Reactor physics analysis of the effects of U-236 poisoning on the use of reprocessed uranium in PWR fuel

Final Report

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Abstract

Under some conditions it is economically attractive or environmentally preferable to reprocess spent fuel in order to recover U^{235}. However, unlike natural uranium, recovered uranium (RU) contains U^{236}, the presence of which decreases the reactivity of fresh fuel and needs to be compensated for. This effect is quantitatively described by the penalty factor: the number of units of U^{235} that need to be added to compensate for the presence of U^{236}, per unit of U^{236} in the fuel. This project examines the penalty factor, natural uranium feed requirement, and separative work requirement for two different multi-pass reprocessing scenarios and compares them to a once-through cycle, as appropriate. In the first scenario, RU is added to a natural uranium feed and re-enriched to produce one unit of fresh fuel per unit RU feed; in the second scenario, no natural uranium feed is added.

Compared to a once-through cycle, the more realistic reprocessing scenario decreased the natural uranium feed requirement for making fuel for the second pass by 8%; the savings decreased to 6% for subsequent passes. These savings come at the price of an increased separative work requirement that ranged from 3.7% for the second pass to 6.7% for the seventh pass. The penalty factor itself was found to decrease with increasing pass number for both scenarios, with values ranging from 0.272 to 0.235 for the more realistic scenario. A four-factor analysis suggests that this downward trend is the result of an energy self-shielding effect from the large 5.48 eV absorption resonance in U^{236}.

Introduction

Closing the nuclear fuel cycle has indisputable advantages: it increases utilization of resources by allowing reuse of valuable nuclear fuel materials and makes a significant contribution to environmental protection by reducing waste volumes and allowing waste to be processed into safer final forms for disposal. Closing the cycle would certainly be advisable if its drawbacks, such as increased proliferation risk, high costs, etc., can be shown to be acceptable.

While that economic analysis is outside the scope of NE 406, this project considers the technical details of one aspect of the closed fuel cycle: the use of reprocessed uranium in PWR fuel. Obviously, reprocessed uranium will differ significantly from natural uranium, which contains only three isotopes: U^{234}, U^{235}, and U^{238}. During irradiation, several new radioactive isotopes form: U^{232}, U^{233}, U^{236}, and U^{237}. From a reactor physics perspective, the principal difference between fuel made with reprocessed uranium and fuel made with natural uranium is the presence of the U^{236} isotope formed by neutron capture in U^{235}. Renier et al. point out that "[b]ecause of the nonfission capture cross-section of U^{236}, fuel at a given U^{235} enrichment has a slightly smaller reactivity with U^{236} present than without. The reactivity loss persists late in the life of the fuel, so the initial enrichment will have to be slightly higher to compensate for the U^{236}" [1, p. 94].

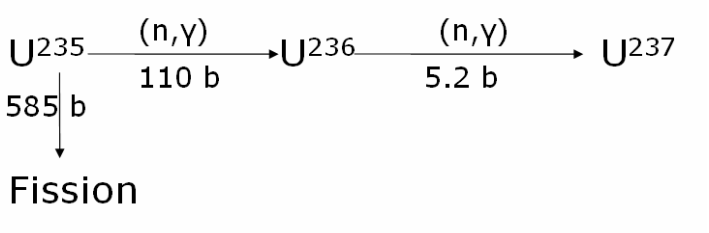


Figure 1: Relevant cross-sections for in-core U^{235} and U^{236} reactions

This additional U^{235} enrichment will require additional separative work, a problem compounded by the fact that the U^{236} in the feed will become enriched as well. One can thus define the U^{236} penalty factor as the amount of U^{235} that needs to be added to overcome the effects of U^{236} and achieve the same discharge burnup, per unit of U^{236}:

$$penalty\_factor = \frac{\Delta massU^{235}}{massU^{236}} \qquad (1)$$

Problem Description

In this project, the penalty factor as well as the separative work and natural uranium feedstock requirements for two different reprocessing scenarios were calculated as a function of pass number and compared to the corresponding values for a once-through cycle, as appropriate. The first scenario is the one used by Renier et al., wherein "the recovered uranium (RU) from a given amount of spent fuel is fed along with sufficient natural uranium to produce the same amount of fresh fuel for the next reactor pass" [1, p. 94]. (Note that this scenario allows as much natural uranium as necessary to be added to the enrichment feed.) As an engineering approximation of this enrichment process, the re-enriched weight fraction of U^{236} was set at 60% of the discharge U^{236} weight percentage from the previous reactor pass, in accordance with Renier et al.'s observation that "about 60% of U^{236} remains in the re-enriched product" [1, p. 94][1]. To more explicitly model the three-component enrichment process, this simplified approximation was then replaced with Benedict et al.'s extension of de la Garza's "matched R cascade" model [2, 3]. The second scenario, which was also analyzed using the matched R cascade model,

[1] Our implementation of this approximation ignores the differences in the weights of the U^{235}, U^{236}, and U^{238}.

represents the other extreme in which no natural uranium is added to the enrichment feed; therefore, the feed is composed entirely of recovered uranium from the previous reactor pass.

Procedure

A three-batch core reloading scheme was used for each pass. In accordance with the linear reactivity model (LRM) discussed in lab 3, the discharge burnup, B_d, was achieved by seeking a critical burnup, B_c, such that $B_c = 2/3 * B_d$. A 4% leakage effect was assumed, so the critical burnup was defined as the burnup at which the reactivity of the infinite core was 0.04. The discharge uranium isotopics were then used as the enrichment feed for the next pass, with (first scenario) and without (second scenario) adding natural uranium to that feed. No chemical shim or control rods were used.

To match Renier et al.'s analysis, the first pass in both scenarios was a low-burnup pass (33,000 MWd/t) representative of legacy spent fuel, followed by some number, N, of high-burnup passes (55,000 MWd/t). N varied by scenario depending on the feasibility of the scenario for high N. For each pass, the U^{235} enrichment necessary to achieve the desired discharge burnup needed to be determined by varying Δ, the weight fraction of U^{235} that needed to be added to overcome the effects of U^{236} poisoning. The nominal enrichment value of 4.38 w/o is the enrichment necessary to achieve 55,000 MWd/t burnup with U^{236}-less fuel.

