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Nuclear Archaeology to Distinguish Plutonium and Tritium Production Modes in Heavy-Water Reactors

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ABSTRACT

Several nuclear archaeology techniques have been proposed to determine historic plutonium production in dedicated nuclear reactors. These methods rely on sampling materials from the reactor core, or specially designed monitor tags, to examine suited isotopic ratios and deduce the amount of plutonium produced. However, some production reactors are capable of producing isotopes other than plutonium, such as tritium. If a reactor was declared to produce tritium, it would be crucial to confirm that it was in fact producing tritium, and not plutonium. In this paper, we describe how isotopic ratios discrepancies could be used to distinguish between plutonium and tritium production modes. We present results obtained from the simulation of reactor lattices inspired by Savannah River Site heavy-water production reactors and show that elements such as hafnium and tungsten can detect undeclared production of plutonium.

Introduction

Tritium production around the world

While the production of fissile materials takes place under international safeguards in non-nuclear weapon states, and would be verified in nuclear weapon states as part of a future Fissile Material Cutoff Treaty, the production of other nuclides, such as tritium, are not typically monitored or constrained. Tritium boosts the explosive fission chain reaction in the first stage of a nuclear weapon and must be regularly replenished due to its short half-life of 12.3 years. There are also some nonmilitary uses such as the introduction of fusion reactors that would require a massive increase in global tritium production.[1] While there are various technologies and methods to produce tritium, countries have historically relied on two main

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Table 1. Annual tritium requirements to maintain nuclear arsenals in weapon states.

	United States	Russia	France	China	U.K.	Pakistan	India	Israel	DPRK
Active arsenal (warheads)[14]	4480	4300	300	270	215	130	120	80	13–30[15]
DSB Task Force estimate	1700 g	1600 g	110 g	100 g	80 g	50 g	45 g	30 g	5–10 g
RDD-8 upper bound	4900 g	4700 g	330 g	300 g	230 g	140 g	130 g	90 g	15–30 g

approaches: detritiating heavy water and exposing lithium targets to a neutron flux in a nuclear reactor. Detritiating heavy water extracts the tritium created when deuterium in heavy water absorbs a neutron. Exposing lithium targets in a nuclear reactor produces tritium because lithium-6 splits into helium-4 and tritium after it absorbs a neutron. Exposing lithium targets to a neutron flux in a nuclear reactor is particularly effective in reactors moderated with heavy water, due to the low parasitic neutron absorption cross section compared to light water.[2] This heavy-water reactor strategy was pursued in the United States (Savannah River Site reactors), Russia (Mayak site, Ludmila LF-2 reactor), France (Celestin I and II), and Israel (Dimona reactor).[3] The United Kingdom imported tritium from the United States and produced its own tritium using graphite-moderated (MAGNOX) reactors, which are now shut down.[4] China also used graphite-moderated reactors (Jiuquan and Guangyuan reactors) to produce tritium until the mid-1980s. India and Pakistan have been producing tritium by detritiating the heavy water used as a moderator and coolant.[5] Additional production paths exist, but these are less effective.[6]

The current tritium production strategy of the United States is based on placing lithium-based tritium producing burnable absorber rods (TPBARs) in pressurized water reactors.[7] Russia continues to use the Ludmila heavy-water reactor, as well as the Ruslan light-water reactor, to produce tritium.[8] France plans to produce tritium in the naval test reactor Réacteur d'essais à terre (RES), a pressurized water reactor that is currently under construction at Cadarache.[9] China might be extracting tritium from its two CANDU reactors.[10] North Korea may be producing tritium in its 5-MWe graphite-moderated reactor or in its light-water moderated research reactor IRT on the Yongbyon site.[11] No changes in production paths are known for India, Pakistan, or Israel.

Table 1 summarizes the estimated tritium requirements for all weapon states based on their current nuclear arsenals, including the annual production rates needed to replenish reservoirs with new stock. The estimated amount of tritium required per warhead is based on two U.S. sources: A 1999 Defense Science Board (DSB) task force report that provides numbers implying an inventory of about 7 g per warhead and The Restricted Data Declassification report (RDD-8) that lists 20 g as an upper limit.[12] Table 2 shows the actual current tritium production capabilities of the five nuclear weapon states of the Nuclear Non-Proliferation Treaty (NPT). For U.S.

Table 2. Annual tritium production in NPT weapon states.

	United States[16]	Russia	France[17]	China	U.K.
Reactor type	1–2 PWR (~1.2 GWt per PWR)	1 HWR (1 GWt) 1 PWR (1 GWt)	1 PWR (150 MW)	2 CANDU (728 MWe each)	Purchase from the U.S.
Tritium production per year	950 (2018) g[18] 1900 (2025) g[19] 3200 (2025) g[20]	4000 g[21]	380 g[22]	190 g[23]	—

production, we show three numbers: an estimate of the production at Watts Bar Unit 1 in 2018, and two possible production strategies that the United States may pursue in 2025. While the two projections for 2025 are consistent with the estimated annual needs to support today's U.S. arsenal, current tritium production is far below these needs, explained by the fact that the United States is currently relying on a tritium reserve.[13] In the case of Russia, we show the maximum production rate since less information is available on the production capability and production schedule of the two Russian reactors. Appendix A summarizes the production estimate for France, however it should be considered a theoretical upper limit of France's tritium production and is therefore above the values found in Table 1. The tritium production estimate from China's two CANDU reactors is also within the required range to support the current nuclear arsenal. The United Kingdom is likely purchasing tritium from the United States under the 1958 U.S.–U.K. Mutual Defense Agreement since all MAGNOX production reactors are shut down.

