

# Computational Analysis of Signatures of Highly Enriched Uranium Produced by Centrifuge and Gaseous Diffusion

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**ABSTRACT.** Different enrichment processes have been used historically to produce highly enriched uranium (HEU) for weapon purposes. The most relevant ones are the gaseous diffusion process and the gas centrifuge. The two exploit different physical principles to separate isotopes of different molecular weight. It could therefore be expected that HEU might carry an isotopic signature that is unique to the enrichment process used to produce the material. Multi-isotope enrichment cascades are generally modeled using the matched-abundance-ratio approach. In this paper, we will present comparisons of the isotopic signatures predicted in gas centrifuge cascades with those predicted in gaseous diffusion cascades by using a modified version of the matched-abundance-ratio cascade code, MSTAR, which accounts for the physical differences in the stage separation factors in the two processes. Additionally, we will present the methodology used by the modified code and discuss representative results for HEU produced from both natural and reprocessed uranium. We find that essentially complete knowledge of the enrichment technologies employed, of the cascade design, and of the mode of operation is required in order to make meaningful (quantitative) statements about expected HEU signatures.

## Introduction

Uranium can be used as fuel for nuclear power reactors and for nuclear weapons. As uranium occurs in nature, it has three isotopes:  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ . The abundance of these isotopes is shown in Table 1. The isotope of interest for producing fission reactions is  $^{235}\text{U}$ , and the uranium must be enriched in that isotope to ~ 3–5% for light-water-cooled power reactors and to ~ 90% for weapons. There are a number of different ways of separating or enriching isotopes, but the two processes used for large-scale enrichment of uranium are gaseous diffusion and gas centrifuge.

Uranium that has been used as fuel in a nuclear reactor, for example, in a plutonium production reactor, can be reprocessed and re-enriched in  $^{235}\text{U}$ , but this irradiated uranium contains additional isotopes, which are produced by the nuclear reactions in the reactor. These isotopes include  $^{232}\text{U}$ ,  $^{233}\text{U}$ , and  $^{236}\text{U}$ , in addition to the  $^{234}\text{U}$  already present. Understanding the unit separation factors for these *minor isotopes* is important in the enrichment of reprocessed uranium.

**Table 1. Isotopic concentrations of natural and Hanford-type irradiated uranium.<sup>1</sup>**

	Natural Uranium	Hanford-type RepU
U-232	-	2.03E-10 at%
U-233	-	3.58E-09 at%
U-234	0.0055 at%	0.0053 at%
U-235	0.7200 at%	0.6010 at%
U-236	-	0.0186 at%

The unit separation in gaseous diffusion is a function of the square root of the ratio of the molecular weights of the components being separated in contrast with the unit separation in a gas centrifuge, where it is a function of the difference of the molecular weights.<sup>2</sup> When analyzing cascades being fed reprocessed uranium for production of low enriched uranium (LEU, less than 20% <sup>235</sup>U), this distinction may be small, but in the case of producing HEU (greater than 20% <sup>235</sup>U), one might reasonably expect the two processes to produce rather different concentrations of the minor isotopes. For designing cascades for enriching multi-component mixtures of uranium, the difference in assays of minor isotopes may be of little importance. However, one might be interested in ascertaining how a particular sample of enriched material was produced. In that case, the minor isotopes might provide *forensic signatures*, which could identify the separation process or ultimately even the origin of the material.

In this paper, we explore how the separation factors of minor isotopes may differ between gaseous diffusion and gas centrifuge, and we present example calculations of enriched LEU and HEU. For the mathematical and numerical analysis, the matched abundance ratio or M\* (read M-star) cascade theory is used. This theory was first suggested by de la Garza.<sup>3,4</sup>

### **Matched-Abundance-Ratio Cascades**

In a cascade separating a binary mixture, the assay of the desired isotope in the up-flowing stream from a stage is matched to the assay of the down-flowing stream from the stage above. This results in a *no-mixing* cascade or *ideal* cascade, which has the desirable feature of minimum inter-stage flow. In a multi-component mixture, the ideal cascade is generalized to a matched-abundance-ratio cascade, which will become an ideal cascade when the mixture is binary.