Table 1: Details of multi-pass scheme for both reprocessing scenarios

Pass number	Number of batches in refueling scheme	Discharge burnup, B_d [MWd/t]	Initial enrichment [w/o U^{235}]
1	3	33,000	2.9
2	3	55,000	$4.38 + \Delta_2$
…	…	…	…
N	3	55,000	$4.38 + \Delta_N$

Reactor physics model

Materials and data

The fuel was composed of stoichiometric UO_2 with 10 g/cm^3 density. The U^{235} enrichment and the U^{236} content varied for each pass. The moderator was light water with 1 g/cm^3 density. The clad was Zirconium with 6.5 g/cm^3 density. No control rods, chemical shim, or burnable poison were introduced to the system. The reactor parameters are summarized in Table 2.

Table 2: Reactor core operating parameters

Moderator density	1 g/cm^3
Fuel density	10 g/cm^3
Clad density	6.5 g/cm^3
Moderator temperature	550 K
Fuel temperature	925 K
Fuel diameter	0.5 cm
U-235 enrichment	Varied
Boron concentration	0 a/o
Control rods	Out
Burnable poison	None

Geometry

A three-batch core was assumed in this project. A structure representing one eighth of a 15x15 PWR assembly (see Figure 2) was used to simulate this core. Reflecting boundaries were used to simulate an infinite structure constructed by repeating this unit.

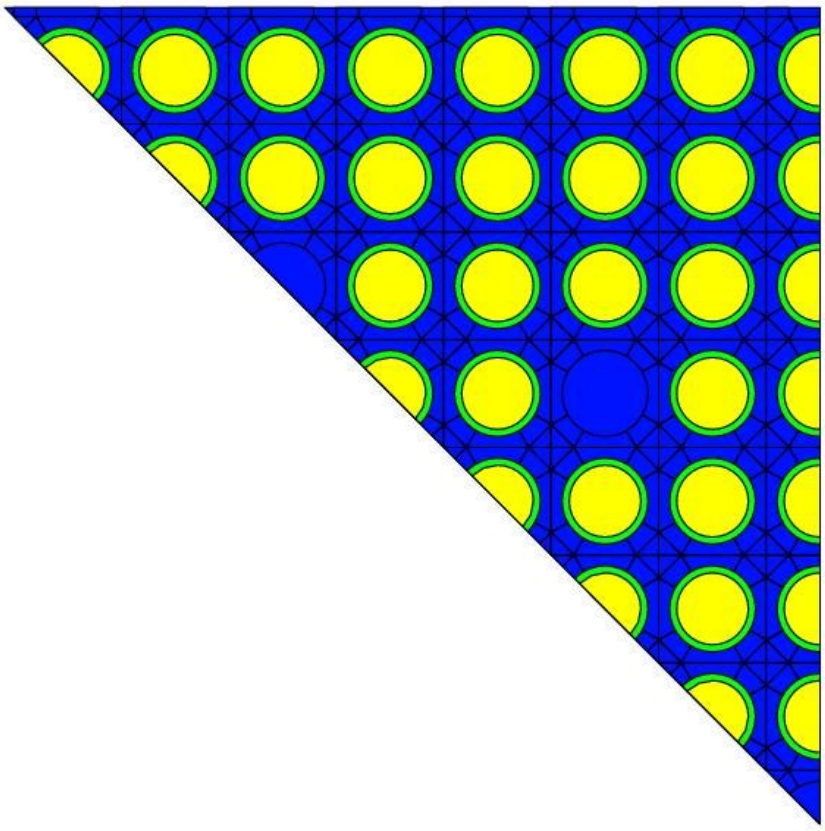


Figure 2: 1/8 of a reactor core assembly

In Figure 2, the fuel is represented by the yellow circles, the clad is the surrounding green rings, and blue area represents the moderator.

Reprocessing and enrichment model

The matched R cascade enrichment model for multi-component uranium mixtures enforces four constraints: the conservation of mass for U^{235}, U^{236}, and total uranium (Equations 2-4) as well as a cascade constraint derived from requiring that the weight ratio of U^{235} and U^{238} be equal whenever two streams are mixed in the cascade (Equation 5).[2] Benedict et al. show that these constraints lead to a coupled system of four equations in four unknowns [2]. The notation for this system is given in Table 3, followed by the equations themselves.

Table 3: Notation for matched R cascade equations

Material stream	Flow rate	U-235 weight fraction	U-236 weight fraction	U-235 to U-238 weight ratio
Product	P	y_{5p}	y_{6p}	R_p
Feed	F	z_{5f}	z_{6f}	R_f
Tails	W	x_{5w}	x_{6w}	R_w

$$F = \frac{y_{5p} - x_{5w}}{z_{5f} - x_{5w}} P \qquad (2)$$

$$W = \frac{y_{5p} - z_{5f}}{z_{5f} - x_{5w}} P \qquad (3)$$

$$Fz_{6f} = Py_{6p} + Wx_{6w} \qquad (4)$$

$$\frac{Fz_{6f}}{R_z^{1/3}} = \frac{Py_{6p}}{R_p^{1/3}} + \frac{Wx_{6w}}{R_w^{1/3}} \qquad (5)$$

R_p, R_w, and R_z are the U^{235} to U^{238} weight ratios in the product, tails, and feed, respectively. The separative work required for the matched R cascade can be calculated with Equation 6:

$$SepWork = P(2y_{5p} + 4y_{6p} - 1)\ln R_p + W(2x_{5w} + 4x_{6w} - 1)\ln R_w - F(2z_{5f} + 4z_{6f} - 1)\ln R_f \qquad (6)$$

[2] Apparently this constraint "minimizes total internal flow" in the cascade [2, p. 695].

Thus, for some arbitrary product flow rate P, if one assumes a U^{235} tails fraction x_{5w} and specifies the U^{235} product fraction y_{5p} and feed fractions z_{5f} and z_{6f}, then the system is entirely determined. The U^{236} fraction in the product and tails (y_{6p} and x_{6p}, respectively) can then be solved for and used to specify the material definition for the next pass in the reactor physics model. This was the process used for the second reprocessing scenario, with the feed fractions set by the discharge isotopics of the previous pass.