The need to verify non-production of plutonium with up-stream verifications

Virtually any type of production reactor can, and was used to, produce both tritium and plutonium. For example, in heavy-water reactors, driver fuel rods of highly enriched uranium can provide the neutron flux to irradiate target rods of either lithium (to produce tritium) or natural or depleted uranium (to produce plutonium). In the future, states could therefore secretly produce plutonium and attempt to avoid detection by other states by declaring production of tritium or other non-restricted isotopes. Regarding the past, states may also misinform other states of having produced more tritium than they did, concealing past plutonium production and excess plutonium stocks.

Routine inspections under IAEA Comprehensive Safeguards Agreements can detect deceptive activities when they occur. In non-nuclear weapon states, the IAEA conducts safeguards in all reactors, including research reactors that are declared to be producing isotopes other than nuclear materials. In reactors with power levels larger than 25 MWth, the IAEA verifies the absence of uranium targets for plutonium production using measures such as physical inventory verification and containment and

surveillance.[24] Specifically, the IAEA verifies that undeclared fertile materials are neither inserted nor removed from the reactor core.[25]

The verification standard of a future Fissile Material Cutoff Treaty (FMCT) may be lower than today's Comprehensive Safeguards Agreements. An FMCT would, at a minimum, prohibit the production of fissile materials for nuclear-weapon purposes, including in nuclear weapon states. While the production of unirradiated direct use materials would be verified, i.e., plutonium separation and uranium enrichment, there is no consensus on up-stream verification. This refers to fuel cycle activities up-stream of enrichment and reprocessing facilities, for example verifying plutonium production in reactors, i.e., before the plutonium is separated from the spent fuel. In contrast to restricting verification to unirradiated direct use materials in declared reprocessing plants, up-stream verification could detect produced plutonium to be separated in undeclared facilities.[26] There is recognition of the importance of verifying the non-production of plutonium in reactors under an FMCT, including those declared not to be producing plutonium.[27]

In the case of up-stream verification that includes verifying reactors producing tritium in irradiation targets, verifying the non-production of plutonium under an FMCT could be based on the methods the IAEA uses today. Should such comprehensive verification measures, which provide the highest level of confidence, not be agreed upon in the FMCT context, less intrusive measures could be envisaged that could also provide confidence. Similar approaches may be possible in nuclear weapon states absent an FMCT, that agree to limited monitoring as a transparency measure.

Nuclear archaeology

Distinguishing plutonium production from other production modes is relevant not only for future efforts to verify ongoing production during routine inspections, but also to verify past fissile material production. Nuclear archaeology reconstructs a state's fissile material production history. The method could significantly strengthen measures to verify the completeness of fissile material declarations and ensure that a state did not produce more fissile materials than it declared as part of an arms-control agreement. Under Comprehensive Safeguards (INFCIRC/153), for instance, the IAEA applies safeguards to all sources of special fissionable materials in peaceful nuclear activities to verify that these materials are not diverted to nuclear weapons or nuclear explosive devices. The verification tools of INFCIRC/153 are not sufficient, however, to detect undeclared materials and activities. The Additional Protocol strengthened the verification system in 1997. IAEA safeguards do not explicitly envision methods of nuclear

archaeology. The IAEA has, nevertheless, certain experience in this regard from its verification activities after South Africa joined the NPT in 1991.[28]

A new measure to distinguish production modes both for ongoing and past production could be based on methods that were, to a limited extent, developed in the context of nuclear archaeology. These methods consist of measuring isotopic ratios from samples taken at a nuclear facility (e.g., directly from the reactor core) in order to find the total production of fissile materials, which often also requires forward-simulations of the operations of the facility.[29] In the case of graphite-moderated production reactors, a technique named GIRM (Graphite Isotopic Ratio Method) relies on isotopic ratios measured from samples of the graphite moderator to obtain the neutron fluence and calculate lifetime plutonium production.[30] For reactors that use heavy water as a moderator, similar techniques have been proposed based on the forensic analysis of structural materials.[31]

This paper explores the potential of measuring isotopic ratios in samples to distinguish different modes of reactor operation. The analysis focuses on distinguishing plutonium from tritium production but could also be relevant for identifying the production of isotopes other than tritium. In the future, specially designed monitor tags could be inserted into the reactor core for routine verification of reactor operations. To verify past fissile material production, suitable structural materials from the reactor core must be identified. This paper considers heavy-water reactors due to their relevance for past and ongoing isotope production, but the proposed technique may also be applicable to other reactor types.[32]

Nuclear archaeology for production mode authentication

Our proposed method assumes that the neutron flux spectrum in the core changes if the host surreptitiously switches the production mode. This change in the neutron spectrum is in turn going to affect the reaction rates of certain isotopes as their spectrum-averaged (one-group) microscopic cross-sections change. Ratios of these isotopes will therefore be different from those expected if the country operated the reactor in the declared mode. Looking at the difference between ratios calculated for a declared campaign and actual measured ratios could potentially enable inspectors to detect any deviation from the declared production history. Since these ratios are used to distinguish a production history based on production modes (plutonium vs. tritium), we will call them mode ratios. A difference in the total flux level would also lead to a difference in these mode ratios over time. However, by examining all results against fluence (the product of flux and time) as opposed to time, our method is independent of the total flux level.