Following Von Halle,<sup>5</sup> in a multi-component mixture of  $J$  components, let the  $k^{\text{th}}$  component be designated the “key” component, and let the abundance ratio of each component be defined in terms of the key component by

$$R_i = \frac{x_i}{x_k}; i = 1, 2, \dots, J, \quad (1)$$

where  $R_i$  designates the abundance ratio of the  $i^{\text{th}}$  component and  $x_i$  denotes the mole fraction of the  $i^{\text{th}}$  component in the mixture. The overall separation factor for a stage in a cascade is defined as

$$\alpha_i = \frac{R_i'}{R_i''}; i = 1, 2, \dots, J, \quad (2)$$

where the superscript (') denotes a quantity in the up-flowing stream leaving a stage and the superscript (") denotes a quantity in the down-flowing stream leaving a stage. This concept is illustrated in Figure 1, which depicts two adjacent stages in the enriching section of a cascade. In the figure,  $L_n$  is the up-flow from stage number  $n$ . From the definition, the overall stage separation factor for the key component,  $\alpha_k$ , is unity. In gas centrifuge plants, it is common for cascades to be comprised of centrifuges of the same design and each would be operated at feed flow rates producing the same separation factor. In this analysis, the overall stage separation factors are assumed to be constant throughout the cascade. That is, all  $\alpha_i$  are assumed to be independent of the stage number.

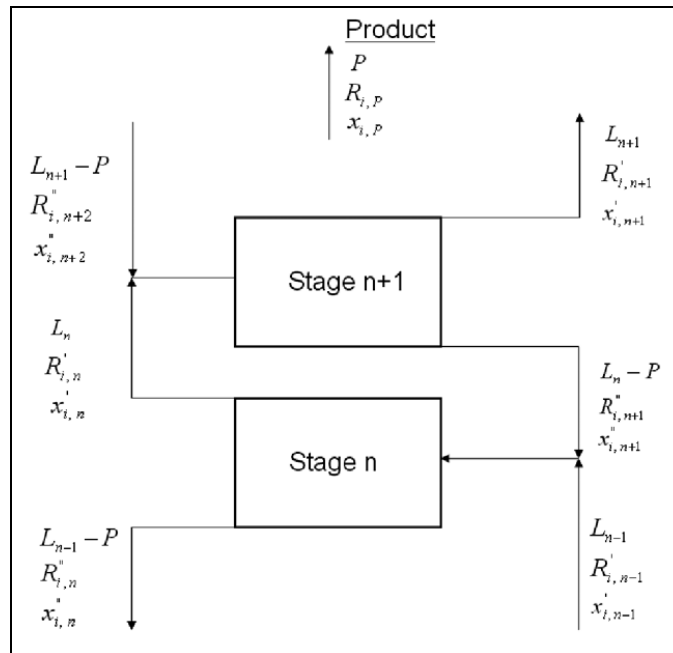


Figure 1. Two adjacent stages in the enriching section of a cascade.

The enriching section of the cascade is comprised of all the stages above the feed point, and the stripping section all stages below the feed point. Material balances are taken about both the enriching and stripping sections to describe their performances. The resulting equations are then used to describe the performance of the overall cascade.

Computer programs, written in Visual Basic, have been developed by Von Halle to solve these M\* cascade equations.<sup>6</sup> The following input is required: (1) the concentrations of all the isotopes in the feed stream, (2) the concentration of <sup>235</sup>U in the both the product and tails stream, (3) either the feed rate or the product rate, and (4) the overall stage separation factor for each isotope. The program then calculates the number of required stages in both the enricher and the stripper, and the interstage flow rates.