The matched R cascade enrichment model was used for both reprocessing schemes. However, modeling the first reprocessing scenario, the one used by Renier et al., requires decomposing F into F_R and F_N, where F_R is the mass flow rate of recovered uranium from the previous reactor pass and F_N is the mass flow rate of natural uranium. Natural uranium is added to the feed at whatever rate is required to meet Renier et al.'s condition: $F_R = P$. This adds one variable (F_N) and one constraint ($F_R = P$) to the above system of equations, so the system should again be fully determined by the specification of an arbitrary product flow rate, a U^{235} tails fraction, a desired product U^{235} enrichment, and the U^{235} and U^{236} fractions in the last reactor pass's discharge vector. Thus, solving the new system will again give product and tails fractions for U^{236}, in addition to the required flow rate of natural uranium.

Results

Excess uranium requirement

Figure 3 plots the additional U^{235} weight fraction required to compensate for U^{236}, as a function of the pass number, for the first scenario using the engineering approximation of setting the U^{236} weight fraction at 60% of its discharge value.

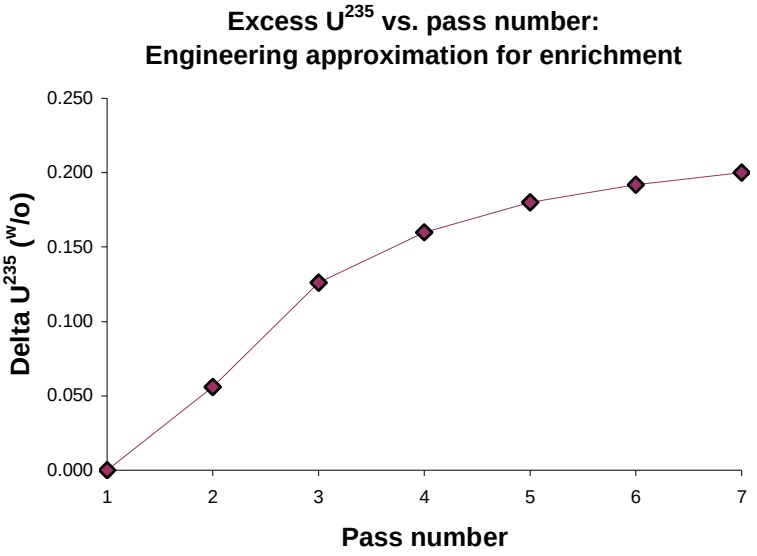
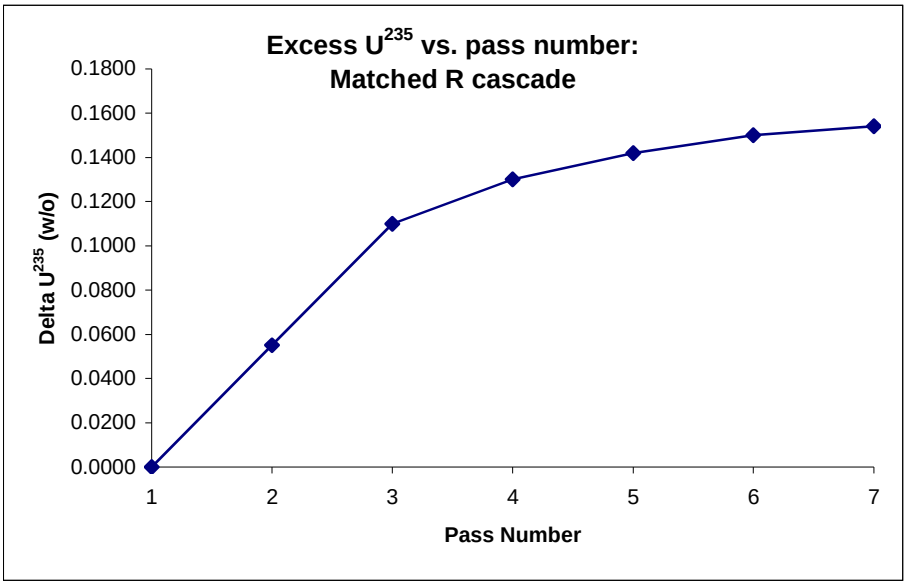
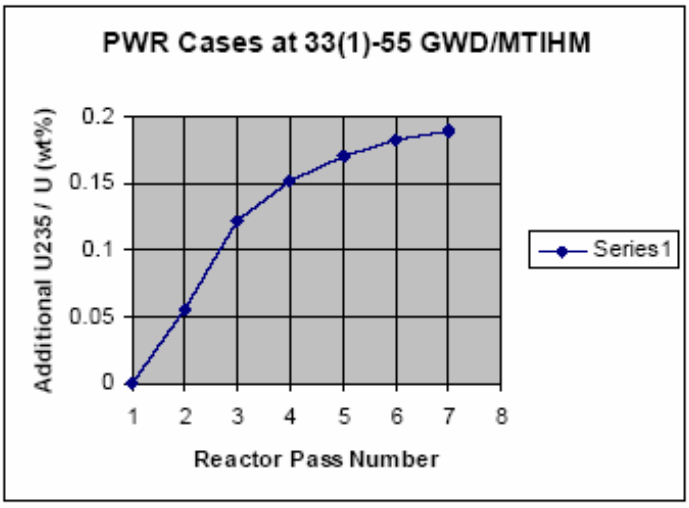


Figure 3: The extra U^{235} needed to compensate for the presence of U^{236} in the first scenario, under the engineering approximation of the re-enrichment process

When the matched R cascade enrichment model was used, the results (Figure 4a) showed that the simplified approximation was causing an overestimate of the amount of U^{235} required to compensate for U^{236}. However, the same trend overall trend seems to hold and to match Renier et al.'s trend (Figure 4b):



(a)



(b)

Figure 4: The extra U^{235} needed to compensate for the presence of U^{236} in the first reprocessing scenario (a) Results of this study under the matched R cascade model (b) Renier et al.'s result (image from [1])

The results of the second scenario, which represents the extreme case of reprocessing using only spent fuel as the re-enrichment feed, showed that this scenario is less practical. The amount of U^{236} increased very rapidly in the beginning until most of the fuel became U^{236} on the fourth pass. Figure 5 plots the weight percentages of the three uranium components U^{235}, U^{236}, and U^{238} against pass number.