To determine which types of isotopes should be used in the mode ratios so that differences in neutron spectra are detectable, we look at the dependence of the microscopic one-group cross-section on the neutron spectrum φ(E):

$$\begin{cases} \sigma = \dfrac{1}{\varphi}\displaystyle\int \sigma(E)\varphi(E)dE \\ \varphi = \displaystyle\int \varphi(E)dE \end{cases}$$

It is important to note that, even if the neutron spectra of two systems are different, the one-group cross-sections might not necessarily reflect this difference: The energy-dependent cross-section $\sigma(E)$ could be large in regions where both spectra are similar, but small in regions where the spectra are different. Obtaining one-group cross-sections that are different due to differences in the neutron spectra requires multiplying the neutron spectra by a cross-section that will amplify the regions of differences. We can establish a list of desired features for isotopes for mode ratios:

- Isotopes must be stable or have a long half-life (of the order of at least hundreds of years) so that their ratios' evolution only evolve with fluence.
- Isotopes considered for a ratio need to be from the same element. In addition, they should not be produced by isotopes of other elements.
- Energy-dependent cross-sections of the suitable isotopes must fold with the neutron spectrum in such a way that the resulting one-group cross-section is sensitive to neutron spectra differences.

To test the concept, we performed depletion calculations based on reactor models inspired by the Savannah River Plants' (SRP) MARK 22 assemblies charge (tritium production) and MARK 15 assemblies charge (plutonium production) which are both arranged within hexagonal lattices. Figure 1 shows a cross-section of both assemblies. The power density was set to 37 kW/l for the MARK 22 design and 40 kW/l for the MARK 15 design. Their life cycles are 200 days and 50 days, respectively.[33]

To model the neutronics and isotopic behavior of each production mode, we used the linkage code MCODE, which couples MCNP5 with ORIGEN2.2.[34] However, MCODE does not track elements that are not important for neutronics, and it therefore excludes many elements that are relevant for nuclear archaeology. Therefore, we used the following method to compute their density evolution with time. The time-averaged flux and one-group cross-sections were computed from the data provided by MCODE for both production modes.

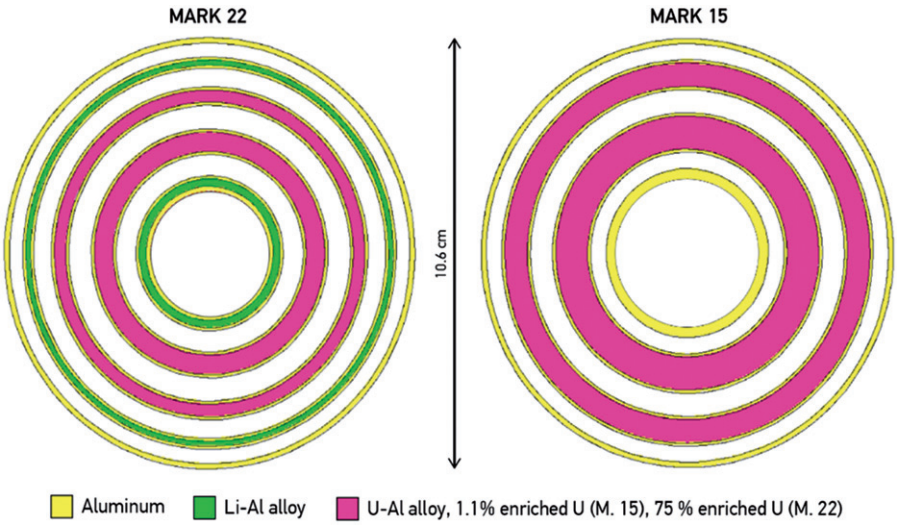

Figure 1. Cross-sectional view of Savannah River Plants' MARK 22 assemblies (for tritium production) and MARK 15 assemblies (for plutonium production).

Then, using these results, Python scripts calculated the isotopic evolution as a function of fluence for the isotopes that are useful for this work. We used infinite lattice configurations for both production charges.

Our model does not specify whether the mode ratios are sampled from structural materials or monitor tags since we are interested in generally identifying viable mode ratios and in quantifying their differences for tritium and plutonium production modes. We consider a scenario where samples would be taken out of or around the outer aluminum sleeve of the assembly. Neutron spectra from other locations such as in the center of assemblies were obtained and do not show significant differences from the one from the outer sleeve. Figure 2 is a plot of the normalized neutron spectra in the outer aluminum sleeves of a MARK 22 and a MARK 15 assembly. Both spectra were taken at the middle point of each charge life cycle respectively, i.e., after 100 days for the tritium production charge and after 25 days for the plutonium production charge. Although they are similar, differences exist at the upper limit of the thermal region and especially in the epithermal region.

Both the differences in geometry and the isotopic composition between MARK-15 and MARK-22 can potentially explain the divergences in the spectra. Starting from the upper limit of the thermal region, with increasing energy, the steeper slope observed for the MARK-15 is explained by the higher concentration of plutonium-239 than in the MARK-22 since the plutonium absorption cross-section in this region has a very broad resonance. In the epithermal region, a higher ratio of metal to water in the MARK-15 can explain that the flux in this energy region is lower than the