In the original version of the M\* program, the overall stage separation factor,  $\alpha$ , for <sup>235</sup>U is given as input, and the stage separation factor per unit mass difference is calculated as

$$\alpha_0 = \alpha^{1/(238-235)}. \quad (3)$$

Then the overall stage separation factor for the  $i^{\text{th}}$  component is calculated as

$$\alpha_i = \alpha_0^{(M_k - M_i)}. \quad (4)$$

For both gaseous diffusion and gas centrifuge, the process gas is UF<sub>6</sub>, and the molecular weights of <sup>235</sup>UF<sub>6</sub> and <sup>238</sup>UF<sub>6</sub> are 349 and 352, respectively. For gaseous diffusion, the overall stage separation factor for <sup>235</sup>U, not including any inefficiencies, is

$$\alpha = \sqrt{352/349} = 1.004289. \quad (5)$$

For a gas centrifuge, the fundamental separation due to the centrifugal force is given by the expression

$$\exp[\Delta M V^2 / 2RT] = \exp\left[\frac{\Delta M}{M} A^2\right], \quad (6)$$

where  $A^2 = M V^2 / 2RT$  is the stratification parameter,  $M$  is molecular weight,  $T$  is absolute temperature,  $\Delta M$  is the difference in molecular weights of the species, and  $R$  is the universal gas constant (8314.4 J/(kg · mole · K)). For the hypothetical “Iguaçu” centrifuge,<sup>7</sup> the peripheral speed is 600 m/s and  $T = 300$  K, Equation (6) yields

$A^2 = 25.4$  for UF<sub>6</sub>. The overall separation factor of a gas centrifuge is determined not only by Equation (6) but by the feed rate, length of the centrifuge, and other parameters of the countercurrent flow in the centrifuge. Typical centrifuges reported in the literature have overall separation factors on the order of 1.6, considerably larger than gaseous diffusion.

However, if these two values of overall separation factor are used in the classical M\* code, the product and tails concentrations of all the isotopes are exactly the same. The only difference is in the number of stages required in the two cascades. This result is due to the calculation of the overall stage separation factor for the minor isotopes through Equation (4). Therefore, the M\* code was modified so the user can prescribe all  $\alpha_i$  in a manner consistent with the separation process being modeled.

### Determination of Separation Factors for Gas Centrifuges

Enrichment of spent reactor fuel by gas centrifuge has been reported in which the Iquaçu centrifuge parameters were used.<sup>8</sup> In that study, a single gas centrifuge was numerically optimized for enrichment of <sup>235</sup>U from a binary mixture of natural uranium. Then, numerical simulations were performed with spent reactor fuel as feed material. The separation factors were computed as functions of feed rate where the concentration of <sup>235</sup>U was maximized with respect to the gradient of the temperature on the rotor wall and drag power of the tails removal scoop. Using the concentrations of the isotopes calculated in the withdrawals streams, the following results were obtained:

$$\frac{\alpha_{232} - 1}{\alpha_{235} - 1} = 1.97, \quad \frac{\alpha_{234} - 1}{\alpha_{235} - 1} = 1.32, \quad \frac{\alpha_{236} - 1}{\alpha_{235} - 1} = 0.66. \quad (7)$$

These ratios were found to vary by less than 1% as the feed rate was varied from 1 to 100 mg/s.

Expanding the right-hand side of Equation (6) in a Taylor series keeping only the first two terms yields

$$\alpha_i - 1 = \left\{ 1 + \frac{\Delta M_i}{M} A^2 \right\} - 1 = \frac{\Delta M_i}{M} A^2, \quad (8)$$

where  $\Delta M_i$  is the difference in molecular weight between isotope  $i$  and the key isotope, in this case <sup>238</sup>U.

Use Equation (8) for the isotopes <sup>232</sup>U, <sup>234</sup>U, <sup>235</sup>U, <sup>236</sup>U, <sup>238</sup>U, so  $J = 5$ , and take the key component to be <sup>238</sup>U, with  $k = 5$ :

$$\frac{\alpha_i - 1}{\alpha_{235} - 1} = \frac{M_i - M_k}{M_{235} - M_k} = \frac{\Delta M_i}{3}, \quad i = 1, 2, \dots, 5. \quad (9)$$

More terms could be taken in the Taylor series, but this equation predicts values of 2, 4/3, and 2/3, which compares very well with the results of the numerical study presented in Equation (7). If the separation factor for <sup>235</sup>U is given, the separation factors for the other components can be determined from this equation as follows:

$$\alpha_i = 1 + \frac{\Delta M_i}{3} (\alpha_{235} - 1), \quad i = 1, 2, \dots, 6. \quad (10)$$

So we see the separation factors are proportional to mass differences, which is consistent with results reported elsewhere.<sup>9</sup> Equation (10) will be used to simulate enrichment by gas centrifuge.

