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Isotopic Signatures of Weapon-Grade Plutonium from Dedicated Natural Uranium–Fueled Production Reactors and Their Relevance for Nuclear Forensic Analysis

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Abstract – *We report neutronics calculations for the most important natural uranium–fueled reactor types historically used for weapons plutonium production. These include an early design of the Hanford-type graphite-moderated and light-water-cooled reactor used in the United States; the Calder Hall–type graphite-moderated and gas-cooled reactor used in the United Kingdom; and the NRX-type heavy-water-moderated and light-water-cooled reactor, originally developed in Canada for civilian purposes but later used in India and Pakistan for military plutonium production. We show that while it is possible in principle to identify with a high level of confidence weapon-grade plutonium compositions produced in other types of reactors, e.g., light-water-cooled or fast neutron reactors, it is difficult to distinguish among plutonium compositions generated in dedicated production reactors fueled with natural uranium. This suggests that efforts to determine the origin of weapon-grade plutonium for a nuclear forensic analysis could well remain inconclusive without access to databases based on actual samples of the nuclear material.*

I. INTRODUCTION

This paper gives an overview of isotopic signatures that can be expected for weapon-grade plutonium compositions (previous results of this research are published in Ref. 1). The purpose of this analysis is to quantify the range of isotopic variations and, ultimately, to understand the relative importance of *predictive* versus *empirical* isotopic signatures—both of which are relevant for nuclear forensic analysis.

The results presented below are primarily based on neutronics calculations for common reactor types used for plutonium production. We do not discuss the age of the material or the time since last purification as indicators. These characteristics could be determined using chronometric methods without resolving ambiguities regarding the origins of the material.[a]

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II. PLUTONIUM PRODUCTION REACTOR TYPES

Plutonium isotopics are determined primarily by the burnup of the uranium fuel in which the plutonium was originally produced. In general, however, the isotopics also depend on the reactor type and the operating history of the reactor. Therefore, even if two plutonium samples

[a]Plutonium isotopics and concentration of other trace nuclides can change significantly with the age of the material, primarily due to the decay of ^{241}Pu, which has a half-life of only 14.4 yr. The resulting ^{241}Am/^{241}Pu ratio is a good indicator of the time that has elapsed since the material was last purified.

TABLE I

Natural Uranium–Fueled Plutonium Production Reactors by Category, Representative but Incomplete List of Facilities

	Graphite Moderated		Heavy Water Moderated	
	H_2O Cooled	Gas Cooled	H_2O Cooled	D_2O Cooled
United States	Hanford	—	—	—
Russia	Tomsk-7	—	—	—
United Kingdom	—	Calder Hall	—	—
France	—	G-Series	—	—
China	Jiuquan	—	—	—
Israel	—	—	—	Dimona
India	—	—	Cirus/NRX	Dhruva
Pakistan	—	—	Khushab[a]	—
North Korea	—	Yongbyon	—	—

[a]Light water cooling of the Khushab reactor, i.e., its similarity to the Indian Cirus reactor, has been reported but is unconfirmed.[3]

are similar in one respect—for example, they may both have the same percentage of ^{239}Pu—their origin may still be identified based on an analysis of selected isotope ratios.

Virtually any reactor type can be used for the production of weapon-grade plutonium by limiting the burnup of the uranium fuel, but plutonium production is maximized in natural uranium–fueled reactors. High-purity graphite or heavy water has to be used with natural uranium fuel for moderation to minimize parasitic neutron absorption. Reactor designs that permit continuous refueling are preferred for dedicated production reactors in order to facilitate frequent discharge and reloading of fuel elements for extraction of the plutonium. Table I lists the main reactor types that have been or are being used for dedicated plutonium production.[2]

As shown in Table I, graphite-moderated designs have played the dominant role in early nuclear weapon programs, in particular in those countries that later became the weapon states of the Non-Proliferation Treaty. Heavy-water-moderated reactors were built in Israel, India, and Pakistan. We have carried out infinite-lattice neutronics calculations for the three most important types of production reactors. Their unit cells are illustrated in Fig. 1.

II.A. Hanford Type

In total, nine reactors were operated at the Hanford site between 1944 and 1987. These reactors were all graphite moderated and light water cooled, but their designs evolved over time. Also, the power levels of all Hanford reactors increased significantly to accelerate plutonium production.[b] We modeled the first and conceptually most simple design, the B Reactor, in which 2004 process tubes, each 8.5 m long, penetrate the reactor core. Additional design information is listed in Table II (Refs. 4 and 5). The core contains ~300 tonnes of uranium and has an average power density in the fuel uranium of ~20 W/cm^3 for the initial power level of 250 MW(thermal). The production reactors used in Russia and China were reportedly virtually identical to the original early U.S. reactors operated at the Hanford site.[6][c] We do not attempt to model these reactors here separately.