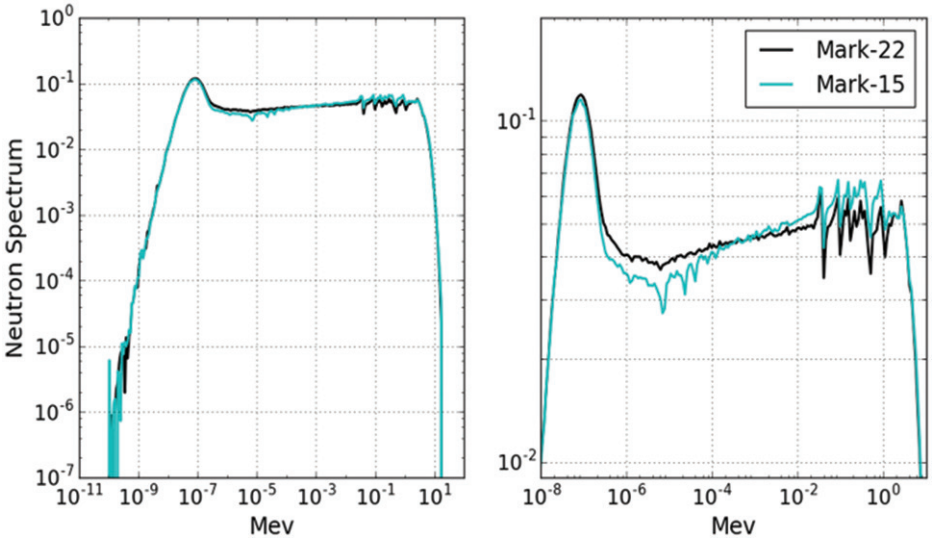

Figure 2. Neutron flux spectrum in the outer sleeves of the MARK 22 and the MARK 15. The plot on the right shows the thermal, epithermal, and resonance regions of the spectra.

MARK-22 spectrum. Neutrons will spend more time in the metal and therefore be more likely absorbed than they would in the MARK-22 geometry. The noticeable absorption peaks in the same regions present for the MARK-15 but not for the MARK-22 stem from a higher concentration of uranium-238 in the MARK-15. Finally, the smaller fast spectrum of the MARK-22 is due to lithium, which has a strong and broad resonance in the fast region.

Because of the observed difference in the epithermal region, isotopes that have a large cross-section in the epithermal relative to the thermal region display a difference in their one-group cross-sections and thus in their reaction rates. Observing the set of criteria listed above for selection of adequate isotopes, ratios between isotopes that are related by (n, γ) reactions are considered here. After a comprehensive evaluation, we selected several elements. Among them, the most promising for production campaign authentication are hafnium and tungsten. Both are familiar in nuclear reactor engineering with hafnium being known for its good mechanical and corrosion resistance properties and its potential for fluence monitoring. Figure 3 displays the (n, γ) cross-section of hafnium-178 and tungsten-182. We can see that they both present a peak in the epithermal region, which makes them good indicators for distinguishing between the MARK 22 and MARK 15 charges. They also have multiple stable isotopes.

The magnitude of the cross-section is another important factor. Ratios that involve high cross-sections will tend to have very short dynamic fluence ranges. Beyond that range, the isotopic ratio reaches an equilibrium where no information can be gained about the production mode. These

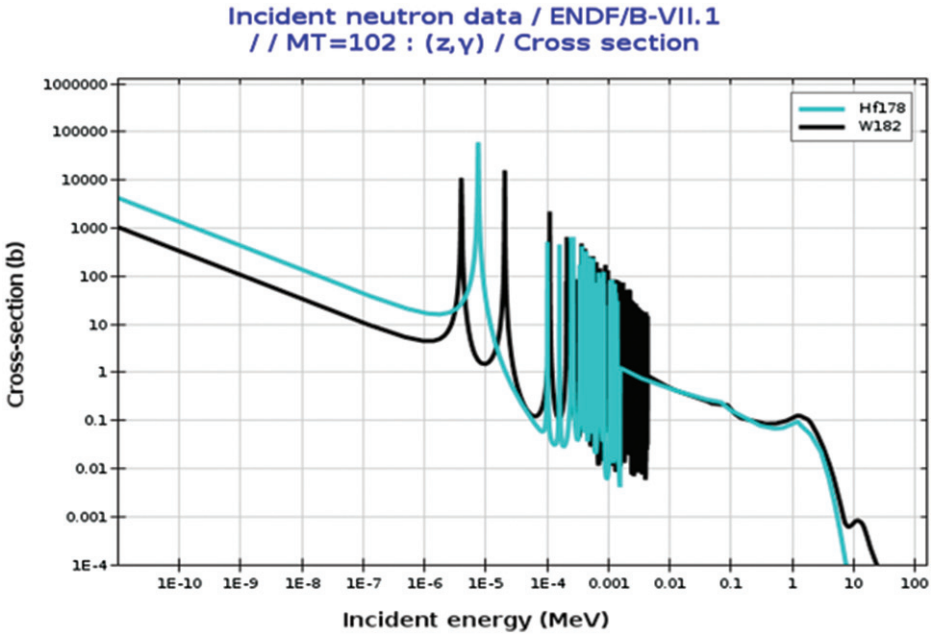

Figure 3. (*n*, γ) cross sections for hafnium-178 and tungsten-182. This plot was generated with Janis-4.0.

ratios will be only useful to verify production modes on short fluence scales. We use ratios with low cross-sections for the large fluence scenario. Figure 4 shows the differences of some isotopic ratios observed from a tritium-only production campaign and a plutonium-only production campaign. This graph helps determine the fluence range in which a specific ratio can be used to verify a declared production mode. In the case of the power density of the SRP reactors, hafnium has isotopes that can be used for about one year (177/176 and 179/177) and between two years and four years (178/176 and 180/178). The tungsten ratio 184/182 will give useful information after six years of operation. Cadmium, which has isotopes with very large (n, γ) cross sections, can be used for the time range of 30–60 days.

While Figure 4 presents useful information, it relies on an extreme scenario in which the host was only producing plutonium during a set of production campaigns declared for tritium production only. In practice, that scenario is unlikely. The host would probably be hiding periods of plutonium production within declared tritium production campaigns. The following discussion shows how mode ratio differences can still be used in this situation.

