



Plutonium Production under Uranium Constraint

Erik Branger^a , Peter Andersson^a , Vitaly Fedchenko^b, Sophie Grape^a ,
Cecilia Gustavsson^a , Robert Kelley^b and Débora Trombetta^a

^aDepartment of Physics and Astronomy, Uppsala University, Uppsala, Sweden; ^bStockholm International Peace Research Institute, Stockholm, Sweden

ABSTRACT

Production rates of fissile materials are often used to independently assess the number of nuclear warheads a state may possess. One key constraint of a plutonium-based nuclear weapons program is the availability of natural uranium, where a shortage of uranium will constrain plutonium production in the fuel cycle. Recycling of the reprocessed uranium can be used to mitigate such a shortage. Furthermore, since military reactors operate in short cycles to ensure that the plutonium is weapon-grade, it may be possible to operate them using slightly depleted uranium, provided that there are sufficient reactivity margins. Using slightly depleted or recycled uranium, the plutonium production can increase by a factor 2–5 as compared to a once-through scenario, for the same input of natural uranium. For future assessments of a state's plutonium production, a uranium constraint should only be considered if there is clear evidence that no nuclear fuel cycle involving uranium recycling is implemented, or if evidence exists that the recycling is insufficient to mitigate the constraint.

ARTICLE HISTORY

Received 22 March 2023
Accepted 5 December 2023

KEYWORDS

Fuel cycle; plutonium production; uranium recycling; reactor modelling; slightly depleted uranium

Introduction

If a current non-signatory state of the Nuclear Nonproliferation Treaty (NPT) were to decide to join it, the state will be required to submit a declaration of all nuclear material in its possession, and there will be a need for a thorough, independent verification of the declaration. For the Treaty on the Prohibition of Nuclear Weapons (TPNW), or for proposed treaties such as the Fissile Material Cutoff Treaty (FMCT), a verification regime accompanying the treaties has yet to be specified, and it is reasonable to assume that one important task for the responsible authority equipped with the mandate to verify the new treaties would include verification of initial declarations. This will, among other things, require verification of the throughput of the various facilities used to produce nuclear material.

CONTACT Erik Branger  Erik.branger@physics.uu.se  Department of Physics and Astronomy, Uppsala University, Ångströmlaboratoriet, Lägerhyddsvägen 1. Box 516, 751 20 Uppsala, Sweden.

This article has been corrected with minor changes. These changes do not impact the academic content of the article.

© 2023 Taylor & Francis Group, LLC

Multiple constraints exist on the nuclear material production in a state producing nuclear weapons, which may affect an independent assessment of the material production. One significant constraint is the availability of natural uranium. The uranium is required to fuel plutonium-producing reactors, for a state-building plutonium-based weapons, or to make highly-enriched uranium (HEU), for uranium-based weapons. Independent assessment to determine the number of nuclear weapons a state possesses is often done by assessing the state's production rate and available stocks of fissile material and then converting it in an estimate of a number of nuclear warheads by assuming fissile materials requirements per warhead.¹ HEU is also a component of secondaries in thermonuclear weapons, which also needs to be considered when judging the number of weapons a state may have produced.² In addition, the majority of military naval nuclear vessels rely on HEU for fuel. There is little possibility for states to clandestinely import large amounts of unsafeguarded uranium for a domestic weapons program, and such states will have to rely on domestically available resources, which could severely limit the available supply.

While a uranium constraint may severely hinder a nuclear weapons program, possibilities exist to recycle and re-use the uranium, which can partly mitigate the shortage.³ For civilian use of reprocessed uranium, options and knowledge exist, but there is less information published regarding the military use of reprocessed uranium.⁴ Furthermore, military reactors operate on short irradiation cycles, achieving significantly lower burnups as compared to civilian reactors, to ensure that the quality of the plutonium is weapon-grade. Due to the short cycles, there may be sufficient criticality margins that military reactors can operate using slightly depleted uranium (SDU), having a uranium-235 content below that of natural uranium. This opens up new possibilities to implement recycling in a military context to better utilize the available uranium.

This paper simulates selected scenarios of nuclear fuel cycles for a military nuclear program, aimed at plutonium production. The chosen scenarios represent different levels of investments in facilities and technical know-how, to assess if the natural uranium consumption can be lowered through recycling. We have chosen to study a heavy water moderated reactor, as such reactors have historically been proposed or used for plutonium production in existing and defunct nuclear weapons programs, such as those in India, Israel, Pakistan, Sweden, Switzerland, and the United States. A civilian CANDU reactor is simulated, as civilian designs are well optimized for economy, including uranium utilization. Thus, using a CANDU model will provide a lower limit on the tolerable uranium-235 contents when using SDU fuel. Military reactors are typically smaller and thus have more neutron leakage, reducing the criticality margins, and requiring a uranium-235 content closer to natural. The aim of this work

is to provide an assessment on how much the natural uranium consumption can potentially be lowered, without focusing on specific states or any implemented fuel cycles. We expect that similar cycles are feasible for states having other reactor types capable of using natural uranium or SDU for fuel, such as graphite-moderated reactors, though determining the level of material saving in such cases will require modeling those fuel cycles.

Historic and current uranium constraints

In the early days of nuclear power, uranium was believed to be a scarce resource, with few mineral deposits of sufficient concentration available to make mining economically justifiable, and much effort was spent on developing breeder reactors. These breeder reactors would be fueled by plutonium while converting enough uranium-238 to plutonium to create equivalent or more plutonium fuel than was consumed. Through breeder reactors and advanced fuel cycles, the uranium shortage would be significantly mitigated for civilian nuclear power. However, as more uranium resources were discovered and exploited, the uranium constraint never materialized, and interest in breeder reactors waned.⁵

While there is little official data on the Soviet Union's fissile material production, it has been assessed that during the early days of their nuclear weapons program, during the period 1950–60, the uranium shortage was a major constraint on the Soviet nuclear program. Not until the '60s and '70s had sufficient production centers in Kazakhstan, Uzbekistan, and Russia been constructed that the shortage could be overcome, with significant natural uranium imports from Czechoslovakia, East Germany, Hungary, and Bulgaria.⁶

In the early 2000s, India was assessed as having insufficient indigenous uranium to fuel both their military and civilian nuclear reactors. Around this time, India put a significant portion of their civilian nuclear program under international safeguards, and as a result, safeguarded uranium could be imported to cover parts of the civilian need. This freed up sufficient uranium resources that the military program could continue unimpeded.⁷

Recently, Pakistan has been assessed as suffering from a uranium shortage.⁸ The perceived uranium shortage has been taken into account in assessments of the Pakistani nuclear weapons arsenal.⁹ However, Pakistan's nuclear program appears to be expanding, with four operating military reactors at the Khushab site and recently added enrichment capabilities.¹⁰ Thus, it can be questioned whether the uranium constraint is real. One aim of this work is to investigate if such uranium shortage can be mitigated through advanced fuel cycles, where uranium is recycled to extend the available supply.

