

## Methods to Vary the U<sup>234</sup> Isotope Concentration of Uranium

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### Description of Processes Leading to U234 Concentration of Natural Uranium:

Uranium 238 (U<sup>238</sup>) undergoes alpha decay to thorium 234, which undergoes beta decay to protactinium 234, which undergoes beta decay to uranium 234. The half-life of U<sup>238</sup> is T<sub>238</sub>=4.468\*10<sup>9</sup> years and the half-life of U<sup>234</sup> is T<sub>234</sub>=2.445\*10<sup>5</sup> years. The two isotopes of uranium are essentially chemically identical so they will not be separated by any geo-chemical process. Therefore, in deposits of uranium, these two isotopes would be expected to exist in radioactive equilibrium and the ratio of concentrations would equal the ratio of half-lives, that is, the concentration of U<sup>234</sup> should be 53.8 ppm. However, the route from one uranium isotope to another passes through two non-uranium species that are chemically distinct and these might be separated. Thus, different mines can produce different U<sup>234</sup> concentrations because of different local conditions [1] and these different U<sup>234</sup> concentrations can be used to identify the source of a sample of uranium. The United States used U<sup>234</sup> concentration measurements to argue that uranium found in Libya was of North Korean origin. How reliable is this method of source identification, particularly if the country of origin wishes not to be identified? Below we discuss different ways to alter U<sup>234</sup> concentrations to disguise the origin of a mass of uranium.

### Simple Mixing:

Natural uranium has a U<sup>234</sup> content ranging from about 48.4 parts per million (ppm) to 62.1 ppm [1]. For n samples of Uranium each with concentration c<sub>i</sub> of a given isotope and of amount a<sub>i</sub>, the concentration of the total combined sample c<sub>final</sub> is given by:

$$\frac{\sum_{i=1}^n (c_i \cdot a_i)}{\sum_{i=1}^n a_i} = c_{\text{final}} \quad (1)$$

If we were to combine samples from a mine with a relatively high concentration of U234 and a mine with a relatively low concentration, this equation then takes the simple form:

$$\frac{60\text{ppm} \cdot a_1 + 49\text{ppm} \cdot a_2}{a_1 + a_2} = c_{\text{final}} \quad (2)$$

Thus, c<sub>final</sub> for U<sup>234</sup> can then have any value in the range 49ppm to 60ppm. This is true for any such combination of samples with different concentrations of the same isotope.

## U234 Concentration Alteration from Gas Centrifuge Cascades:

Uranium enrichment facilities that utilize gas centrifuge cascades can yield a product with a number of  $U^{235}$  concentrations. Low Enriched Uranium (LEU) is defined as Uranium with a  $U^{235}$  concentration of .00711 to .2, Highly Enriched Uranium (HEU) has a  $U^{235}$  concentration above .2, and Weapons-Grade Uranium has a  $U^{235}$  concentration of .9 or higher. The waste from these enrichment processes is generally .001 to .004  $U^{235}$ . Most of the following math can be found in [2]. Variables are defined as follows:

$c_F =$   $U^{235}$  concentration of the feed for a uranium cascade (.0711 for natural uranium).

$c_w =$   $U^{235}$  concentration of the waste from a uranium cascade (generally .001-.004).

$c_p =$   $U^{235}$  concentration of the product from a Uranium cascade.

$c_{Fm} =$   $U^{234}$  concentration of the feed for a Uranium cascade (48.4ppm-62.1ppm).

$\Delta\mu_m =$  The atomic mass difference between U234 and U235 (-1).