Figure 5 shows the relative difference observed at the end of the campaigns for several isotope ratios versus the fraction of fluence dedicated to plutonium production. We show three cases with inspections occurring

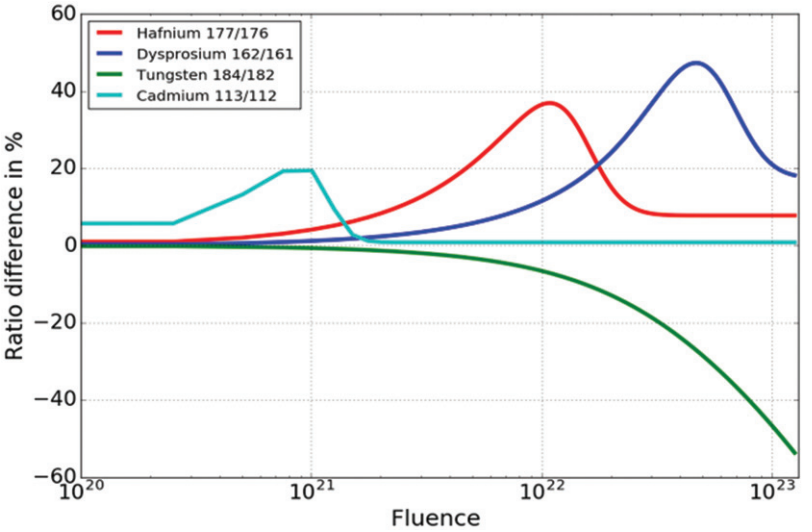

Figure 4. Ratios difference between a tritium-only and a plutonium-only production mode for various isotopic ratios. The *x*-axis uses a log scale.

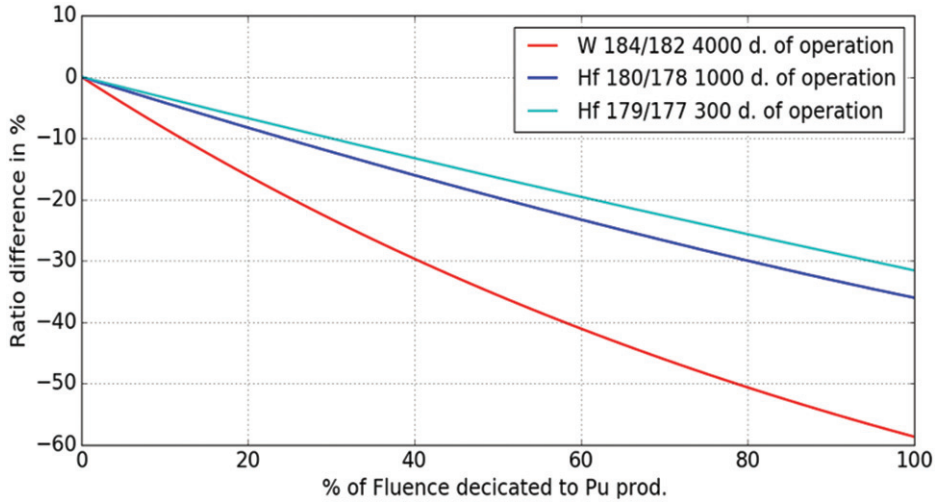

Figure 5. Relative difference in ratios observed between exclusive tritium production and devoting the amount indicated on the *x*-axis to plutonium production.

after 300 days, 1,000 days, and 4,000 days. As expected, the larger the part of fluence dedicated to plutonium, the larger is the difference in the mode ratio.

Figure 6 shows isotope fractions and their ratios' evolution versus fluence for two scenarios. In the first scenario (a), the host declared a 300-day campaign of tritium production but produced plutonium instead. Assuming a total power of 2,400 MW, the 300-day period dedicated to plutonium production enables the host to produce about 626 kg of plutonium