Fuel cycles

Any state with an indigenous nuclear weapons program will need to construct and operate several nuclear fuel cycle facilities. For a state manufacturing plutonium-based weapons, this includes facilities such as uranium mining, conversion, fuel fabrication, a reactor to produce plutonium, and reprocessing facilities.¹¹ If the reactor is heavy water moderated, or graphite moderated, then the reactors can be built to operate on natural uranium. Hence enrichment facilities are not required, which can reduce resource use with respect to finances, personnel, infrastructure, and R&D. However, for a state with a more ambitious nuclear weapons program, enrichment capabilities may still be desired and can be part of the indigenous fuel cycle.

The simplest fuel cycle is a once-through cycle, where plutonium is recovered from the reprocessed spent fuel, and all other reprocessed material is treated as waste. While being cheaper and simpler than recycling the uranium, it requires more input natural uranium. Three main scenarios implementing fuel cycles will be studied in this work, corresponding to different levels of ambition, available resources, possibility of using recycled uranium, and available infrastructure. All studied scenarios require at a minimum all facilities needed for a domestic once-through cycle, including uranium mining, conversion, fuel fabrication, irradiation in a reactor, and reprocessing to extract the produced plutonium. The principal difference between the scenarios is the use of uranium enrichment, and blending, or mixing, the reprocessed uranium with natural or enriched uranium, as summarized in Table 1.

The key features of the fuel cycles in the scenarios are schematically illustrated in Figure 1. In scenario 1, no enrichment occurs, and no reprocessed uranium is recovered, as it is a once-through fuel cycle for comparison and benchmarking. Scenario 2 assumes that no enrichment capabilities are available, hence the enrichment step in Figure 1 is skipped, and natural uranium is added to the fuel fabrication step. Scenario 3 assumes that enrichment capabilities exist so that the product or blendstock can be enriched to an arbitrary level, and that the fuel fabrication stage in Figure 1 will blend reprocessed uranium with enriched uranium. Scenario 3 replaces the material consumed by the reactor with the blendstock, hence the enrichment of the blendstock is determined by the reactor calculations but is

Table 1. Summary of the key differences between scenarios for recycling of uranium and the uranium enrichment needs.

	Recycling approach	Anticipated enrichment level	Enrichment capacity needed
Scenario 1	No recycling	Only natural uranium used	None
Scenario 2	Blending reprocessed and natural uranium	SDU in the reactor, natural uranium for blending	None
Scenario 3	Blending reprocessed and enriched uranium	SDU or natural uranium for the reactor, 40%–60% enriched for blendstock	Low

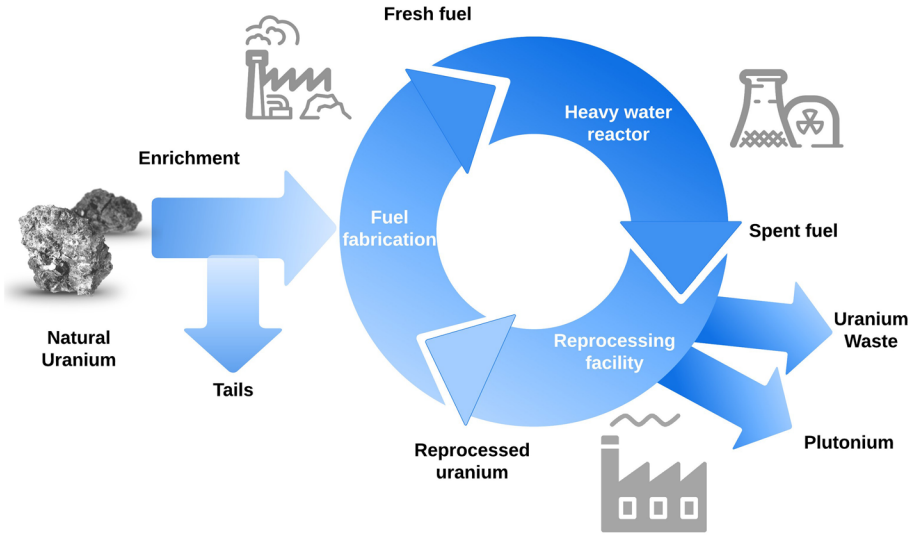


Figure 1. A simple closed fuel cycle, representing scenario 2 and 3 studied in this work.

likely to be between 40 and 60 wt%. Note that [Figure 1](#) contains the key components of a military nuclear fuel cycle studied in this work and much additional infrastructure is needed for a complete weapons program.¹²

Fuel cycle mass balance

As seen in [Figure 1](#), the uranium mass balance must be monitored at several facilities to study the implemented fuel cycle. The key isotopes that will be tracked are uranium-235 and uranium-238. Less abundant isotopes will be tracked when needed. The uranium material balance in the enrichment step is described by [Equations 1](#) and [2](#), for the masses (m_i) and the enrichments (e_i) of material i .

$$m_{product} = m_{feed} + m_{tail} \quad (1)$$

$$e_{product} \cdot m_{product} = e_{feed} \cdot m_{feed} + e_{tail} \cdot m_{tail} \quad (2)$$

[Equation 1](#) details the total uranium mass balance, and [Equation 2](#) does the same for uranium-235, using the uranium masses and enrichments of each flow. To calculate the separative work units (SWU) needed to enrich the material, a value function $V(e)$ is introduced, which is a function of the enrichment e , as described by [Equation 3](#).

$$V(e) = (2e - 1) \ln \left(\frac{e}{1 - e} \right) \quad (3)$$

Using this value function, the SWU requirements can be calculated by Equation 4, relating the enrichments and masses of the uranium streams to the SWU.

$$SWU = m_{product} \cdot V(e_{product}) + m_{tail} \cdot V(e_{tail}) - m_{feed} \cdot V(e_{feed}) \quad (4)$$

For the fuel fabrication step, a mass balance for the total uranium is given by Equation 5, and for the uranium-235 in Equation 6 as a function of the input and output enrichments.

$$m_{fuel} = m_{product} + m_{reprocessed} \quad (5)$$

$$e_{fuel} \cdot m_{fuel} = e_{product} \cdot m_{product} + e_{reprocessed} \cdot m_{reprocessed} \quad (6)$$

The amount of uranium in the used fuel depends on the input mass and the amount of uranium consumed through fission or transmutation, according to Equation 7. The amount of uranium fissioned and transmuted is provided by the burnup simulations. The simulations also provide the amounts of plutonium produced, which will slightly differ from the transmuted uranium, as the transmutation also produces other elements.

$$m_{used} = m_{fuel} - m_{fissioned} - m_{transmuted} \quad (7)$$

Finally, the reprocessing step is assumed to be able to extract all uranium from the used fuel, but the amount of reprocessed uranium may exceed what is needed for the fuel cycle. If this happens, it will be discarded as waste, as described by Equation 8.

$$m_{waste} = m_{used} - m_{reprocessed} \quad (8)$$

In the case that more uranium isotopes need to be tracked, they can be handled similarly to uranium-235 in the equations above, by keeping track of the total uranium amount and the fraction of each isotope in each material in each step. In all studied fuel cycles, the abundance of minor uranium isotopes is expected to be negligible, though this will also be simulated and verified.

Scenario 1

In Scenario 1, a once-through cycle is implemented and will be used as a reference for comparison with the subsequent scenarios. In this scenario, the state has all infrastructure for a once-through cycle but has not developed any enrichment capabilities. With respect to Equations 1–8, all values are uniquely determined, allowing the plutonium produced as a function of input natural uranium to be calculated.