### Determination of Separation Factors for Gaseous Diffusion

For gaseous diffusion, we use Equation (5) to determine the separation factors for all the isotopes:

$$\alpha_i = \sqrt{M_k/M_i}, i = 1, \dots, J, \quad (11)$$

where  $M_i$  is the molecular weight of each isotope and  $M_k$  is the molecular weight of the key component. For  $^{238}\text{UF}_6$ ,  $M_k = 352$ . Equations (4) and (11) yield almost exactly the same values for  $\alpha_i$ , and either may be used to simulate enrichment by gaseous diffusion.

### Results of Calculations

We performed M\* calculations to simulate enriching natural uranium and reprocessed uranium to 93%  $^{235}\text{U}$  by both gaseous diffusion and gas centrifuge as described earlier. The concentrations of the feed material for the two cases are given in Table 1. In all calculations, the  $^{235}\text{U}$  tails concentration was 0.3%. For the gas centrifuge calculations, we have chosen  $\alpha_{235} = 1.60$ , and we have used Equation (10) for the separation factors for the other isotopes. This gives results different from traditional M\* calculations, which used Equation (4). The net effect is shown in Figure 2 for the isotope  $^{232}\text{U}$  enriched in a centrifuge cascade using the traditional and the modified M\* code. Counterintuitively, the  $^{232}\text{U}$  concentration at first increases *faster* for the *lower* separation factor because it is closer to the value the cascade is optimized for (1.60 for  $^{235}\text{U}$ ). This trend reverses only in the final stages of the cascade, when the  $^{235}\text{U}$  enrichment is already about 90%. In this example, the final concentration of  $^{232}\text{U}$  is about 10% lower for the modified M\* calculation, which uses the smaller separation factor (2.20 vs. 2.56) for this isotope. Note that about 80% of the total  $^{232}\text{U}$  present in the feed is extracted in a cascade designed production of weapon-grade uranium, compared to only about 50% of the initial  $^{235}\text{U}$  that leaves the cascade in the product.

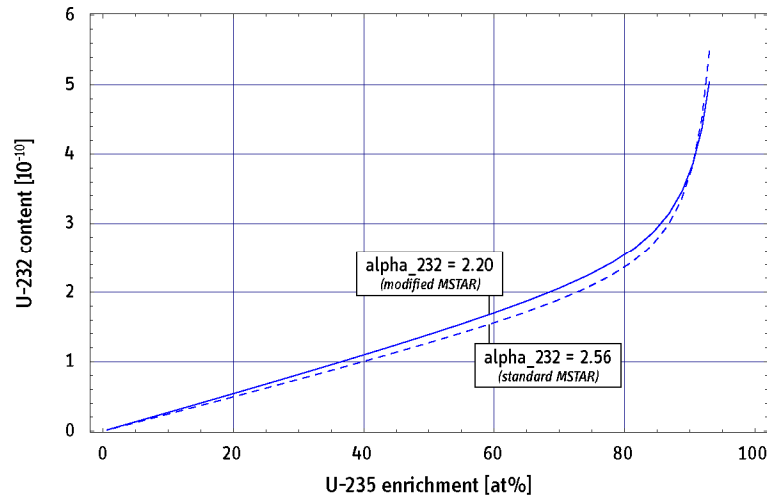


Figure 2. U-232 vs U-235 in a centrifuge cascade using the traditional and the modified M\* code.

Similarly, Figure 3 shows the concentration of several uranium isotopes relative to the final concentration of the respective isotope in the product stream of the centrifuge cascade as a function of the stage number in the enriching section. These simulations, carried out with the modified M\* code, predict that the relative increase of concentration per stage behaves quite differently for the various isotopes. Again, since the cascade is

optimized for  $^{235}\text{U}$  enrichment, the concentration of other isotopes increases less effectively in the lower stages of the enricher. Only once the efficiency of  $^{235}\text{U}$  enrichment, i.e. or  $^{238}\text{U}$  depletion, drops in the final stages of the cascade, the enrichment of the lighter isotopes becomes much more efficient.