We also define the following matched-abundance ratios that specify the relative abundance of  $U^{234}$  after a cascade in terms of the fuel, product, and waste concentrations of  $U^{235}$  [2]:

$$R_F = \left( \frac{1 - c_F}{c_F} \right)^{1 - \frac{2}{3} \cdot \Delta\mu_m} \quad R_P = \left( \frac{1 - c_P}{c_P} \right)^{1 - \frac{2}{3} \cdot \Delta\mu_m} \quad R_w = \left( \frac{1 - c_w}{c_w} \right)^{1 - \frac{2}{3} \cdot \Delta\mu_m} \quad (3)$$

And the ratio:

$$f := \frac{R_F - R_P}{R_w - R_F} \quad (4)$$

It is then possible to calculate the  $U^{234}$  concentration of the product :

$$c_{pm} = \frac{c_p - c_w}{c_F - c_w} \cdot \frac{c_{Fm}}{1 + f} \quad (5)$$

Using the mass conservation equation analogous to (1) where W, P, and F=P+W are the amounts of Waste, Product, and Feed.:

$$c_F \cdot F = c_p \cdot P + c_w \cdot W \quad (6)$$

We can then calculate the amount of feed necessary to produce a given amount of product ( $U^{235}$ ), set P=1 and solve for W (with  $c_F$ ,  $c_p$ ,  $F=1+W$ , and  $c_w$ ) to yield :

$$W = \frac{c_p - c_F}{c_F - c_w} \quad (7)$$

In Table 1 we present the  $U^{234}$  concentrations of products from  $c_p=(.01,.02,.03,.04,.05,.2,.9)$  and  $c_{fm}=(49\text{ppm}, 60\text{ppm})$  at the specified  $U^{234}$  and  $U^{235}$  concentrations. The values for F, P, W, and SWU (Separative work units to produce 1kg) have also been included.

**Table 1.** Product Concentrations of  $U^{234}$  (cpm324),  $U^{235}$  (cp235), amounts of Feed/Waste/Product, and Separative Work Units to produce 1kg of product material at a given concentration. This includes initial  $U^{234}$  concentrations of  $c_{fm} = (49 \& 60 \text{ ppm})$ .

	<i>cfm=49ppm</i>	<i>cfm=60ppm</i>				
<b>cp235</b>	<b>cpm234</b>	<b>cpm234</b>	<b>F</b>	<b>P</b>	<b>W</b>	<b>SWU</b>
0.01	72	89	1.57	1.00	0.57	0.38
0.02	155	190	3.52	1.00	2.52	2.19
0.03	239	292	5.48	1.00	4.48	4.31
0.04	323	395	7.44	1.00	6.44	6.54
0.05	407	498	9.39	1.00	8.39	8.85
0.2	1672	2047	38.75	1.00	37.75	45.75
0.9	7580	9282	175.73	1.00	174.73	227.34

When looking for wider ranges of concentration for  $U^{234}$ , one must also look at the concentration of the waste products from typical  $U^{235}$  enrichment processes. To determine this concentration a similar technique as before is utilized, but set W=1 and then solve for P:

$$P := \frac{c_w - c_F}{c_F - c_p} \quad (8)$$

Then, take these values of F, P, W and put them into the new equation for concentrations of  $U^{234}$ :

$$c_{wm} = \frac{c_{Fm} \cdot F - c_{pm} \cdot P}{W} \quad (9)$$

The results of this calculation are presented below in Table 2. In this case, the SWUs and amounts of Fuel are those necessary to produce 1 kg of waste.

**Table 2.** Waste Concentrations of  $U^{234}$  (cwm234) ,  $U^{235}$  (cp235), amounts of Feed/Waste/Product, and Separative Work Units to produce 1kg of waste material at a given concentration. This includes initial  $U^{234}$  concentrations of cfm = (49 and 60 ppm).

cp235	cfm=49ppm	cfm=60ppm	F	P	W	SWU
	cwm234	cwm234				
0.01	7.60	9.31	2.77	1.77	1.00	0.67
0.02	6.91	8.46	1.40	0.40	1.00	0.87
0.03	6.62	8.11	1.22	0.22	1.00	0.96
0.04	6.46	7.91	1.16	0.16	1.00	1.02
0.05	6.36	7.78	1.12	0.12	1.00	1.05
0.2	6.01	7.36	1.03	0.03	1.00	1.21
0.9	5.90	7.23	1.01	0.01	1.00	1.30

As seen in Tables 1 and 2,  $U^{234}$  concentrations can range anywhere from 5.90 to 9281.56. If one were to also consider the fact that different cascade schemes yield different concentrations of  $U^{235}$  in the waste (.001-.004), then this range is even wider.