Scenario 2

In Scenario 2, it is assumed that the state has all infrastructure for a once-through cycle, but has not developed any enrichment capabilities, and wants to recycle the reprocessed uranium to mitigate a uranium constraint. If the military reactors are designed to operate with natural uranium fuel, the reprocessed uranium may have a uranium-235 fraction too low to be useful in new reactor fuel (i.e., only a once-through cycle is feasible). However, if there are sufficient margins to criticality, the military reactors can operate on SDU fuel for short reactor cycles. For civilian reactors, optimized for economics including uranium utilization, such margins exist, and if a military reactor is inspired by a civilian one, or has been further rebuilt to allow better margins to criticality, a reactor operation using SDU may be possible.

The scenario consists of blending the reprocessed uranium with natural uranium, to make new fuel material. The uranium-235 fraction of the blended material can be varied from that of natural uranium to that of the once-used uranium, by controlling the fraction of each material in the mixture. The minimum fresh fuel uranium-235 content required depends on the reactor design, thus several initial uranium-235 contents will be studied, and it is expected that the minimum tolerable initial uranium-235 content for the simulated CANDU is below that of a military reactor. Since this scenario blends reprocessed and natural uranium to make fresh fuel, it is likely that not all reprocessed uranium can be used, and any excess will be discarded. With respect to [Equations 1–8](#), once the SDU fuel initial uranium-235 fraction is fixed, all other values are uniquely determined, either from the equations or from the burnup simulations done for each initial uranium-235 fraction studied.

The main advantage of this scenario is that almost all required infrastructure is already present for a state using heavy water reactors to make weapon-grade plutonium. The key changes that need to be made to implement the scenario are to ensure that the reprocessed uranium is sufficiently cleaned from fission products that it can be handled by workers without posing a radiation dose risk. Alternatively, some remote handling capabilities in the reprocessing and fuel manufacturing may mitigate this issue. The fuel fabrication plant must also be able to handle reprocessed uranium, which will be slightly contaminated with fission and activation products. It may be possible to reprocess the uranium again using the available reprocessing infrastructure to obtain the necessary purity, assuming that capacity exists, else additional capacity is needed.

Scenario 3

In Scenario 3, it is assumed that sufficient enrichment capacity is available to supply enriched material for a fuel cycle, in addition to any other

military uses of HEU. In this scenario, the reprocessed uranium is blended with enriched uranium, so that the enriched uranium replaces the uranium consumed in the reactor. The enrichment of the blendstock and amounts of enriched uranium needed depend on the uranium consumption for each simulated fuel initial uranium-235 content. As all reprocessed uranium is recycled, the only uranium waste is the enrichment tails. The scenario will be studied for a reactor operating on natural-equivalent uranium enrichment, and SDU fuel, to identify if any margins to criticality can be used to further improve the efficiency of the fuel cycle also for this scenario. With respect to the mass balance, once the uranium-235 fraction of the fresh fuel and enrichment tails are selected, all other masses can be determined from the fuel cycle mass balance [Equations 1–8](#) and the burnup calculations. Several different tail uranium-235 contents will be simulated, corresponding to different enrichment capabilities.

The simulated fuel cycle in this scenario involves blending the reprocessed uranium with enriched uranium, rather than re-enriching the reprocessed uranium. Such blending has been done for civilian applications and is an option also for military fuel cycles. From a material usage perspective, the cycles are similar in efficiency.¹³ However, blending with enriched material solves some practical issues. There is no risk that any remaining fission products from the reprocessed uranium will be deposited in the enrichment plant, which makes maintenance and operation more difficult. The build-up of uranium isotopes existing in trace amounts, such as uranium-234 and uranium-236 is also slower when blending with enriched material, as those isotopes would be enriched together with uranium-235 should the reprocessed uranium be enriched using an enrichment technology that is based on mass differences, such as centrifuges.¹⁴ Should a more exotic enrichment process such as laser enrichment be used, that can select individual isotopes, such trace isotopes would be less of a concern in the enrichment step.

Simulations

For the benchmarking, a modern CANDU reactor was chosen, as it is a heavy water moderated reactor, it is well developed, and basic design information is publicly accessible. Additionally, civilian operation of such reactors commonly achieves higher burnups than what is needed for military use, showing that there are sufficient margins to criticality that using SDU uranium could be feasible. The CANDU reactor is larger than most military reactors, which reduces neutron leakage and increases margins to criticality, and effort has been put into improving uranium utilization for economic reasons. Hence, the CANDU provides an upper limit to the achievable uranium savings in the simulated scenarios, since military

reactors will likely have a higher neutron leakage, resulting in a higher minimum uranium-235 content of the fresh fuel.

Depletion calculations were performed using Serpent2.¹⁵ An axially infinite unit cell containing one 37-pin fuel bundle was implemented, corresponding to a modern CANDU6 design, and is shown in Figure 2. The bundle was simulated with reflective boundary conditions; thus, the simulated core is infinite in all directions. Note that a real CANDU has CO₂ in the space between the pressure tube and the calandria, however as that has little impact on the neutron flux, it was modeled as void.

The dimensions of the fuel bundle are available in open literature and have already been implemented in a benchmark Serpent input file, for a 37-pin bundle design. However, some temperatures were lowered as compared to this input file, as civilian reactors are optimized for power production and run hot, while a military reactor for plutonium production may be operated at lower temperature.¹⁶ The cooler fuel and moderator temperatures also leads to increased plutonium production, which is relevant for this work. The cooler fuel temperature reduces the Doppler broadening of neutron capture resonance peaks, and the cooler moderator and subsequently increased moderator density further increase the moderation, decrease the resonance capture probability. The result is that plutonium is created more slowly but remains weapon-grade for a longer period. The net effect is an increased plutonium production for the same fuel, at the cost of requiring a higher burnup. The temperatures were based on data for the Nuclear Power Demonstrator (NPD) reactor found in the SFCOMPO database, which was judged to have a representative

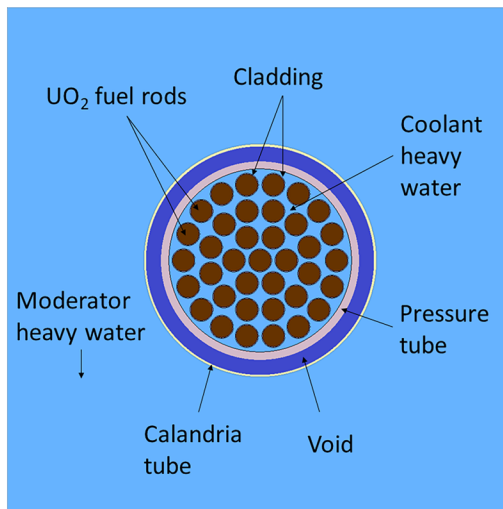


Figure 2. The simulated unit cell geometry, containing one CANDU fuel bundle. Reflective boundary conditions are applied to make an infinite core, with the boundaries matching the ones in the image. The dimensions can be found in Table A1 in the appendix.

power level of a military reactor.¹⁷ The dimensions and temperatures can be found in the [appendix, Table A1](#). Note that for a military reactor, metallic uranium is a fuel option that will also have low temperatures due to good thermal conductivity. However, we here choose to use uranium oxide in the model since it matches the design of the simulated reactor.