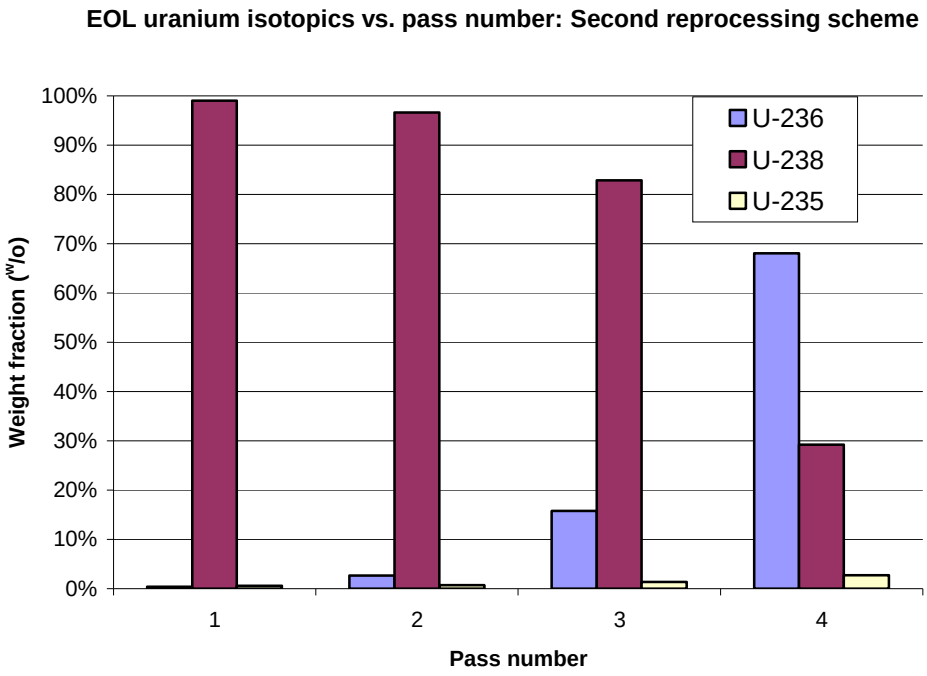


Figure 5: The uranium isotopic vector after burning the fresh fuel (pass 1) and after each pass of burning fuel made with reprocessed uranium for the reprocessing scheme using no natural feed during re-enrichment

To compensate for this rapid increase in U^{236} content, the amount of excess U^{235} required in order to achieve the desired burnup also increased rapidly in the second scenario. Figure 6 shows that a value of Δ = 3.9 w/o U^{235} was reached by the fourth pass; this value is a factor of more than 20 times the delta value for the seventh pass under the first scenario.

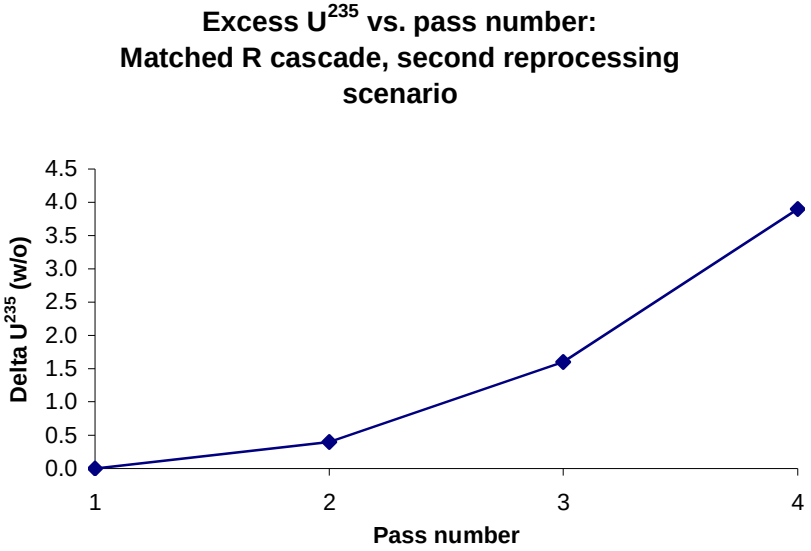


Figure 6: The extra U^{235} needed to compensate for the presence of U^{236} in the second scenario with no natural uranium feed, under the matched R cascade model of the re-enrichment process

Natural uranium feed requirement

Recovering U^{235} from spent fuel in both reprocessing scenarios resulted in an overall savings in the amount of natural uranium needed for the re-enrichment process. While this result is trivial for the second reprocessing scenario (which doesn't use any natural uranium feed after the first pass), the natural uranium savings in the more realistic reprocessing scenario is an important thing to consider, since it represents one of the chief benefits of reprocessing. In this study, the amount of natural uranium required to feed the enrichment process was 8% lower than for the once-through cycle for the second pass, and then stabilized at 6% for passes 3 through 7. Figure 7 plots the natural uranium feed required for the first and second reprocessing scenarios ("Feed = Spent + Nat" and "Feed = Spent," respectively), and for the once-through cycle ("Feed = Nat").

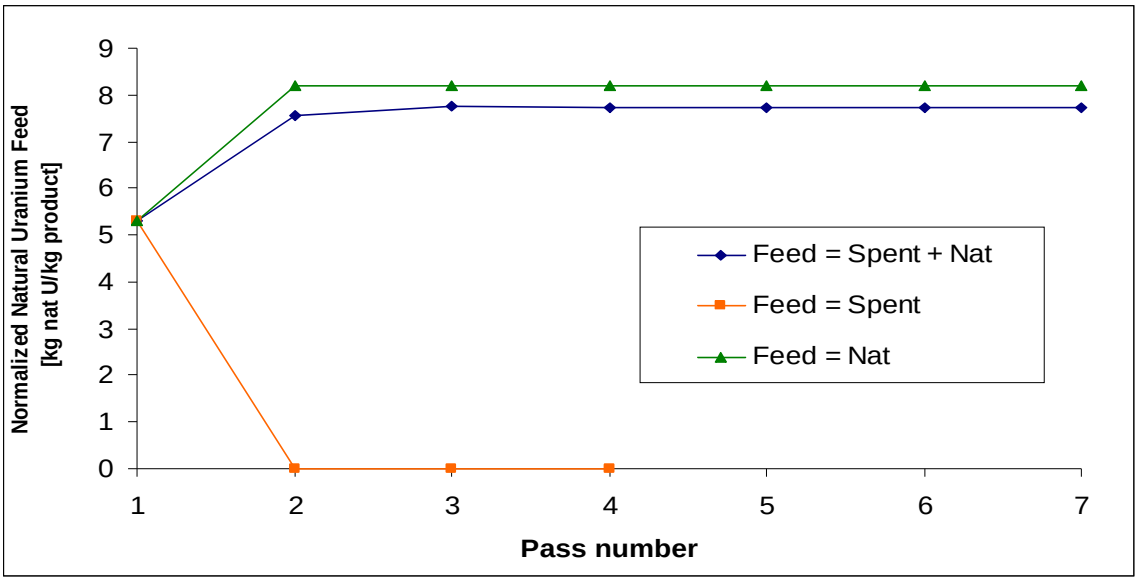


Figure 7: Natural uranium required in the enrichment process per unit of enriched product for the two scenarios as well as the once through cycle