II.B. NRX Type

The NRX is a heavy-water-moderated and light-water-cooled reactor, originally designed as a plutonium-production reactor but used in Canada for civilian purposes.[7] The original NRX reactor went critical in July 1947 with a power level of 20 MW, which was later increased to 40 MW and finally to 42 MW. As shown in Fig. 1, it uses a hexagonal lattice. The total uranium inventory in the core is ~9.5 tonnes (54 kg/fuel assembly).[8] For a reference power level of 40 MW(thermal), the power density in the fuel is ~80 W/cm^3. As part of the Atoms for Peace Program, India obtained a copy of the NRX reactor (CIRUS) (Ref. 9), which went critical in 1960 and became fully operational in 1963. Reportedly, Pakistan's 50-MW Khushab-I reactor, which has been operational since 1998, is also similar to NRX/CIRUS. Pakistan is currently building two additional plutonium production

[b]For example, the power levels of the B, D, and F Reactors were increased from 250 MW to eventually >2000 MW(thermal). The production reactors at Hanford evolved into the Hanford N Reactor, a design that optimized plutonium production concurrent with 860 MW(electric).

[c]In contrast to the early Hanford reactors, the Russian reactor variant used highly enriched (90% ^{235}U) spike fuel for radial and axial power flattening. However, >96% of the core inventory is standard natural uranium fuel. Similarly, Hanford reactors used slightly enriched fuel in the radial spike ring to flatten flux distributions.

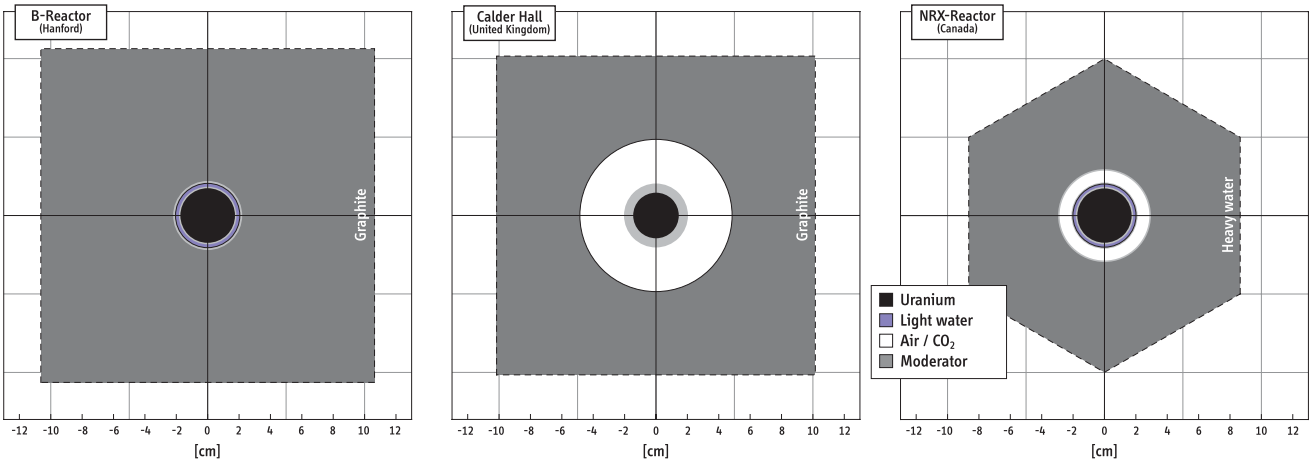

Fig. 1. Reactor geometries of three important types of natural uranium–fueled plutonium production reactors. These models are used for all MCNP simulations and burnup calculations. Shown are the unit cells with the fuel, coolant, and moderator. The Hanford-type and Calder Hall–type reactors are graphite moderated, but one is light water cooled, and one is gas cooled. The NRX-type reactor is heavy water moderated and light water cooled.