The reactor simulations also provide information about the criticality of the infinite core. It is assessed that a core with $k_{\text{inf}} > 1.045$ throughout the reactor cycle can sustain criticality, which means that a maximum of $(k_{\text{inf}} - 1)/k_{\text{inf}} = 4.3\%$ of the neutrons may leak out of the finite core.¹⁸ Should another neutron leakage be assumed, the minimum k_{inf} must be adjusted accordingly. For a reactor operating with batch refueling, where the entire core is replaced at the same time, the limits on k_{inf} must be followed at all times. For a reactor that allows on-line refueling, the changes in k_{inf} are relatively low over time, with small reactivity changes anytime a single fuel bundle is replaced. The effect of online refueling on the criticality can be extrapolated from the simulations. For a core consisting of assemblies of varying criticality, the average of all individual assembly k_{inf} is an indicator of the total core criticality.¹⁹ Hence, the average criticality of the fuel assemblies in the core, having uniform burnup between 0 and the terminal one, is the same as the average criticality of one assembly from a burnup of 0 to the terminal burnup. Thus, the criticality limit to the burnup for on-line refueling is then that the assembly criticality must be above 1.045 on average over its lifetime.

The Serpent2 calculations also include the depletion of the fuel material, and the isotopic composition of the uranium and plutonium. For the mass balances, both the total amounts and the isotopic vector are required, although the result will look primarily at the total plutonium production. The burnup calculations were done in steps of 20 MWd/tU to a burnup of 2000 MWd/tU. From the results, the last burnup step when the plutonium was still weapon-grade (Plutonium-239 $\geq 93\%$) was determined and selected as the true end of the reactor cycle. We have chosen to use the definition of weapon-grade plutonium of Plutonium-239 $\geq 93\%$, ensuring that all scenarios use the same boundary condition, to allow a fair comparison. Comparing this to a real example, France started plutonium production in 1959 with plutonium containing 99.1% plutonium-239, and until 1977 the fraction of plutonium-239 in the produced plutonium dropped steadily to 89.9%, with a total average plutonium-239 contents of 95.3% on all material produced.²⁰

The Serpent2 calculations also include the uranium isotopes that were to be recycled in the simulated fuel cycle, to be used with [Equations 1–8](#). For both natural uranium and the enriched material, uranium-234, uranium-235 and uranium-238 were included, and a natural uranium uranium-235 content of 0.71% has been used. Uranium-234 was modeled in

a simplified manner as being present with 1% the uranium-235 abundance, for both natural and enriched uranium. This considers that it occurs in trace amounts in natural uranium, and that it is enriched together with uranium-235 in a centrifuge. For the recycled uranium, all uranium isotopes may build up, thus all uranium isotopes present were tracked throughout the simulated recycling of the uranium. Uranium recycling was modeled for ten consecutive irradiation and reprocessing cycles, to track whether these isotopes would start to influence the reactor operation. For the enrichment, tail uranium-235 contents of 0.1% to 0.4% in steps of 0.1% were simulated for each scenario, representing varying SWU availability, and that a state with a uranium constraint may be motivated to go for a lower tail uranium-235 content to better utilize the uranium.

For each scenario, the material balance was evaluated to calculate the use of natural uranium. This includes tracking material needed to produce enriched material, tracking material when blended, and tracking uranium that is discharged as waste that could not be further used in the scenario. The result is an estimate of the mass of plutonium produced per input mass of natural uranium. We have also assumed that there are no losses in the reprocessing, both for uranium and plutonium. Note that while a reprocessing plant may have a significant holdup of material, this does not imply that the material is lost, only that its availability is delayed. Losses of less than 0.01% have been demonstrated, hence assuming negligible losses provides an upper limit on the possible plutonium production.²¹ Should material be lost in the cycle, the material balance needs to be recalculated to take this into account, as input natural uranium is needed to cover the losses.

Results

Criticality simulations

The result of the criticality simulations is shown in [Figure 3](#). For natural uranium, k_{inf} is always above 1.089, and on average at 1.0925, significantly above the criticality limit of $k_{\text{inf}} > 1.045$. Hence, such a fresh core will require the use of control rods or other neutron absorbing material to suppress the multiplication to sustain criticality, and using SDU for the fresh fuel together is feasible for this reactor design. Simulations were done for initial uranium-235 content between 0.57 and 0.71 wt%, in steps of 0.01 wt%. For the simulated cases using batch refueling, the criticality limit $k_{\text{inf}} > 1.045$ imposes a minimum initial uranium-235 content of 0.64 wt%, and for on-line refueling, the corresponding content is 0.62 wt%. The uranium-235 content of the used fuel drops by 0.12–0.14 wt% as compared to the initial one at the terminal burnup. Data on the criticality

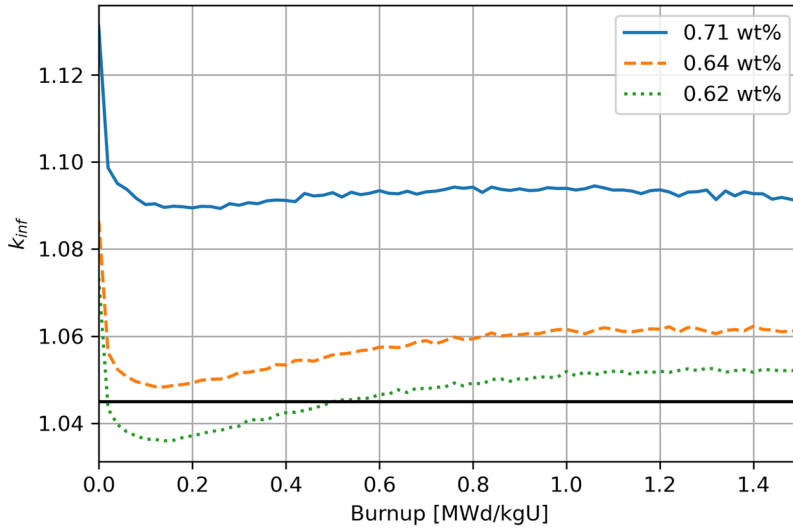


Figure 3. The criticality of the simulated reactor as a function of burnup, for three different initial uranium-235 contents, corresponding to natural uranium, the minimum initial uranium-235 contents for a batch-refueled and for an online refueled reactor. The $k=1.045$ minimum criticality limit is shown in the plot.

for each simulation, as well as the uranium-235 content at terminal burnup, can be found in Table A2 in the appendix. The calculated k_{inf} in Table A2 can also be used to calculate the maximum tolerable neutron leakage for each initial uranium-235 content, for comparing the effect of other assumptions on the neutron leakage. Note also that if natural uranium is used, the uranium-235 content of the used fuel is 0.577 wt%, well below the minimum tolerable initial uranium-235 content. Thus, recycled material cannot be used solely in a reactor, and additional reactivity needs to be provided by adding uranium-235 in some way. The isotopic composition of the plutonium can be found in Table A3 in the appendix.