In Table 2, the product concentrations of weapon-grade uranium (93%  $^{235}\text{U}$ ) enriched by gas centrifuge and gaseous diffusion are presented. In the case of natural-uranium feed, the difference for  $^{234}\text{U}$  concentration between the two methods is shown to be on the order of 5%. In the case of irradiated uranium from a Hanford-reactor-type production reactor, the differences for the minor isotopes concentrations range from 5% to more than 20%. The difference for the lighter isotopes increases with decreasing molecular weight and are all much smaller than the difference for the heavier isotope  $^{236}\text{U}$ , which features the most significant signature in our simulations.

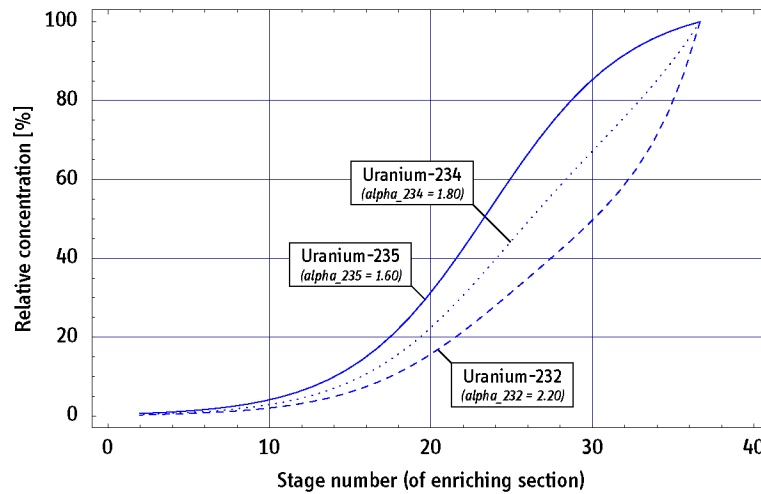


Figure 3. Relative concentration of various uranium isotopes as a function of stage number.

Table 2. Product concentrations in gas centrifuge and gaseous diffusion cascades producing HEU from natural uranium (top) and reprocessed Hanford-reactor-type uranium (bottom).

Isotope	Gas Centrifuge	Gaseous Diffusion	Difference
U-234	0.890 at%	0.934 at%	-4.7%
U-235	93.0 at%	93.0 at%	-
U-232	5.04E-08 at%	5.49E-08 at%	-8.14%
U-233	8.20E-07 at%	8.88E-07 at%	-7.65%
U-234	1.058 at%	1.121 at%	-5.68%
U-235	93.0 at%	93.0 at%	-
U-236	1.524 at%	1.250 at%	+21.87%

## Conclusions

We have extended the functionality of the cascade code MSTAR, which simulates multi-isotope enrichment cascades using the matched-abundance-ratio technique. In the modified code, the separation factors for all isotopes can be specified independently, allowing the user to determine the impact of different enrichment processes on the abundance of the minor uranium isotopes. We have proposed expressions to determine sets of separation factors for the gas centrifuge and the gaseous diffusion process and used those with MSTAR to determine isotopic signatures for weapons-grade uranium produced from natural and irradiated uranium. The most significant signature is obtained for the  $^{236}\text{U}$  concentration in weapons-grade uranium produced from reprocessed uranium. Our simulations predict that the  $^{236}\text{U}$  content should be about 20% higher for material enriched with centrifuge technology, which *could* be a relevant indicator for an assessment of the potential origin of the sample. The results of our simulations should be benchmarked against (or validated with) experimental data, which would however require knowledge of all relevant sample properties and process characteristics that were used at the time of production.

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<sup>1</sup> Adapted from “Signatures of Plutonium and Uranium,” Appendix F in *Nuclear Forensics: Role, State of the Art, Program Needs*, report by the Joint Working Group of the American Physical Society (APS) Panel on Public Affairs and the American Association for the Advancement of Science (AAAS), Center for Science, Technology and Security Policy, Washington, D.C., February 2008, Table F.2.

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