Separative work requirement

The separative work needed for enrichment when RU is added to natural uranium (first scenario) is higher than that of enriching only natural uranium due to the U^{236} penalty *and* because the U^{236} itself gets enriched in the process. Figure 8 shows that the separative work requirement is 3.6% higher for the second pass of the first reprocessing scenario than for the once-through cycle. This difference keeps increasing with the pass number (as U^{236} continues to accumulate) and reaches 6.7% at the seventh pass. The separative work requirement when only spent fuel was used as the enrichment feed increased rapidly and reached about double the separative work of the once-through cycle by the fourth pass.

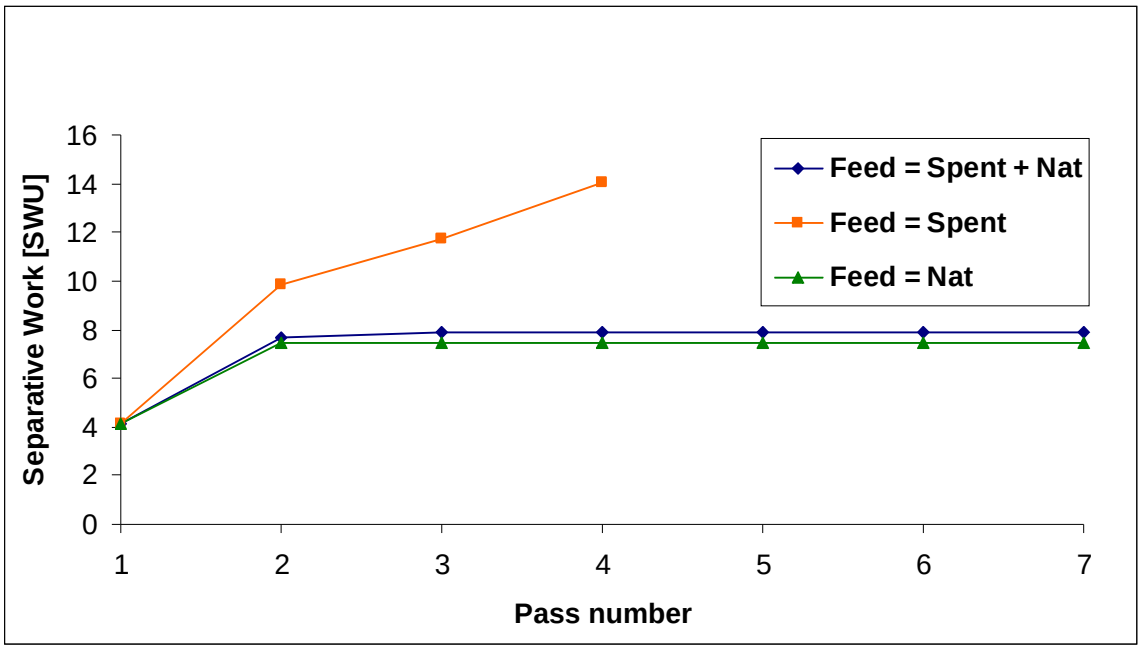


Figure 8: The separative work requirement for enrichment for both reprocessing scenarios as well as the once-through cycle

Penalty factor

The penalty factor was found to decrease with the number of passes, i.e., with the U^{236} content. Figure 9 shows the penalty factor as a function of pass number for both scenarios. It decreases rapidly for the second scenario because of the rapid change in U^{236} content for that scenario (see Figure 5).

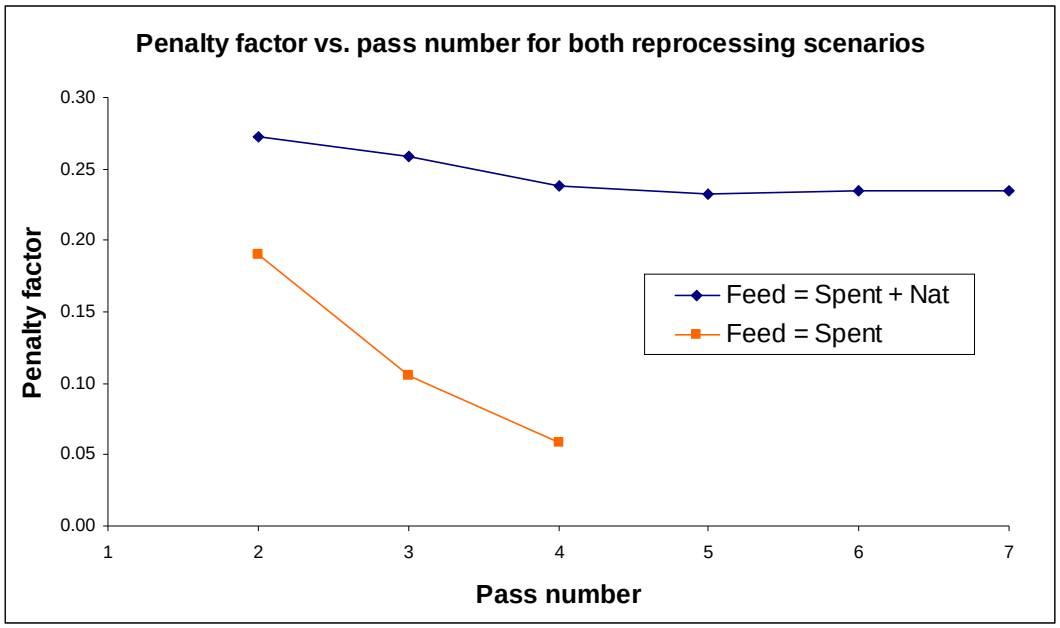


Figure 9: The penalty factor as a function of the pass number for both scenarios. Note the decrease in penalty factor with pass number (see below for explanation).