TABLE II

Basic Reactor Design Data

	Hanford B	Tomsk-7	NRX	Calder Hall
Uranium slug diameter (cm)	3.48	3.40	3.46	2.92
Cladding thickness (cm)	0.09	0.10	0.10	0.58
Width of cooling channel (cm)	0.22	0.20	0.18	2.81
Flow tube diameter (cm)	4.10	4.00	4.22	9.70
Aluminum tube thickness (cm)	0.15	0.16	0.10	—
Lattice pitch (cm)	21.27	20.00	17.30	20.32

reactors, Khushab-II and Khushab-III, at the same site. Their design power levels are unknown, but they too seem to be similar to the original Canadian design.[10,11] All these reactors in India and Pakistan are or will be used for military production of weapon-grade plutonium.

II.C. Calder Hall Type

In the 1950s, the United Kingdom built numerous dual-use facilities that were used for both electricity and military plutonium production. These reactors were graphite moderated and gas cooled and used a unique cladding material (magnesium nonoxidizing or magnox alloy). The power levels of the four Calder Hall reactors varied between 180 and 240 MW(thermal) [or 42 to 54 MW(electric)] (Ref. 12; for additional information see also Ref. 2, pp. 59–66). The core inventory is on the order of 100 tonnes of uranium, which corresponds to an average power density of 40 W/cm^3 in the fuel. North Korea later adapted the Calder Hall design, apparently based on information published in the 1950s, and built a downsized [20- to 25-MW(thermal)] version of the reactor in the 1980s. This production reactor is now closed down, and its cooling tower was demolished in June 2008. A larger Calder Hall–type reactor at the Yongbyon site was never completed.

All calculations below have been carried out with the MCODE computer code system[13–15d] linking the Monte Carlo particle transport code MCNP and the ORIGEN2 general point-depletion code.[16–18] Most importantly, MCODE regularly updates the fuel composition in MCNP, which is used to determine spectrum-averaged one-group cross-section data for all relevant nuclides. MCODE uses an optimized averaging technique based on the predictor-corrector method to improve the accuracy of the burnup calculations. For a given nuclide composition or burnup zone, a first transition matrix is generated for time t_n with the effective cross-section and neutron-flux data calculated by MCNP. These data are used by ORIGEN2 to calculate an estimated nuclide composition for the end of the time step t_{n+1} (predictor run).

Based on a second MCNP run for this new material composition, a second transition matrix is generated and used in a consecutive ORIGEN2 calculation, starting with the original nuclide composition, to obtain a second composition for t_{n+1} (corrector run). The final nuclide composition, i.e., the composition to be used as the initial composition for the next burnup step, is obtained in

[d]MCODE is used with the kind permission by its author, Zhiwen Xu.

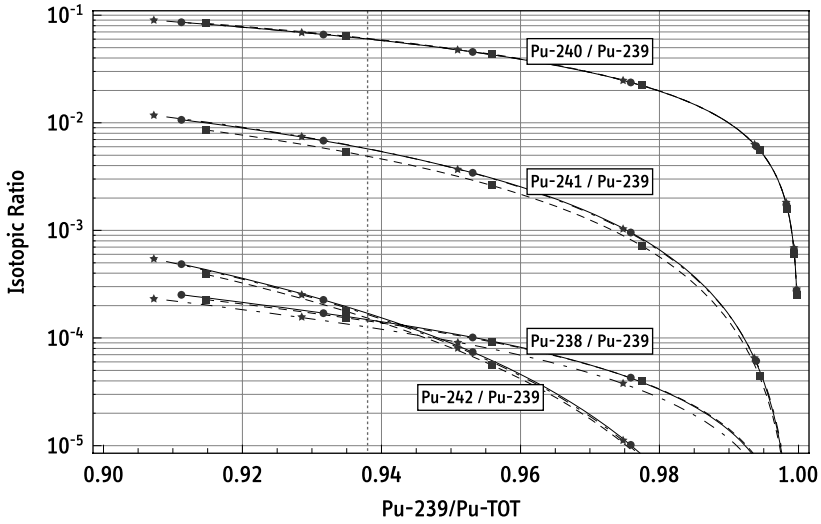

Fig. 2. Isotopic ratios for various plutonium isotopes as a function of ^{239}Pu content. The compositions of Hanford-type (solid line, circles), NRX-type (dashed line, squares), and Calder Hall–type plutonium (dot-dashed line, stars) are shown. Results are based on MCODE simulations. The power density in the fuel was 40 W/cm^3 in all cases. The dashed vertical line indicates the target condition used to define weapon-grade plutonium (93.8 wt% ^{239}Pu) in this paper.

taking the average number densities of both runs for all nuclides tracked explicitly with MCNP. Previous analyses have shown that this approach is more effective than those used by other burnup codes, such as MOCUP and MONTEBURNS, and provides most accurate results for a given total number of MCNP calculations.[14]

In order to avoid isotopic dissimilarities that could result from the value of the power density alone, we have selected a default power density of 40 W/cm^3 for the three reactor types—a value that is compatible with all reactor designs considered here. Given that the neutron spectra are very thermal and that the uranium-rod diameters are rather similar for all designs (2.9 to 3.5 cm), we cannot expect large differences in the isotopics of the plutonium built up in the fuel.