It is quite trivial to have a sample with more or less Uranium 234 just by combinations of products and wastes. To make a combined sample resemble natural Uranium it is necessary to combine samples in such a way that the  $U^{235}$  concentration is .7% and the  $U^{238}$  concentration 99.3%. If one were to combine the product and waste from the same cascade the material would resemble the original natural sample, but the  $U^{234}$  concentration would not have changed. If we were to take product and waste concentrations from different enrichment processes, it is possible to increase and even decrease the  $U^{234}$  concentration of the resultant combined material while keeping the  $U^{235}$  and  $U^{238}$  concentrations at levels resembling natural Uranium.

To determine the requisite amounts of material required to give a sample resembling natural Uranium, solve equation (6) for a given value of  $c_p$ , set  $c_F$  to the desired  $U^{235}$  concentration (i.e. that of natural uranium), and note that the  $c_w$  (or waste concentration) is the same for all processes. In this case  $c_p=.002$ . It is then possible to vary the amount of  $U^{234}$  by picking from different waste and product combinations. This is due to the different  $U^{234}/U^{235}$  relative concentrations for each product and waste. In Tables 3 and 4 we show possible  $U^{234}$  concentrations for combinations of waste and product that yield a  $U^{235}$  concentration of .00711. Each of these tables shows combinations of product and waste for one initial  $U^{234}$  concentration. Further combinations of these waste and product samples from different initial  $U^{234}$  concentrations are also possible.

**Table 3.** Resulting concentrations of  $U^{234}$  from combinations of product (cp) and waste cw(cp) from different cascades such that the  $U^{235}$  content is equal to that of Natural Uranium. The initial  $U^{234}$  Concentration is assumed to be 49ppm.

cw(cp)	cp						
	0.01	0.02	0.03	0.04	0.05	0.2	0.9
cw(0.01)	49.0	49.0	49.8	50.0	50.1	50.6	50.7
cw(0.02)	48.7	48.8	49.2	49.4	49.5	49.9	50.0
cw(0.03)	48.6	48.7	49.0	49.1	49.2	49.6	49.7
cw(0.04)	48.6	48.6	48.9	49.0	49.1	49.4	49.6
cw(0.05)	48.6	48.4	48.8	48.9	49.0	49.3	49.5
cw(0.2)	48.4	48.3	48.5	48.6	48.7	49.0	49.1
cw(0.9)	48.4	44.1	48.4	48.5	48.6	48.9	49.0

**Table 4.** Resulting concentrations of  $U^{234}$  from combinations of product (cp) and waste cw(cp) from different cascades such that the  $U^{235}$  content is equal to that of Natural Uranium. The initial  $U^{234}$  Concentration is assumed to be 60ppm.

cw(cp)	cp						
	0.01	0.02	0.03	0.04	0.05	0.2	0.9
cw(0.01)	60.0	60.0	61.0	61.2	61.4	61.9	62.1
cw(0.02)	59.7	59.7	60.3	60.5	60.6	61.1	61.2
cw(0.03)	59.6	59.6	60.0	60.2	60.3	60.7	60.9
cw(0.04)	59.5	59.5	59.8	60.0	60.1	60.5	60.7
cw(0.05)	59.5	59.2	59.7	59.9	60.0	60.4	60.6
cw(0.2)	59.3	59.1	59.4	59.5	59.6	60.0	60.1
cw(0.9)	59.2	53.9	59.3	59.4	59.5	59.9	60.0

From these tables it is evident that combinations of products and wastes from different cascades can yield samples of the same  $U^{235}$  concentrations but differing  $U^{234}$  concentrations. If such an enrichment scheme were then reapplied to the resultant samples it would be possible to increase or decrease the  $U^{234}$  concentrations to even further amounts. However, such a scheme would be excessively cost-intensive and only result in a small shift in the  $U^{234}$  concentration. It would be a way to decrease the  $U^{234}$  concentration (an effect unobtainable by other methods), but for other purposes its end result would not justify the cost.