To probe the physics behind this behavior, the multiplication factor and each term in its four-factor decomposition (normalized by their values without U^{236}) were plotted against increasing U^{236} content in Figure 10. For this simplified analysis intended to illustrate just the effects of U^{236}, only BOL values were considered, and no U^{235} was added (U^{236} simply replaced U^{238} in the fuel).

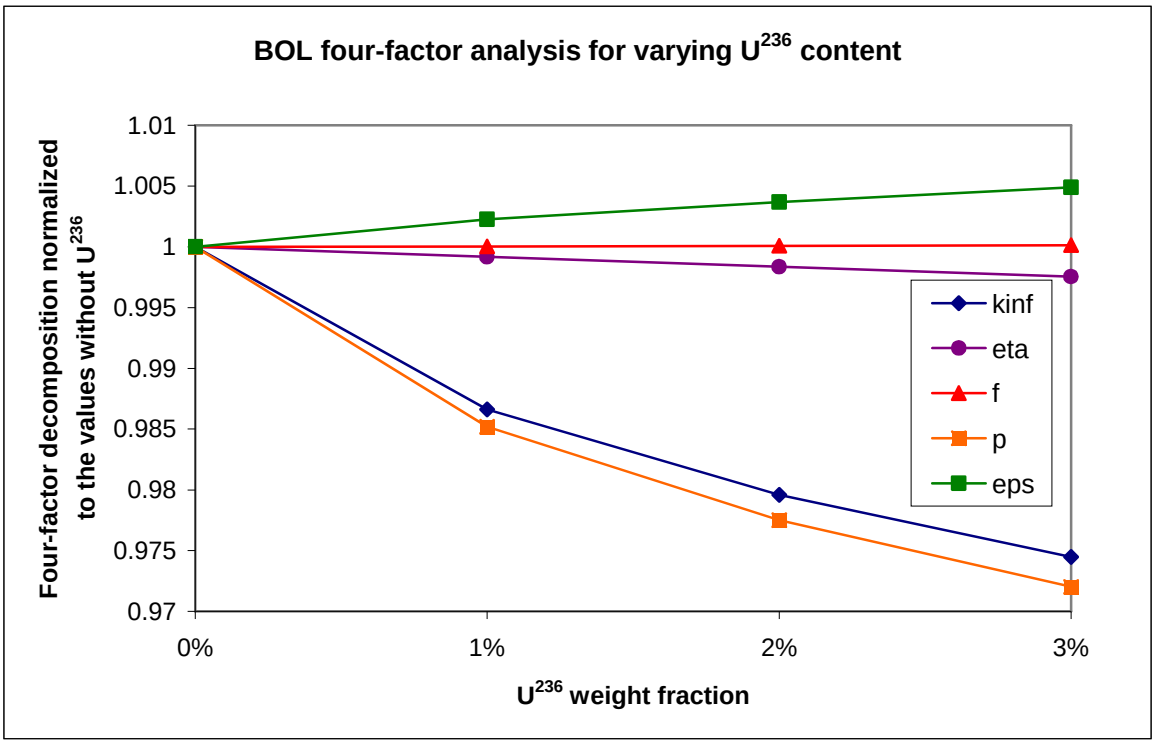


Figure 10: The four factors normalized to their values without U^{236}, as a function of the U^{236} content (no U^{235} compensation)

The behavior of each of these terms is fairly easy to understand. The thermal absorption cross-section of U^{236} is slightly higher than that of the U^{238}, which explains the slight increase of the thermal utilization term and decrease of the regeneration factor term with increasing U^{236} content. The slight increase of the fast fission factor term with U^{236} content can be explained by comparing the fission cross-section of U^{236} with that of U^{238}.

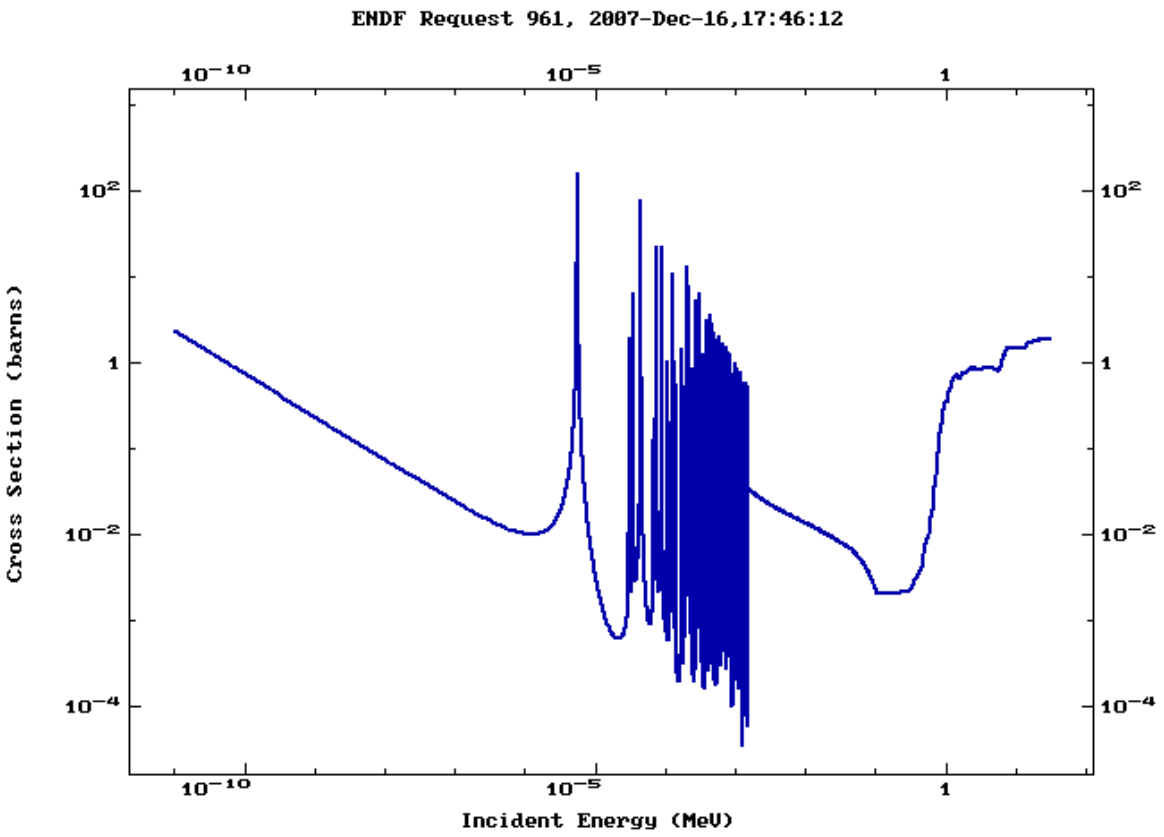


Figure 11: U^{236} fission cross-section (image from [4])