Historically, because of uranium-availability constraints, several nuclear weapon states reportedly have recycled irradiated uranium, after extraction of the plutonium, and used it to fabricate fuel for a second (or third) plutonium production run. In addition to the reference calculations, where natural uranium is used for the fresh fuel, we therefore also analyze the impact of this uranium-recycling strategy on the isotopics of the plutonium discharged from the modeled reactors.

III. RESULTS

Figure 2 shows the main results of the neutronics calculations for selected isotopic ratios as a function of ^{239}Pu content. Even for increasing fuel burnup and, thus, decreasing ^{239}Pu fraction in the material, the compositions are remarkably similar, which is due to the similarity of important effective cross-section values and ratios. Spectrum-averaged one-group cross sections for the main uranium and plutonium isotopes, as calculated by MCNP, are listed in Table III. For reference purposes, Table III also includes data for a typical pressurized water reactor (PWR), as well as effective plutonium production rates for the various reactor types.

A manifold of isotopic ratios is available to characterize plutonium compositions and samples. Below, we will discuss those plutonium ratios that are most relevant for nuclear forensic analysis.

Figure 3 shows isotopic correlations for two selected ratios, ^{242}Pu/^{240}Pu versus ^{238}Pu/^{TOT}Pu, identified by Mayer, Wallenius, and Ray as one of the most characteristic signature combinations.[20] Specifically, the ^{238}Pu/^{TOT}Pu ratio is an indicator of the hardness of the neutron spectrum, whereas the ^{242}Pu/^{240}Pu ratio is a measure of exposure or burnup.

The empirical data shown are based on mass-spectrometric analyses of plutonium samples from diverse origins. Most of these samples, however, correspond to high-burnup fuels (^{240}Pu $> 10\%$) and may be less relevant for a forensic analysis, in which weapon-grade plutonium might be intercepted or recovered. To assess the capabilities of nuclear forensic analysis in this situation, we compared plutonium compositions that are identical in one important aspect, making an analysis more challenging, but also more meaningful. These compositions are characterized by a selected reference value of

TABLE III

Selected Spectrum-Averaged One-Group Cross Sections as Calculated by MCNP*

Cross Section	Hanford		NRX/Cirus		Calder Hall		PWR (for reference only)	
	BOL	EOL	BOL	EOL	BOL	EOL	BOL	EOL
^{235}U (b)								
Capture	23	22	29	28	25	24	7.9	7.6
Fission	128	121	162	156	137	129	33.1	31.3
^{238}U (b)								
Capture	0.91	0.88	1.05	1.02	0.99	0.96	0.83	0.83
Fission	0.07	0.08	0.07	0.08	0.06	0.07	0.10	0.10
^{239}Pu (b)								
Capture	107	96	124	115	119	107	54.4	49.3
Fission	226	206	271	254	248	227	93.4	85.1
^{240}Pu (b)								
Capture	271	217	267	228	297	244	230	210
Fission	0.48	0.48	0.46	0.47	0.44	0.44	0.61	0.61
Plutonium production rate (g/MWd)	0.83		0.77		0.85		0.48	

*Listed are values for beginning of life (BOL) (after one effective full-power day) and end of life (EOL), which is reached in our simulations when the ^{239}Pu fraction has dropped to 93.8 wt%. Effective plutonium production rates have been determined with MCODE neutronics calculations and are listed for reference purposes only. The continuous-energy cross-section data for the MCNP libraries are based on ENDF/B-VI Release 5 data [MCLIB-E6, NEA-1651/001 (Ref. 19)].

93.8 wt% for the isotope ^{239}Pu.[e] Again, Fig. 3 illustrates the main results, and Table IV lists the plutonium isotopics in the fuel of the modeled reactors, when this target condition is reached.[f]

III.A. Effects of Uranium Recycle

The burnup of the fuel for the target ^{239}Pu content of 93.8 wt% is on the order of only 1200 MWd/t HM. At this level, the uranium recovered from the spent fuel still contains ~0.6% of the isotope ^{235}U. As mentioned, nuclear weapon states have sought to maximize the use of uranium resources for fissile material production and reportedly used irradiated uranium in consecutive plutonium production runs.