#### Uranium 234 from Plutonium 238:

Another possible source of  $U^{234}$  is from the alpha decay of Plutonium 238:  ${}_{94}\text{Pu}^{238} \rightarrow \alpha + {}_{92}\text{U}^{234}$  [4]. The half-life for this decay is  $t_{\text{half}}=87.7$  years, so substantial portions of an older plutonium sample could potentially be  $U^{234}$ .

Plutonium with a high concentration of the isotope  $\text{Pu}^{238}$  is not generally usable in nuclear weapons. Safeguards on it are less stringent than those on plutonium with a high concentration of  $\text{Pu}^{239}$ . NASA utilizes High- $\text{Pu}^{238}$  Plutonium for nuclear reactors. The concentration of  $\text{Pu}^{238}$  in an RTG, for instance, is generally about 69% [5].

Table 5 gives the amounts of U234 that would gradually appear in a sample of Pu238 initially equal to 1000 grams.

**Table 5.** Amounts of Plutonium 238 and resulting Uranium 234 from alpha decay.

Years of Decay	grams U234	grams Pu238
0	0	1000
1	7.87	992.13
5	38.75	961.25
10	75.99	924.01
15	111.80	888.20
20	146.21	853.79
30	211.09	788.91
50	326.44	673.56
87.7	500.00	500.00

If one were to simply mix a sample of Plutonium with Uranium, it would be easy to tell that the sample had been altered because natural Uranium doesn't contain Plutonium. However, since Plutonium doesn't combine with Fluorine in the same manner as Uranium (UF<sub>6</sub>), it is conceivable that the Plutonium would not show up once the Uranium was in the form of Uranium Hexafluoride. It is, therefore, possible to simply add older Plutonium 238 to a Uranium sample and get UF<sub>6</sub> with higher concentrations of U<sup>234</sup> without the telltale Plutonium.

One major problem with this possible method would be the decay products from other isotopes of Plutonium. In the aforementioned RTG, the plutonium has significant isotope concentrations of Pu<sup>236</sup>, Pu<sup>239</sup>, Pu<sup>240</sup>, Pu<sup>241</sup>, and Pu<sup>242</sup> in addition to Pu<sup>238</sup>. Many of these isotopes can decay to some isotope of Uranium (Table 6).

**Table 6.** Example isotope concentrations, half-lives, decay products (U<sup>n</sup> Daughter), and amount of daughter U<sup>n</sup> isotopes potentially in product after 10 years from fuel for RTG [5].

Isotope	Concentration (%)	Half-Life (yrs)	U <sup>n</sup> Daughter	U <sup>n</sup> in Product (10yrs)
Pu <sup>236</sup>	.0000011	2.851	U <sup>232</sup>	1*10 <sup>-8</sup>
Pu <sup>238</sup>	69.294	87.7	U <sup>234</sup>	5.5*10 <sup>-3</sup>
Pu <sup>239</sup>	12.230	24,131	U <sup>235</sup>	3.5*10 <sup>-6</sup>
Pu <sup>240</sup>	1.739	6,569	U <sup>236</sup>	1.8*10 <sup>-5</sup>
Pu <sup>241</sup>	.270	14.4	U <sup>237</sup> , U <sup>233</sup>	<1*10 <sup>-3</sup>
Pu <sup>242</sup>	.0955	37,800	U <sup>238</sup>	1.7*10 <sup>-5</sup>

A few things can be determined from these calculations:

- (1) U<sup>234</sup> concentrations in the range of 48.4 to 62.1 ppm can be created by the combination of natural uranium from different mines.
- (2) Enriched/depleted uranium can be combined so that the resultant sample equals the U<sup>235</sup> and U<sup>238</sup> concentrations of natural uranium but has a shifted U<sup>234</sup> concentration. The effect is, however, small and probably not practical.
- (3) Large existing stocks of Pu<sup>238</sup> will produce significant amounts of U<sup>234</sup> and could be added to natural Uranium to produce a higher U<sup>234</sup> concentration. But, the presence of

uranium daughter products from other plutonium isotopes could keep samples from resembling natural uranium.

Sources:

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