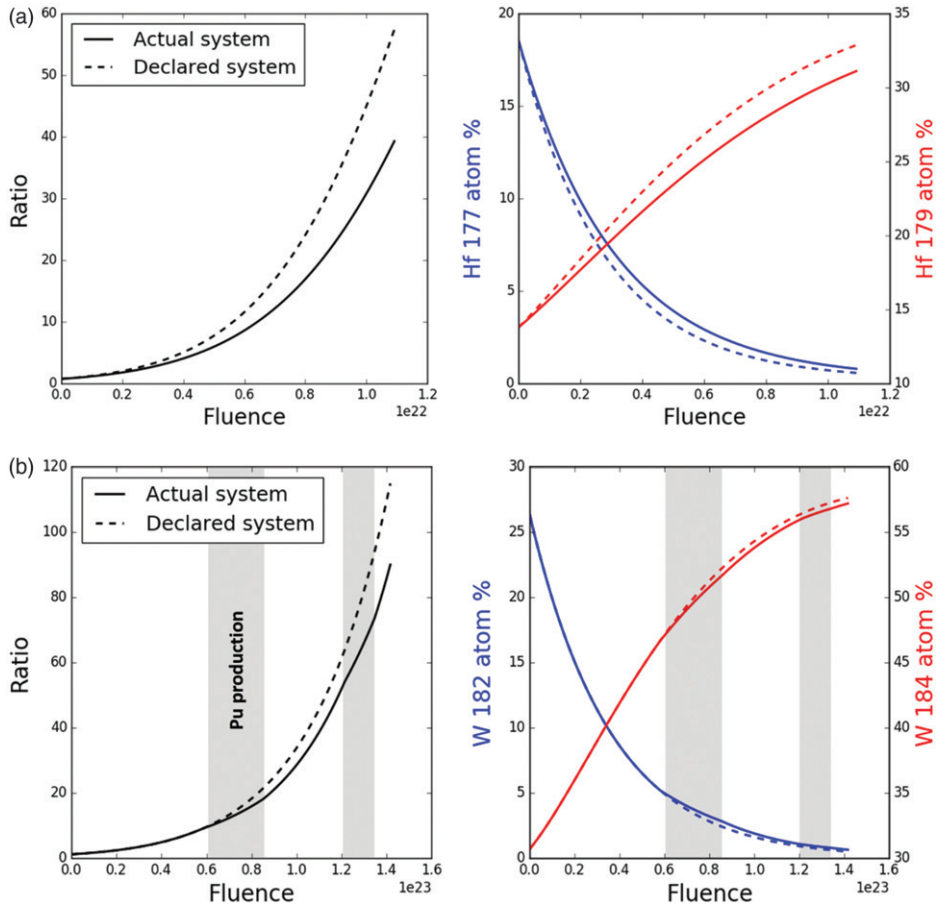

Figure 6. Ratio and isotopic fraction evolutions of (a) hafnium isotopes for a production campaign of 300 days where the host produced plutonium only instead of tritium and (b) tungsten isotopes for a set of campaigns over 4000 days where the host has hidden some plutonium production campaigns (represented by black shades in the graphs).

(95% plutonium-239) with our design characteristics. The mode ratio measured at the end of these 300 days would be 31% smaller than what inspectors would expect. This would constitute a typical scenario in the context of routine verification implemented for countries already part of a treaty. In the second scenario (b), inspectors want to verify the correctness of a declaration covering a production history of 4,000 days (∼11 years). The host declares that only tritium was produced during that time span, but two plutonium production campaigns are hidden between tritium campaigns: one of 800 days and another one of 400 days. Using the same total power of 2,400 MW for the reactor, this corresponds to a total plutonium production of 2.5 tons. Using tungsten isotopes for the mode ratio, inspectors would measure a value that is 21% lower than expected. This scenario could be relevant in the context of a country joining a treaty and where

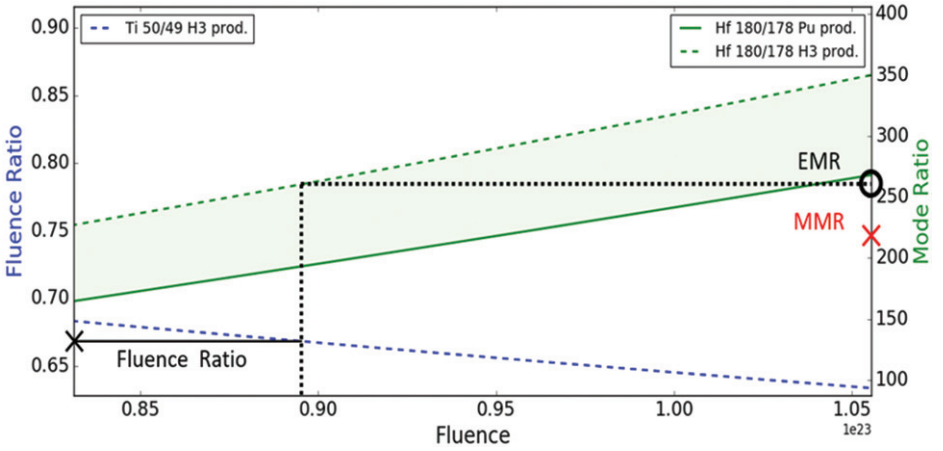

Figure 7. Ratio evolutions for a pure plutonium mode and a pure tritium mode against fluence. The titanium ratio is used as fluence ratio while the hafnium ratios are used as mode ratios. The MMR in this figure is assumed to be sampled in a scenario where plutonium was produced.

inspectors want to confirm the non-production of undeclared plutonium over the past production history of a reactor.

Proposed implementation for inspections

While the previous section dealt with demonstrating the feasibility and the possibilities of using mode ratios to verify a production history, this section proposes an implementation strategy that inspectors could follow in heavy-water reactors.

One should compare all mode ratios at the same fluence level and avoid referencing time, as it would enable the host to adjust the neutron flux and thereby change the measured mode ratios. Using fluence as the reference for all results means that in addition to measuring the mode ratio, the inspectors must measure the fluence of the same material they are sampling. This can be done by using another isotopic ratio, here referred to as the fluence ratio, with a low dependence on production modes (e.g., titanium-50/titanium-49). The following points summarize the process of measuring and comparing ratios:

- First, the inspectors calculate the evolution of the expected fluence ratio (EFR) and the expected mode ratio (EMR) against fluence using information on the reactors' parameters that correspond to the declared campaign. The dashed blue and green curves respectively represent these two evolutions as in Figure 7. These curves will serve as reference ratio evolutions for the inspectors. The solid green curve shows the evolution of the mode ratio for a plutonium-only production. It only serves

as a landmark to know in what direction the mode ratio will tend to move when plutonium is produced.

- Second, inspectors obtain the measured fluence ratio (MFR) to deduce the expected fluence using the EFR evolution curve. Once the fluence is known, they can identify the expected EMR from the curve. The black, dashed line in Figure 7 represents this step.
- Third, the inspectors obtain the measured mode ratio (MMR), indicated by a red cross in Figure 7, compare it with the EMR and conclude whether it agrees with the production history as declared by the host.

These three steps are plagued with uncertainties. During the neutronics simulation, various approximations in the model and uncertainties on input parameters will create uncertainties on the EFR and the EMR evolution curves. These include nuclear data uncertainties, reactor design and operation uncertainties, physical model approximations, or statistical and computational uncertainties. Uncertainties will also exist when the inspectors obtain the MFR and the MMR because of the inherent uncertainties of the technology used to measure these ratios (such as those encountered for mass spectrometry) and because of uncertainties on the exact location of the sample used to make these measurements.[35] It is critical to consider these uncertainties when assessing the conformity or the violation of a declared mode of production. Specifically, we can use a hypothesis testing method where we accept or reject the hypothesis that the declaration is correct based on the MMR. Similarly, given a robust assessment of the involved uncertainties and by assessing false positives and false negatives, one can assess how much undeclared plutonium production is possible without being able to reliably reject the hypothesis of a correct declaration.

