intermediate enrichment could be obtained by interpolation. However, it must be noted that the uncertainties in these ratios are probably too large to warrant a detailed quantitative analysis. Depleted

uranium would be expected to have a  ${}^{236}U/{}^{232}U$  ratio of roughly 10<sup>8</sup>, since  ${}^{232}U$  is more effectively prevented from reaching the tails output of the enrichment cascade.

## 5.0 Further Work

The approximate relationships derived in this work can clearly be improved, but not without significant effort. Should future events make such improved estimates desirable, the following areas of work could be expected to lead to the greatest improvements in our ability to predict <sup>232</sup>U abundances.

- The detailed fuel cycle used within the United States should be studied to determine its impact on the above calculations. Examples of parameters that should be studied include the time between fuel fabrication and irradiation, the time between irradiation and reprocessing, and the detailed operational mode of the enrichment cascade during uranium enrichment.
- The fate of <sup>232</sup>U and <sup>236</sup>U during isotope separation by technologies other than thermal diffusion should be carefully studied.
- The <sup>232</sup>U enrichment in the cascade should be estimated using numerical codes rather than simple bounding estimates as used above.
- Research should be done to improve the quantitative relationship between <sup>232</sup>U content and <sup>236</sup>U content.

## 6.0 References

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