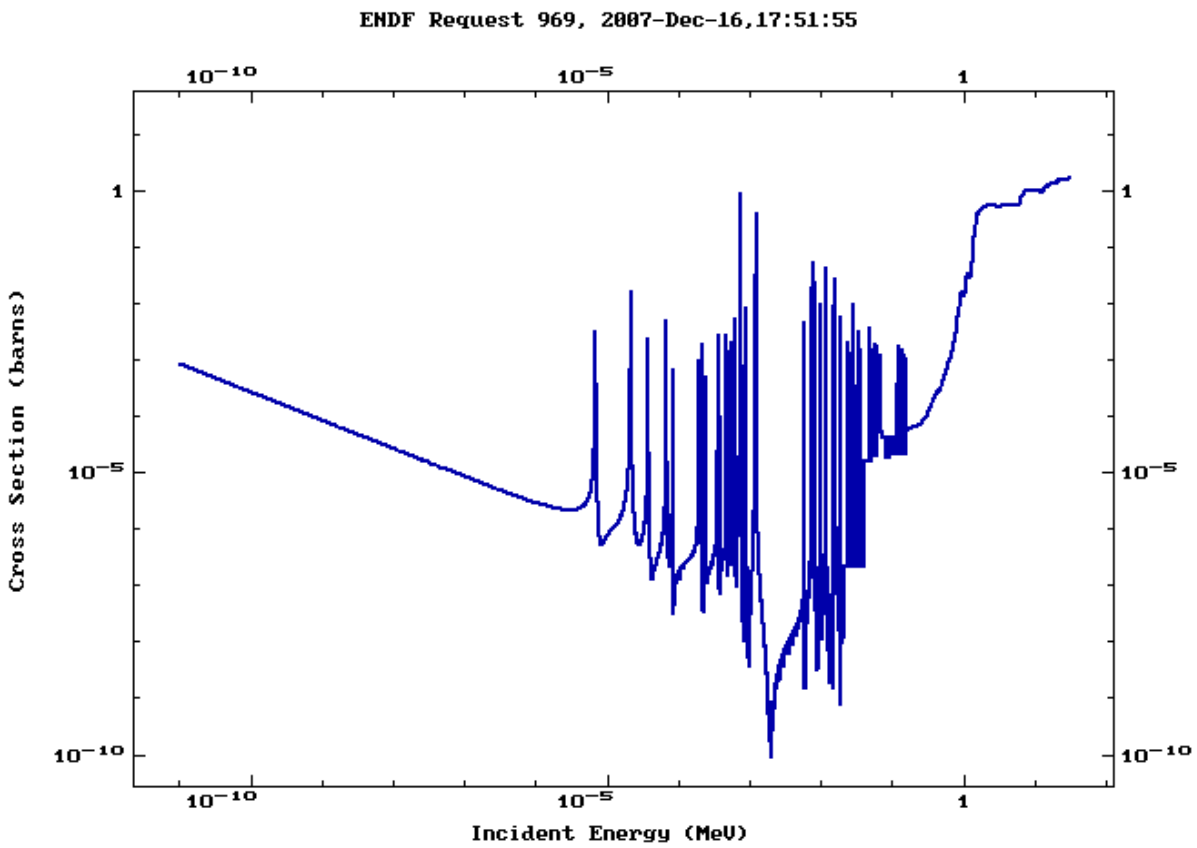


Figure 12: U^{238} fission cross-section (image from [4])

Note that the fission cross-section of U^{236} (Figure 11) is slightly higher than that of U^{238} (Figure 12) at high energies. A very strong U^{236} resonance at 5.48 eV (see further discussion below) exists in the fission cross-section as well[3]. Both these effects contribute to the increase in the fast fission factor term with increasing U^{236} content.

The most important observation about Figure 10, of course, is that the multiplication factor follows the dominant resonance escape probability term's decrease with increasing U^{236} content. The magnitude of the slope of that term's curve decreases by about 28% in the region between 2% and 3% U^{236} content from the slope in the region between 1% and 2%. This behavior can be

[3] Thus, we can conclude that the fission resonance partial width , Γ_f, for this resonance must be appreciable, though a published value is hard to find. Comparing Figure 11 and 13, we can guess that Γ_f is less than the radiative capture partial width, $\Gamma\gamma$, for which Baumann et al. report a value of 32.5 mV [5].

explained by comparing the U^{236} parasitic (n,γ) absorption cross-section to that of U^{238} (Figures 13 and 14).