Another important aspect of these inspections is the frequency of implementation. In fact, the time scope of an inspection is in part constrained by the technical feasibilities of nuclear archaeology. As seen in Figure 4, mode ratios can be used only for specific fluence ranges. To convert this to a time range necessitates knowing the specific power of the reactor and so time range of ratios must be calculated on a case-by-case basis. For example, using the specific power of production reactors like those of the SRP, the use of isotopes of hafnium could be used for annual inspections as their associated time range is about a year to a few years. Ratios of tungsten, on the other hand, are useful only at very high fluence. For the specific powers of the SRP reactors, the time range of usability of tungsten is around 6 years and more. Tungsten could be used in a context where a country accepts undergoing inspections after a decade of no inspections as illustrated in Figure 6. In addition, the sampling procedure is also a deterministic factor that frames inspection schedules. Structural material can

only be sampled if discharged or when the reactor is shut down. Concerning monitor tags, some designs would allow for withdrawal at any time but since it is still in a conceptual stage, more research needs to be done to determine the modalities of monitor tag sampling procedures.[36]

Conclusion and discussion

This paper has presented a concept that uses techniques from nuclear archaeology to develop a method to identify and distinguish between plutonium and tritium production modes in heavy-water reactors. The technique relies on the fact that different production modes will result in characteristic neutron spectra, which produce differences in certain isotopic ratios present in structural materials or a monitor tag. It may be possible to use this technique to verify the correctness and completeness of a country's declared past plutonium production, or it can be implemented to confirm the absence of undeclared plutonium production under a Fissile Material Cutoff Treaty.

This paper is mainly conceptual and does not develop or recommend a specific sampling procedure (structural material or monitor-tag sampling). Further research needs to examine the limitations imposed by each of these sampling procedures. This is especially true for structural material sampling. First, to what extent permanent or semi-permanent materials are available for specific reactor designs needs to be examined. One cannot rely on materials that are frequently discharged. Furthermore, some elements may not be present in adequate concentrations in structural materials. Typical cladding materials such as zircalloy contain traces of hafnium and tungsten, but aluminum does not. Results for other common trace elements (titanium, iron, chromium, nickel, and strontium), some of which are contained in stainless steel, were less promising than results for hafnium or tungsten, but it may still be possible to use these elements in certain situations (verification of production history).[37]

Another issue that needs to be studied concerns the spatial dependence of the neutron flux spectrum. Because the spectrum varies across the reactor core, the mode ratio will depend on where the sampling is done. Inspectors therefore need to know the sampling locations to calculate the expected mode ratios for all relevant locations in the reactor. If inspectors are unable to determine the sampling location, for example if they measure ratios from a discharged assembly's outer sleeve, the method can be severely limited as the local neutron spectrum will not be known with sufficient accuracy. Furthermore, uncertainties in the design and operational parameters of a specific reactor will result in uncertainties in the expected isotopic ratios for different production modes. Therefore, it must be

examined how accurately reactor data need to be known to enable forward-simulations that are sufficiently accurate to translate measured ratios to production modes. Specifically, a robust uncertainty assessment is required to examine which production scenarios can reliably be distinguished.

Finally, it is also important to verify that the results obtained in this work can be generalized to other designs of heavy water-moderated production reactors and to other types of production reactors. Part of the trends observed here may be specific to the designs used in this paper, others may be more general. As explained previously, the spectra differences stem from geometry differences as well as from composition differences in the two production charges. The method presented in this work should first be tested on designs like the Savannah River Plant heavy-water reactors, the LF-2/Ludmila reactor in Russia, or the Celestin I and II in France. Further works could also consider other types of production reactors such as the graphite-moderated reactors used in North Korea today.

Acknowledgments

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Notes

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Appendix A: Calculation of France's annual tritium production

This appendix estimates France's annual tritium production in the RES reactor based on the expected excess reactivity of the core and the corresponding number of neutrons available for capture in breeding targets. To estimate the excess reactivity of the RES reactor, we calculate the infinite multiplication factor using the standard four-factor formula:

$$k_{inf} = \varepsilon p f \eta_T$$

with ε the fast fission factor, p the resonance escape probability, f the thermal utilization factor, and η_T the reproduction factor. We assume that the neutronics of the RES reactor are similar to those of a standard LWR. Nevertheless, since the fuel of the RES is more enriched (6% vs 3–5%), we adjust the f and η_T factors in the four-factor formula using parameter values listed in Tables A-1 and A-2.

Table A-1. Fuel parameters for 6% enrichment.

Fuel parameters for 6% enrichment	
Fuel density	2.34E + 28 atoms m^{-3}
Fuel volume fraction (V^{fuel})	0.376
Fuel fission cross-section ($\sigma^{fuel}_{T,\ f}$)	35.1 barns
Fuel capture cross-section ($\sigma^{fuel}_{T,\ \gamma}$)	8.40 barns
Fuel nubar (ν)	2.44

Table A-2. Other material parameters.