For the scenarios involving uranium recycling additional simulations were run, where the uranium was recycled and used in ten irradiation cycles, tracking all uranium isotopes throughout the fuel cycle. However, since the cycles were chosen to minimize the growth of the minor isotopes, the results are similar in all scenarios, though the build-up of minor uranium isotopes is most pronounced in scenario 3, where all minor isotopes are recycled. In scenario 3, the only isotope that noticeably increase in abundance is uranium-236, which after 10 recycling may be up to 0.2 wt% of the uranium. However, as uranium-236 behaves much like uranium-238 which is much more abundant, its presence will have a negligible effect on the criticality. Uranium-236 can also capture two neutrons to eventually form plutonium-238, which is not desired in weapon-grade plutonium. However, since the intermediate nuclei neptunium-237 is removed at each

reprocessing step, the plutonium-238 production is suppressed, and negligibly impacts the results. Hence, from a criticality and plutonium production point of view, multiple recycles are feasible, although the efficiency of the cycles will be lowered somewhat if uranium is discarded after a number of recycles. For the other tracked uranium isotopes, they are too rare to affect reactor operations or the plutonium production, though their presence may be useful in determining if the plutonium was produced using recycled uranium. The uranium isotopic composition for the first and the tenth cycle can be found in [Table A4](#) in the [appendix](#).

Scenario 1

For comparison and benchmarking, the results of a once-through fuel cycle with natural uranium are presented. With the implemented core model, the plutonium is weapon grade up to 1300 MWd/tU (Plutonium-239 $\geq 93\%$), which matches values in literature.²² The uranium-235 content of the used uranium is 0.577 wt%. Finally, the plutonium production is calculated as 1001 grams of plutonium per metric ton of natural uranium (gPu/t.nat.U).

Scenario 2

The plutonium production as a function of initial fuel uranium-235 content is shown in [Figure 4](#).

As seen in [Figure 4](#), for the lowest acceptable initial uranium-235 content of 0.62 wt% for an online-refueled reactor, the plutonium production is 1742 gPu/t.nat.U. The corresponding minimum initial uranium-235 content for a batch-operated reactor is 0.64 wt%, resulting in a plutonium production of 1569 gPu/t.nat.U. Note that these values apply to an operational cycle, and neglects additional uranium required to start the cycle. At an initial enrichment of 0.62 wt%, the fresh fuel consists of 44% reprocessed uranium and 56% natural uranium, which is the cause of the natural uranium savings in this scenario. In general, if the initial enrichment is lowered, the plutonium stops being weapon-grade at slightly lower burnups. For an initial enrichment of 0.62 wt%, the burnup limit is 1180 MWd/tU, as compared to 1300 for initial natural uranium. This is expected, since for a reactor with less uranium-235 a higher neutron fluence is required to obtain the same burnup, and plutonium isotopic degradation therefore occurs earlier.

Scenario 3

[Figure 5](#) shows the plutonium production as a function of the selected fuel initial uranium-235 content for four different enrichment tail

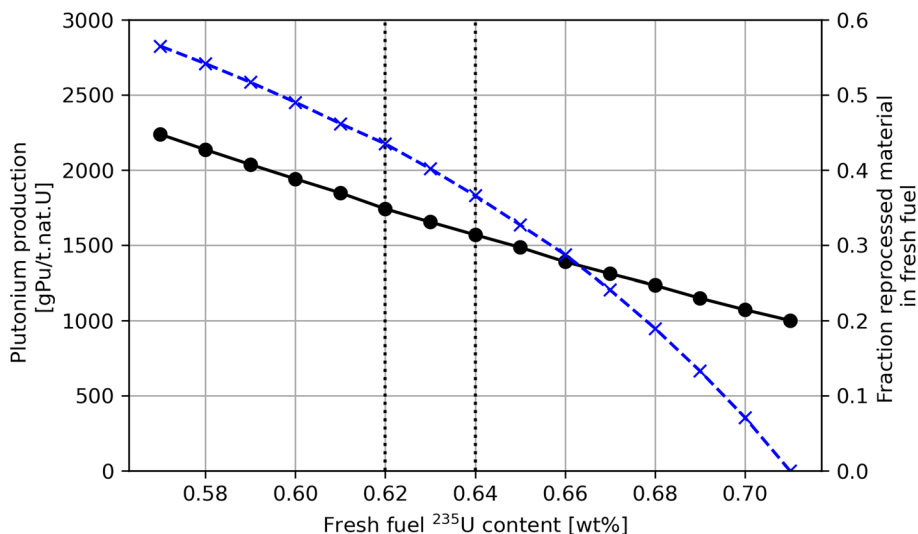


Figure 4. The plutonium production (solid line, left y-axis) and fraction of reprocessed uranium in the fresh fuel (dashed line, right y-axis) for scenario 2. The minimum initial uranium-235 content for a batch refueled and an on-line refueled reactor are marked with dotted vertical lines.

uranium-235 contents. As before, initial uranium-235 content limits of 0.62% for on-line refueling and 0.64% for batch refueling apply to the simulated reactor.

As can be seen in Figure 5, the plutonium production is further increased in this scenario as compared to scenario 2. For the simulated tail uranium-235 contents, a plutonium production between 2326 and 5096 gPu/t. nat.U is achieved, where the highest production is for the lowest tail uranium-235 content and an initial fuel uranium-235 content of 0.62 wt%. The plutonium production does not vary strongly with the fresh fuel uranium-235 content, hence this scenario will be much less reliant on using reactor designs with good margins to criticality, although a lowered initial uranium-235 content is advantageous. When 1 ton of fuel has been irradiated in the reactor up to the burnup limit, between 2.2 and 2.5 kg of uranium will be consumed (fissioned or transmuted), depending on the chosen initial uranium-235 content, that needs to be replaced using the enriched blendstock. The required blendstock enrichment is also similar for the different initial uranium-235 content simulated, and is between 52–55 wt%, to ensure that the blended, fresh fuel has the required initial uranium-235 content. The natural uranium requirement to make the enriched blendstock in all simulated cases ranges between 190 and 430 kg, where the lowest value corresponds to a low tail uranium-235 content and a low fresh fuel uranium-235 content. The SWU usage is calculated to be

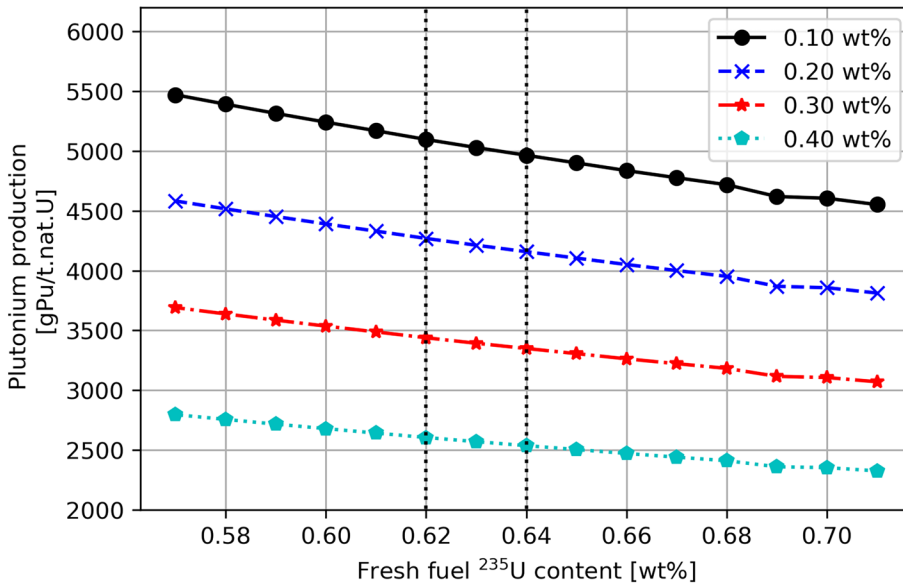


Figure 5. The plutonium production in scenario 3, as a function of initial uranium-235 content. Four different tail uranium-235 contents of 0.1 wt%–0.4 wt% were simulated. The minimum initial uranium-235 content for a batch refueled and an online refueled reactor are marked with dotted vertical lines.