The impact of this strategy on the plutonium isotopics is shown in Fig. 4. The results are based on MCODE neutronics calculations using uranium recovered from the respective reactors from the first production cycle.

Comparatively high ^{238}Pu contents are the most significant feature of plutonium produced with recycled uranium. For all reactor types modeled here, the ^{238}Pu increases by ~25% (to >0.015% in all cases, with ^{239}Pu at 93.8 wt%), which itself could be sufficient to conclude that this specific production strategy has been pursued. The elevated ^{238}Pu content is a result of two consecutive neutron captures in ^{236}U and ^{237}Np, facilitated by the initial ^{236}U content in the recycled uranium. The results, however, also show that the characteristic differences in the isotopic ratios do *not* increase when recycled uranium is used.

Figure 4 provides representative error margins for predictive isotopic signatures obtained with neutronics calculations. To evaluate the impact of variations in the parameters of the models or operating conditions, we added a low concentration of natural boron in the moderator to simulate the presence of control rods and—in separate simulations—also varied the power density in the fuel. As shown in Fig. 4, allowing for these modifications adds uncertainties in the isotopic signatures that are on the order of ±5% and would further complicate an evaluation of the data. Especially in the case of recycled uranium, in which the uncertainties and errors would propagate from the first to the second run, the plutonium compositions recovered from the three reactors would not be distinguishable based on predictive signatures obtained with neutronics calculations alone.

[e] Different countries have defined weapon-grade plutonium differently, i.e., targeted a preferred fuel burnup and ^{239}Pu content, balancing weapon-relevant characteristics and availability of uranium ore. Here, we assume that prior information on the expected isotopics of the plutonium, which could point to the origin of the material, is unavailable.

[f] As shown in Table IV, the ^{241}Pu/^{TOT}Pu ratio appears as another potential signature. The short half-life of the isotope, however, would require a separate age determination and introduce additional uncertainties for a nuclear forensic analysis seeking to identify a particular reactor type or facility.

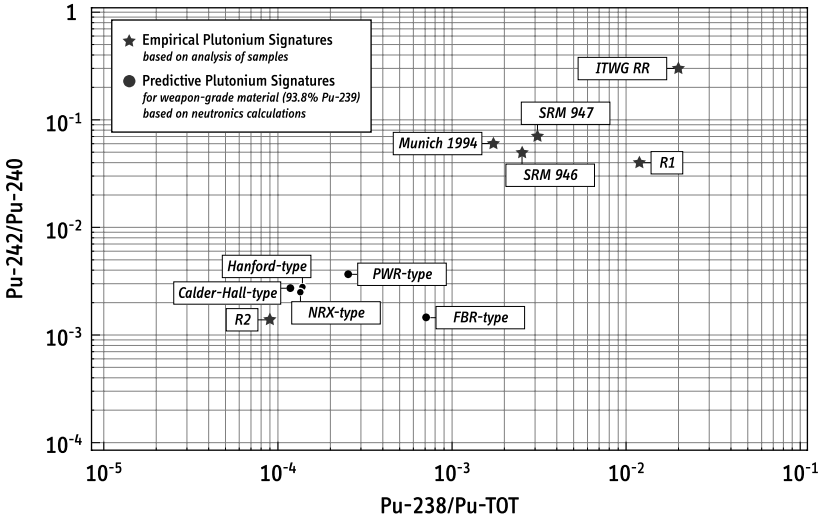

Fig. 3. Isotopic ratios for selected plutonium compositions. Most data points obtained for actual plutonium samples (*) correspond to high-burnup fuels. Weapon-grade plutonium compositions (•) are based on MCODE neutronics calculations for typical operating conditions. Some reactor types are easy to distinguish, e.g., light water from fast neutron reactors, but dedicated production reactors are not. Sample data (*) from Mayer, Wallenius, and Ray.[20]

TABLE IV

Weapon-Grade (93.8 wt%) Plutonium Compositions in the Fuel of Various Reactor Types at Time of Discharge*

	Hanford	NRX/Cirus	Calder Hall	PWR (for reference only)
^{238}Pu (%)	0.014	0.013	0.012	0.025
^{239}Pu (%)	93.80	93.80	93.80	93.80
^{240}Pu (%)	5.63	5.71	5.63	5.23
^{241}Pu (%)	0.54	0.46	0.54	0.93
^{242}Pu (%)	0.016	0.014	0.015	0.018
Target burnup (for reference only) (MWd/t)	1174	1241	1118	2523

*All data from MCODE neutronics calculations. PWR values are for reference purposes only. Total may not add up to 100% due to rounding.