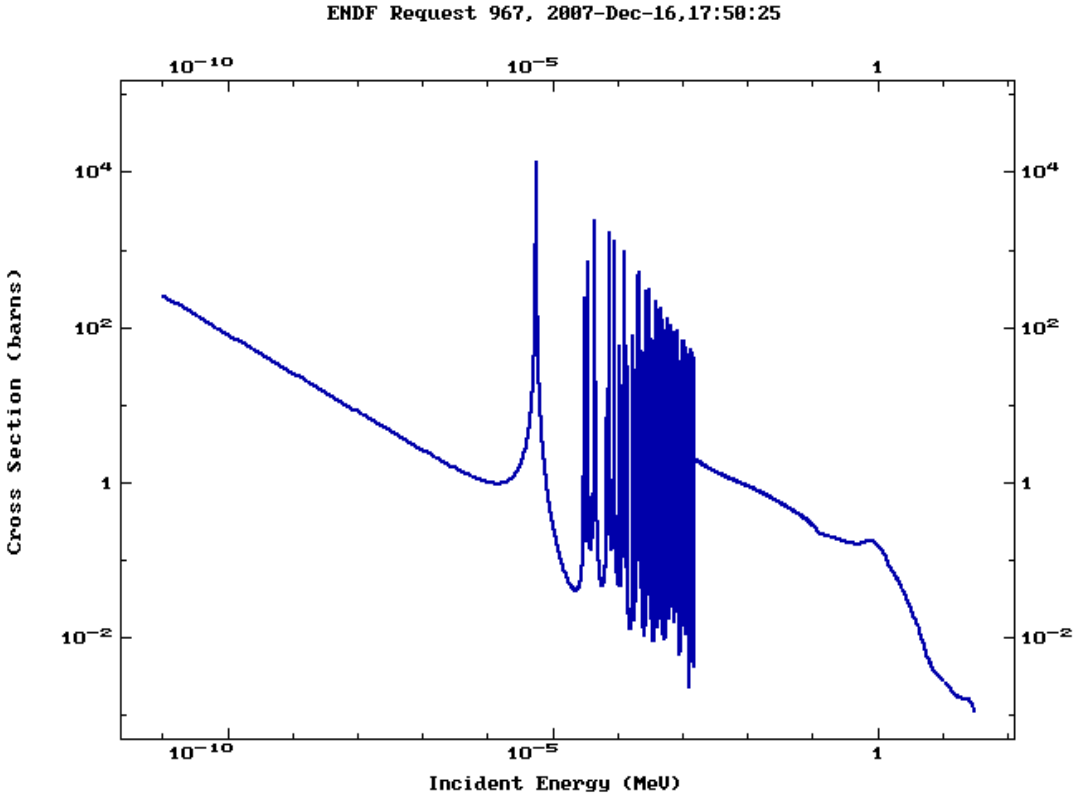


Figure 13: (n,γ) cross-section of U^{236} (image from [4])

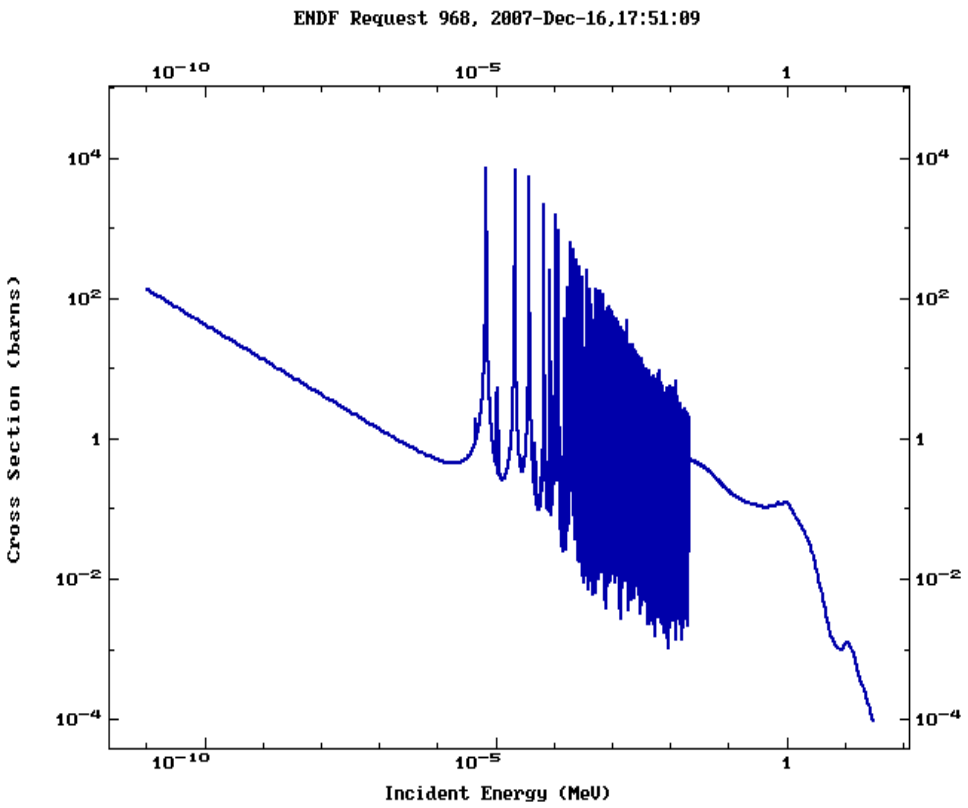


Figure 14: (n,γ) cross-section of U^{238} (image from [4])

Figure 13 shows that U^{236} has a strong resonance at 5.48 eV with a maximum value greater than 10^4 b. The decrease in the slope of the resonance escape probability term in Figure 10 is an energy self-shielding effect; the 5.48 eV resonance becomes less important as more and more U^{236} is added because the resonance depresses the neutron flux at the resonance energy, making subsequent absorptions at that energy less likely. *Thus, one expects a gradual decrease in the poisoning effects of U^{236} on a per mass U^{236} basis.* This explains why the penalty factor decreases with increasing pass number.

Conclusion

As expected, using RU in PWR fuel reduces the natural uranium feed requirement for both reprocessing scenarios considered in this study but increases the separative work requirement. While the "no natural feed" scenario is fairly unrealistic, the scenario Renier et al. suggest yields a 6-8% feed savings for a 3-7% increase in the separative work requirement, depending on the number of passes being used. An economic analysis of this data would allow fuel cycle researchers to identify a price of natural uranium for which reprocessing becomes economically viable for some given cost of separative work.

More interesting to the reactor physicist is that the penalty factor was found to decrease with increasing pass number. For the realistic reprocessing scenario, it ranged from 0.272 for the second pass to 0.235 for the seventh pass. A four-factor analysis showed that this behavior is dominated by the resonance escape probability term. U^{236}'s 5.48 eV absorption resonance is most responsible for the poisoning effect of U^{236}, but due to energy self-shielding, the poisoning effect decreases on a per mass basis as more and more U-236 accumulates in the RU.

References

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[5] Baumann, N. P., et al. (1968). "Resonance Parameters for ^{236}U from Integral Measurements," *Nuclear Science and Engineering* **32**, 267.