between 50 and 100 SWU per kg of plutonium, where a lowered fresh fuel uranium-235 content leads to a lowered SWU requirement, as the required blendstock enrichment and amount is slightly lowered. Depleting the tails more requires more SWUs but also produces more plutonium for the same input uranium, resulting in only modest changes on the order of 15% in SWU usage for a selected fuel initial uranium-235 content.

Discussion

Material savings

The maximum plutonium production for the studied scenarios is presented in Table 2, together with the mass balance solving the system of Equations 1–8. The masses are normalized to 1 ton of fresh fuel, to allow scaling to another reactor size. The results correspond to the lowest initial enrichment that the simulated core could tolerate with on-line refueling of 0.62 wt% for scenario 2 and 3, as the plutonium production increased with decreasing initial enrichment. The optimum tail enrichment is unsurprisingly the lowest possible, which in the simulations was 0.1%. This will however require significant enrichment capabilities, and if unavailable, will require a higher tail enrichment, which lowers the plutonium production per input uranium.

Table 2. The total uranium and uranium-235 content for the maximum plutonium production in each scenario, solving the system of Equations 1–8, the plutonium production and the SWU usage. The masses are normalized to 1 ton of fresh fuel.

Scenario	1	2	3
Natural uranium	1000.0 kg 0.71 wt%	565.0 kg 0.71 wt%	193.1 kg 0.71 wt%
Enrichment tails		–	190.9 kg 0.1 wt%
Enriched U/Blendstock, for fresh fuel fabrication	1000.0 kg 0.71 wt%	565.0 kg 0.71 wt%	2.2 kg 52.50 wt%
Fresh fuel	1000.0 kg 0.71 wt%	1000.0 kg 0.62 wt%	1000.0 kg 0.62 wt%
Used fuel	997.6 kg 0.58 wt%	997.8 kg 0.50 wt%	997.8 kg 0.50 wt%
Reprocessed uranium used for fuel manufacturing	–	435.0 kg 0.50 wt%	997.8 kg 0.50 wt%
Reprocessed uranium discarded as waste	997.6 kg 0.58 wt%	562.8 kg 0.50 wt%	–
Plutonium per fresh fuel	1001 gPu/t.fuel	984 gPu/t.fuel	984 gPu/t.fuel
Plutonium per input natural uranium	1001 gPu/t.nat.U	1742 gPu/t.nat.U	5096 gPu/t.nat.U
SWU consumption	–	–	74 SWU/kgPu

As can be seen in Table 2, the plutonium production can be increased by a factor of just over five in the most productive scenario, for the same natural uranium consumption, as compared to scenario 1. Alternatively, if the throughput is kept constant, the same throughput as scenario 1 can be achieved with one-fifth of the natural uranium usage. For a state having no enrichment capabilities, savings are possible if the reactor can operate on SDU fuel. For states having enrichment capabilities, it is feasible to rely on natural-equivalent uranium-235 content in the fresh fuel. These results clearly show that a uranium constraint can be mitigated with an advanced fuel cycle, also in the context of plutonium-producing reactors operating with short cycles.

Estimates of a state's stockpiles of nuclear material that include a uranium constraint may need revision, unless there is no evidence of uranium recycling. When recycling cannot be excluded, it should be assumed that it can occur, and thus that the supply of uranium is significantly extended, possibly to the extent that it is not a constraint for the state's fuel cycle. Additionally, if a state's fuel cycle is to be evaluated, some additional uranium is required to get the cycle started, as there will initially be no reprocessed uranium available. This will slightly lower the efficiency of the overall cycle as compared to the results in Table 2, which are for fuel cycles having reached equilibrium.

HEU production for nuclear weapons

For a state with a more ambitious nuclear program, there may be other needs for HEU, such as for making the secondary stage of a thermonuclear device, or for other reactor applications such as for nuclear-powered submarines.²³ In this case, the state will need to balance the uranium allocation between the programs. Since such a state will have enrichment capabilities, all scenarios studied here could be implemented. An advanced fuel cycle

may be desirable to minimize the material use for the plutonium production, freeing as much natural uranium as possible for HEU production.

Alternative uranium sources

While the studied recycling scenarios can be used to mitigate a uranium constraint, other potential sources of uranium should not be forgotten. Such sources could include undeclared or smuggled imports, which may be difficult to detect and quantify. They could also include production or declared imports of material that contains low amounts of uranium, which is not declared as uranium-containing material. Fertilizers are one example. Due to the large quantities of fertilizers used by a state, extracting the trace amounts of uranium can provide useful amounts, and this has been accomplished previously.²⁴ Additionally, rare-earth minerals, coal ash, or uranium extracted from seawater may be unconventional sources for undeclared uranium import or production.²⁵ While some of these sources may not be economically viable, for a state that cannot import uranium for military use the economics may be less of an obstacle. Thus, should a state's nuclear program ever be verified, it may be important to also be able to determine the source of the uranium.

Conclusion and outlook

Independent assessment of the number of nuclear warheads a state may possess is often done by analyzing known nuclear infrastructure, to assess the production of nuclear material. Both historically and currently, there are states that are assessed as suffering from a natural uranium shortage, which constrains nuclear material production, and affects the assessment of nuclear material production. Due to the relatively short irradiation cycles and low burnups achieved in a military reactor, there can be margins to the criticality limit, which allows the use of SDU fuel. Such reactors, together with a fuel cycle to recycle and reuse the uranium, can mitigate a uranium shortage. In the simulated reactor, the margins allowed the fresh fuel to have a uranium-235 content of 0.62% for online reloading, and 0.64wt% for batch reloading.

In the simulated irradiation cycles, 2.2–2.5 kg of uranium is consumed per ton of fuel, terminating the irradiation when the resulting plutonium is still weapon-grade. Hence, it is straightforward to replace the consumed material by blending the reprocessed SDU with that much HEU, to make the blended material equivalent to that of the fresh fuel. In the case that enrichment is not available, if the reactor can run on SDU, it is possible to blend the reprocessed uranium with natural uranium, to make fresh fuel with the desired uranium-235 content. Blending with very small

amount of HEU or somewhat more natural uranium allows the reprocessed material to be used repeatedly, using in total less input natural uranium as compared to a once-through cycle.