Overall, the data in Figs. 3 and 4 show that it is possible to distinguish with a high level of confidence weapon-grade plutonium compositions from some basic reactor types. These include fast breeder reactors, light water reactors using low-enriched fuel, and reactors fueled with natural uranium. It is, however, extremely difficult to distinguish among plutonium compositions that were generated in dedicated production reactors fueled with natural uranium if reasonable uncertainties are taken into account for the reactor model and the mode of operation. A nuclear forensic analysis based on predictive signatures alone, i.e., without access to actual samples, could well remain inconclusive in this case. An analysis based on samples is likely to be more conclusive because these would reveal unique features of the material caused by a priori unknown specifications, e.g., the target burnup set by the operator, or other details of the production process.

IV. CONCLUSION

Historically, only a few reactor types have been used to produce weapon-grade plutonium. Moreover, nearly identical designs were sometimes used by more than one country. As a result, isotopic signatures of plutonium isotopics are generally weak identifiers. For this study, we have carried out neutronics calculations for the most important types of dedicated production

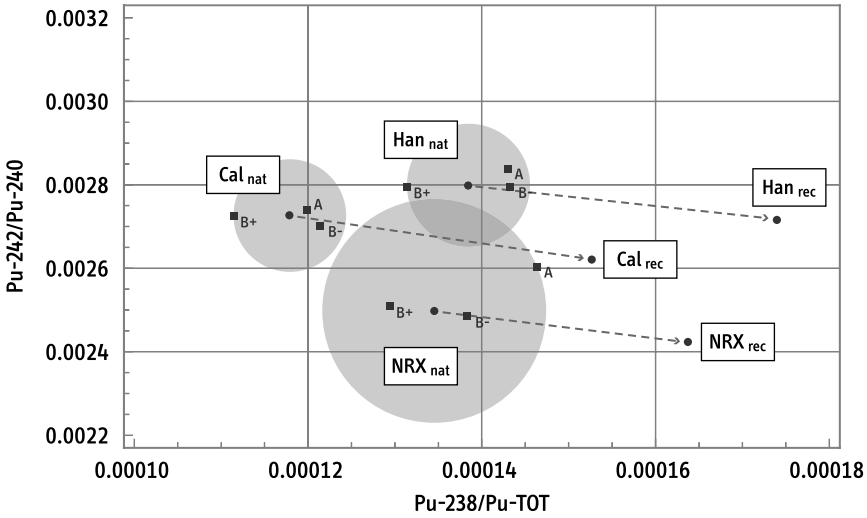

Fig. 4. Impact of using natural versus recycled uranium to fuel different types of production reactors on selected isotopic ratios. The initial ^{236}U content in the recycled uranium facilitates ^{238}Pu buildup in the fuel. The shaded areas indicate error margins that can be expected due to uncertainties in the input or model parameters: presence of neutron absorbers (A), reduced ($\times\frac{1}{2}$, B−) and increased ($\times 2$, B+) power densities. All results are for a target concentration of ^{239}Pu of 93.8% and based on MCODE neutronics calculations using MCNP cross-section data from ENDF/B-VI Release, MCLIB-E6, NEA-1651/001 (Ref. 19).

reactors, generating consistent results for an intercomparison of plutonium compositions. For a given burnup or ^{239}Pu fraction in the material, we find that isotope ratios (such as ^{242}Pu/^{240}Pu or ^{238}Pu/^{TOT}Pu) can vary by up to 20 to 30% between the modeled reactors, but these differences may be too small to determine the origin of a sample if uncertainties in the model and the mode of operation have to be factored in. In other words, considerable additional information has to be available to identify a particular facility based on *predictive signatures*, i.e., based on computer modeling, alone. For this reason, whenever possible, a nuclear forensic analysis seeks to rely on *empirical signatures* obtained from actual samples to perform its task with confidence. The fact that countries generally produced weapon materials under strictly controlled and dissimilar operating conditions has created differences in material properties that nuclear forensics could exploit.

The findings of our analysis therefore reinforce the importance of creating comprehensive databases for nuclear forensic analysis, containing physical samples of nuclear materials, and of establishing a framework for access, control, and use of these data. Such a database could also contribute to the creation of a *global fissile material registry* under future arms-control agreements, which is an idea that has been suggested since the 1990s and that could receive renewed attention in future security debates.

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