Other material parameters	
Cladding density	4.31E + 28 atoms m^{-3}
Cladding volume fraction (V^{clad})	0.088
Cladding capture cross-section	0.191 barns
Moderator density	3.35E + 28 atoms m^{-3}
Moderator volume fraction (V^{mod})	0.535
Moderator capture cross-sections	0.322 barns

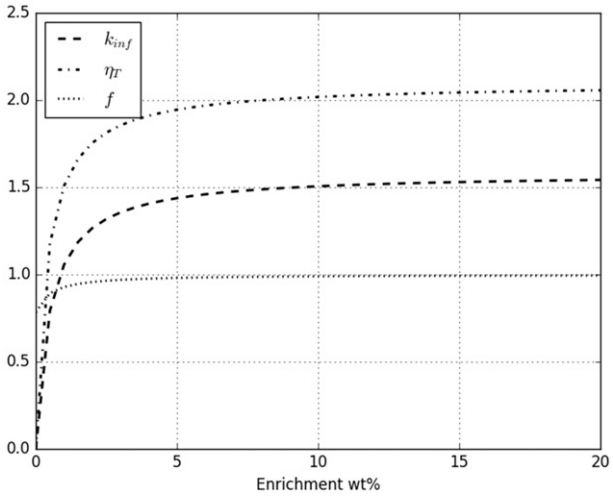

Figure A-1. Evolution of k_{inf}, η_T and f against enrichment in U-235.

The thermal utilization has the following expression:

$$f = \frac{V^{fuel}\left(\Sigma^{fuel}_{T,\ f} + \Sigma^{fuel}_{T,\ \gamma}\right)}{V^{fuel}\left(\Sigma^{fuel}_{T,\ f} + \Sigma^{fuel}_{T,\ \gamma}\right) + V^{mod}\Sigma^{mod}_{T,\ \gamma} + V^{clad}\Sigma^{clad}_{T,\ \gamma}}$$

Using the parameters from Tables A-1 and A-2, we find:

$$f = \frac{0.376 \cdot (35.1 + 8.40) \cdot 2.34}{0.376 \cdot (35.1 + 8.4) \cdot 2.34 + \ 0.535 \cdot 0.322 \cdot 3.35 + 0.088 \cdot 0.191 \ \cdot 4.31} = 0.983$$

Table A-3. Physics parameters.

Physics parameters	
Mev to J conversion factor	1.602E−13 J. Mev^{-1}
Fission energy	200 Mev
Avogadro number	6.022E + 23
Tritium molar mass (M_{H3})	3.00 g. $mole^{-1}$

Table A-4. Reactor parameters.

Reactor parameters	
Power	150 MW
Enrichment (U-235)	6.00%
Capacity factor	75.00%

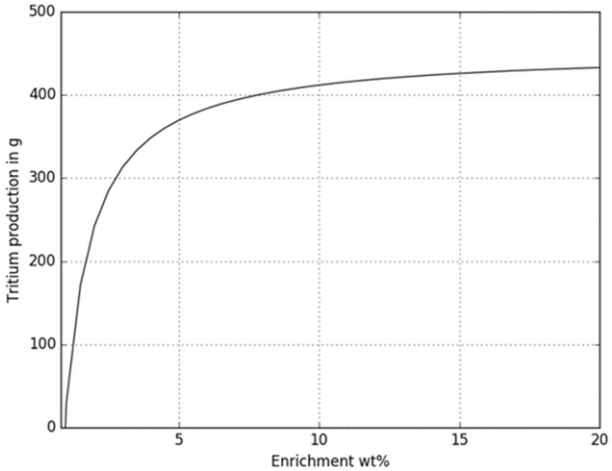

Figure A-2. Annual tritium production in the RES against enrichment in uranium-235.

Next, we calculate η_T:

$$\eta_T = \frac{\nu\sigma^{fuel}_{T,\ f}}{\sigma^{fuel}_{T,\ f} + \sigma^{fuel}_{T,\ \gamma}} = \frac{2.44 \cdot 35.1}{35.1 + 8.40} = 1.97$$

The ε and p factors are less sensitive to fuel enrichment, and we can use values that are typical for light-water reactors, $\varepsilon = 1.03$ and $p = 0.732$. We therefore find:

$k_{inf} = 1.03 \cdot 0.732 \cdot \ 0.983 \cdot 1.97 = 1.46$

Figure A-1 shows how k_{inf}, η_T and f change with enrichment.

The reactivity ρ is equal to $\frac{k_{inf}-1}{k_{inf}} = \frac{1.46-1}{1.46} = 0.315 = 31.5\%$

In a real reactor, neutron leakage from the core will reduce this excess reactivity by several percent. Here, we assume a total loss of 3%, reducing the excess reactivity to 28.5%. For a reactor to operate in a stable manner, ρ has to be zero, and any excess reactivity would usually be absorbed by burnable poisons or control rods. For simplicity, here, we assume that available excess reactivity is absorbed in lithium targets. By calculating the

number of neutrons created per year, we can then find how much tritium is produced per year.

For a reactor power level of 150 MW thermal and the parameter values from Tables A-3 and A-4, the number of fission events per second is:

$$fission\ rate = \frac{150 \times 10^6\ J/s}{200 \cdot 1.602 \times 10^{-13} J} = 4.68 \times 10^{18}\ s^{-1}$$

The number of neutrons produced per second is then:

$$neutron\ rate = fission\ rate\ \cdot \nu = 4.68 \times 10^{18} \cdot\ 2.44 = 1.14\ \times 10^{19}\ s^{-1}$$

Assuming a capacity factor of 75%, the number of neutrons produced per year is:

$$1.14\ \times 10^{19} s^{-1} \cdot 0.75 \cdot 365 \cdot 24 \cdot 3600\, s = 2.69 \times 10^{26}$$

Of these neutrons, 28.5% are available for tritium production:

$$2.69 \times 10^{26} \cdot 0.285 = 7.67 \times 10^{25} = 127\ moles$$

Based on our assumption, each of these neutrons is absorbed in a lithium target and produces exactly one tritium nucleus. The atomic mass of tritium is about 3.0 grams per mole, and the resulting tritium production rate is therefore 381 grams per year. This number should be considered an upper limit.

Figures A-2 shows how annual tritium production varies with fuel enrichment.

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