The three scenarios studied in this work correspond to the availability of different nuclear fuel cycle facilities and infrastructure, primarily the enrichment capacity of a state. For a state with a basic nuclear weapons program based on plutonium produced in heavy-water moderated reactors, the plutonium production per natural uranium input can potentially nearly double if recycling is used, even without enrichment if reactors can operate SDU fuel. For a state with enrichment capabilities, the increase can be up to a factor five in the simulated scenarios, if the tail enrichment is kept low, even when using natural-equivalent uranium fuel. Thus, the potential savings depend strongly on the criticality margins of the reactor and the enrichment capability of a state, which must both be considered. Furthermore, should a state have produced an excess of HEU, this material can also be used as blendstock. This would ensure that plutonium production can proceed without input of natural uranium for extended periods of time, as 2.2 kg of 52.5% enriched HEU is needed to produce 1 ton of fresh fuel in the scenario with the highest plutonium production.

Should the nuclear material of a state be verified under the NPT, or verification regimes related to treaties such as TPNW or FMCT, there may be a need to verify the complete nuclear fuel cycle. This includes determining if uranium recycling has taken place, which fuel cycle was implemented, and verifying it to determine if nuclear material declarations match production. Nuclear archaeology can be a powerful tool here, to investigate facilities and material for verifying their operation.^{23,26} Depending on the fuel cycle implemented, different uranium waste streams arise, which can be measured to determine which fuel cycle was used and how it was operated. An accurate simulation of the fuel cycle can help determine the expected uranium and plutonium masses and compositions, including the presence of minor uranium isotopes that are a sign that reprocessing and recycling have occurred. In addition, the changes in the fresh fuel isotopic composition may also affect the abundance of fission products, which may also serve as a signature to verify the fuel cycle.

Acknowledgements

We would like to acknowledge the Alva Myrdal Centre for Nuclear Disarmament at Uppsala University for supporting this work.

Disclosure statement

No potential conflict of interest was reported by the author(s).

Notes and References

1. Alexander Glaser and Julien Troullioud de Lanversin, "Plutonium and Tritium Production in Israel's Dimona Reactor 1964–2020," *Science & Global Security* 29 (2021): 90–107, <https://doi.org/10.1080/08929882.2021.1988325>; Sherzod R. Kurbanbekov, Seung Min Woo, and Sunil S. Chirayath, "Analysis of the DPRK's Nuclear Weapons Capabilities by Estimating Its Highly Enriched Uranium Stockpile and Natural Uranium Reserves," *Science & Global Security* 27 (2019): 113–123, <https://doi.org/10.1080/08929882.2019.1657608>; Lalitha Sundaresan and Kaveri Ashok, "Uranium Constraints in Pakistan: How Many Nuclear Weapons Does Pakistan Have?" *Current Science* 115 (2018), <https://www.jstor.org/stable/26978355>.
2. Vitaly Fedchenko and Robert Kelley, "Proliferation & Procurement. New Methodology Estimates North Korean Stockpile," *Janes Intelligence Review* 32, no. 9 (2020): 44–49.
3. Alexander Glaser, "Isotopic Signatures of Weapon-Grade Plutonium from Dedicated Natural Uranium–Fueled Production Reactors and Their Relevance for Nuclear Forensic Analysis," *Nuclear Science and Engineering* 163, no. 1 (2009): 26–33, <https://doi.org/10.13182/NSE163-26>.
4. International Atomic Energy Agency, "Use of Reprocessed Uranium: Challenges and Options," *IAEA Nuclear Energy Series*, NF-T-4.4, 2010, <https://www.iaea.org/publications/8010/use-of-reprocessed-uranium-challenges-and-options>
5. Frank N. von Hippel, "The Rise and Fall of Plutonium Breeder Reactors," in *Fast Breeder Reactor Programs: History and Status*. (Princeton NJ: IPFM, Program on Science and Global Security, 2010), https://fissilematerials.org/publications/2010/02/fast_breeder_reactor_programs_.html.
6. Oleg Bukharin, "Analysis of the Size and Quality of Uranium Inventories in Russia," *Science & Global Security* 6 (1996): 59–77, <https://scienceandglobalsecurity.org/archive/sgs06bukharin.pdf>.
7. Zia Mian et al., "Plutonium Production in India and the U.S.-India Nuclear Deal," in *Gauging U.S.-Indian Strategic Cooperation*, edited by Henry Sokolski. (Strategic Studies Institute, US Army War College, 2007), <http://www.jstor.org/stable/resrep11995.7>.
8. Zia Mian, Abdul H. Nayyar, and R. Rajaraman, "Exploring Uranium Resource Constraints on Fissile Material Production in Pakistan," *Science and Global Security* 17 (2009): 77–108, <https://doi.org/10.1080/08929880902975834>.
9. Sundaresan and Ashok, Uranium Constraints in Pakistan, 2018.
10. Sarah Burkland, Allison Lach, and Frank Pabian, *Khushab update*, *Institute for Science and International Security*, 2017, <https://isis-online.org/isis-reports/detail/khushab-update/12>; David Albright, Sarah Burkland, and Frank Pabian, *Pakistan's Growing Uranium Enrichment Programme*. (Institute for Science and International Security, 2018), <https://isis-online.org/isis-reports/detail/pakistans-growing-uranium-enrichment-program/12#images>.
11. U.S. Department of Energy, *Nuclear Fuel Cycle and Weapons Development Process* (chart). (Richland, WA: Pacific Northwest National Laboratory, 2009), <http://plaza.ufl.edu/sjoden/ENU4930/Week2/Non-Prolif-FreezeFrame2009.pdf>.
12. U.S. Department of Energy, 2009.
13. International Atomic Energy Agency, *Use of Reprocessed Uranium: Challenges and Options*, 2010.
14. Vitaly Fedchenko, *The New Nuclear Forensics: Analysis of Nuclear Materials for Security Purposes* (Oxford United Kingdom: Oxford University Press, 2015), https://www.sipri.org/sites/default/files/2018-09/sipri_new_nuclear_forensics_fedchenko_150712.pdf.

15. Jaakko Leppänen et al., “The Serpent Monte Carlo Code: Status Development and Applications in 2013,” *Annals of Nuclear Energy* 82 (2013):142–50. <https://doi.org/10.1016/j.anucene.2014.08.024>.
16. Example input files, https://serpent.vtt.fi/mediawiki/index.php/Collection_of_example_input_files
17. F. Michel-Sendis et al., “SFCOMPO-2.0: An OECD NEA Database of Spent Nuclear Fuel Isotopic Assays, Reactor Design Specifications and Operating Data,” *Annals of Nuclear Energy* 110 (2017): 779–788, www.sciencedirect.com/science/article/pii/S0306454917302104.
18. Example input files, https://serpent.vtt.fi/mediawiki/index.php/Collection_of_example_input_files
19. Peter. G. Boczar, et al., “Recent Advances in Thorium Fuel Cycles for CANDU Reactors,” *Thorium Fuel Utilization: Options and Trends* (2002): 104.
20. International Panel on Fissile Materials, *Global Fissile Material Report 2010: Balancing the Books: Production and Stocks: Fifth Annual Report of the International Panel on Fissile Materials* (Princeton, N.J: IPFM, 2010), https://fissilematerials.org/publications/2010/12/global_fissile_material_report_4.html.
21. Paul E. Faugeras et al., The Marcoule Pilot Plant, CEA-CONF-8524, (1986), https://inis.iaea.org/search/search.aspx?orig_q=RN:18001363.
22. Alexander Glaser, “Isotopic Signatures of Weapon-Grade Plutonium,” 2009.
23. Vitaly Fedchenko and Robert Kelley, “Proliferation & Procurement,” *Janes Intelligence Review*, 2020.
24. Nils Haneklaus, Anastasiya Bayok, and Vitaly Fedchenko, “Phosphate Rocks and Nuclear Proliferation,” *Science & Global Security* 25 (2017): 143–158, <https://doi.org/10.1080/08929882.2017.1394061>.
25. Peter S. Waring et al., “Unconventional Uranium: Prospects, Progress and Challenges,” (Symposium on International Safeguards: Reflecting on the Past and Anticipating the Future, Vienna, Austria, October 31–November 4, 2022).
26. Erin J. Holland et al., “An Introduction to Nuclear Industrial Archaeology,” *Sustainability* 15, no. 7 (2023): 6178, <https://doi.org/10.3390/su15076178>

Appendix

Table A1. Parameters used for the CANDU bundle simulations.

Fuel pellet radius	0.6122 cm
Cladding inner radius	0.6122 cm
Cladding outer radius	0.06540 cm
Lattice	4 rings, the third rotated by 15 degrees. Ring radius: 0 cm, 1.4885 cm, 2.8755 cm, 4.3305 cm
Pressure tube inner radius	5.1689 cm
Pressure tube outer radius	5.6032 cm
Calandria inner radius	6.4478 cm
Calandria outer radius	6.5875 cm
Assembly pitch	28.574 cm
Fuel material	Uranium dioxide
Fuel density	10.4375 g/cm ³
Fuel temperature	683 K
Moderator	D ₂ O
Moderator density	1.082885 g/cm ³
Moderator temperature	311 K
Coolant	D ₂ O
Coolant density	0.81212 g/cm ³
Coolant temperature	537.5 K
Cladding material	98.1858% Zr, 1.3955% Sn, 0.1994% Fe, 0.1196% O, 0.0997% Cr
Cladding density	6.56 g/cm ³
Calandria material	99.7% Zr, 0.16% Mg, 0.11% Cr, 0.06% Ni, 0.0003% B
Calandria density	6.44 g/cm ³
Pressure tube material	99.9% Zr, 0.00021% B
Pressure tube density	6.57 g/cm ³
acelib	sss_jeff311u.xsdata
declib	endf-b-vi-8_decay.dat
nfylib	endf-b-vi-8_nfp.dat
Heavy water thermal scattering library	therm hwtr 0 hwj3.00t hwj3.14t using “tms” for interpolation.

Table A2. Uranium enrichment before and after irradiation, the maximum burnup when the plutonium is still weapon-grade, minimum and average criticality for the uranium blending scenarios, and the maximum allowable neutron leakage for each enrichment level, for both a batch refueled reactor and an online refueled reactor. This work assumes that the criticality must be above 1.045 for a CANDU, which corresponds to allowing 4.3% of the neutrons to leak out of the core.

Initial U235 contents (wt%)	Final U235 contents (wt%)	Terminal burnup (MWd/tU)	Minimum criticality	Average criticality	Maximum neutron leakage, batch refueling (%)	Maximum neutron leakage, online refueling (%)
0.71	0.5770	1300	1.08928	1.092578	8.20	8.47
0.70	0.5692	1280	1.08389	1.087777	7.74	8.07
0.69	0.5596	1280	1.07808	1.082653	7.24	7.63
0.68	0.5518	1260	1.07248	1.07761	6.76	7.20
0.67	0.5440	1240	1.06667	1.072356	6.25	6.75
0.66	0.5361	1220	1.06089	1.067152	5.74	6.29
0.65	0.5266	1220	1.05465	1.061812	5.18	5.82
0.64	0.5187	1200	1.04836	1.056349	4.61	5.33
0.63	0.5109	1180	1.04194	1.051355	4.03	4.88
0.62	0.5031	1160	1.03597	1.045649	3.47	4.37
0.61	0.4935	1160	1.02934	1.039928	2.85	3.84
0.60	0.4857	1140	1.02291	1.034123	2.24	3.30
0.59	0.4779	1120	1.01617	1.028149	1.59	2.74
0.58	0.4701	1100	1.00895	1.022046	0.89	2.16
0.57	0.4623	1080	1.00178	1.015891	0.18	1.56

Table A3. Results for the simulations of plutonium production. The initial uranium-235 content correspond to natural uranium, the criticality limit for batch refueling (0.64 wt%) and online refueling (0.62 wt%). The simulations were run to the maximum burnup where the plutonium is still weapon-grade (Plutonium-239 > 93%), denoted as terminal burnup in the table. The plutonium isotopes values are the mass fraction of the total plutonium contents of the used fuel. The plutonium production as a function of the input fuel amounts is included in the last row of the table.

Initial uranium-235 content	0.71 wt%	0.64 wt%	0.62 wt%
Terminal burnup	1.30 MWd/kgU	1.20 MWd/kgU	1.16 MWd/kgU
Plutonium-236 fraction	1.00E-13	8.15E-14	8.53E-14
Plutonium-237 fraction	2.20E-10	1.97E-10	2.06E-10
Plutonium-238 fraction	7.11E-05	6.59E-05	6.33E-05
Plutonium-239 fraction	9.30E-01	9.30E-01	9.30E-01
Plutonium-240 fraction	6.48E-02	6.51E-02	6.46E-02
Plutonium-241 fraction	4.96E-03	4.88E-03	4.77E-03
Plutonium-242 fraction	1.69E-04	1.68E-04	1.63E-04
Plutonium-243 fraction	1.85E-08	1.95E-08	1.88E-08
Plutonium-244 fraction	1.24E-10	1.30E-10	1.26E-10
Total plutonium	1.0012E+03 gPu/t.fuel	9.9470E+02 gPu/t.fuel	9.8428E+02 gPu/t.fuel

Table A4. Mass fractions of the uranium isotopes in the fuel cycle showing its evolution over the first cycle after having been recycled and irradiated 10 times. The results follow scenario 3, where all uranium isotopes are recycled in every step. For the natural uranium and the enriched blendstock, it is assumed that uranium-234 is present at 1% of the mass of uranium-235, to determine if the trace amounts present will significantly increase in the fuel cycle.

	1st cycle start	1st cycle end	10th cycle start	10th cycle end
Uranium-232	0	1.8638E-14	2.5450E-13	2.8780E-13
Uranium-233	0	4.1828E-11	3.2700E-10	3.5275E-10
Uranium-234	7.1000E-05	6.8420E-05	1.6067E-04	1.5530E-04
Uranium-235	7.1000E-03	5.7727E-03	7.0999E-03	5.7627E-03
Uranium-236	0	2.0420E-04	1.9750E-03	2.1755E-03
Uranium-238	9.930E-01	9.9395E-01	9.9076E-01	9.9190E-01