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Nuclear Archaeology for Heavy-Water-Moderated Plutonium Production Reactors

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There is growing interest in a set of methods and tools that can be used to characterize past fissile material production activities, using measurements and sampling at production and storage sites. This field has been dubbed "nuclear archaeology." The best-established example of nuclear archaeology relies on measurements of the isotope ratios of selected elements in the graphite of graphite-moderated plutonium production reactors. This Graphite Isotope-Ratio Method (GIRM) determines the cumulative neutron fluence through the graphite and thereby estimates the cumulative plutonium production in the reactor. The great limitation of this particular method is that it can only be applied to graphite-moderated reactors, which represent only one class of reactors that have been used for unsafeguarded plutonium production. In this article, we propose to extend this method to non-graphite moderated reactors by analyzing the evolution of relevant isotope ratios in the support structures and other core components of heavy-water moderated reactors. We present results of neutronics calculations for a generic heavy-moderated reactor evaluating the robustness of the method and explore the role of nuclear archaeology for applications in arms-control treaty verification.

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BACKGROUND

As the nuclear weapon states further reduce the size of their arsenals, declarations of historical fissile material production are likely to become a central part of the nuclear disarmament process. Most weapon states now offer increased transparency with regard to their nuclear programs, and some have already made public their fissile material (and nuclear weapon) inventories.[1] Such declarations are valuable as a confidence-building measure, but their verification will be essential if declarations are to serve as a basis for deep cuts in nuclear arsenals. Verification would partly rely on sampling at former production facilities or direct measurements on selected waste materials in order to determine *ex post facto* the quantity of fissile material an installation might have produced over its lifetime. This field has been dubbed nuclear archaeology.[2] The best-established example of nuclear archaeology relies on measurements of the isotope ratios of naturally occurring trace impurities in the graphite of graphite-moderated plutonium production reactors. The method was developed and demonstrated by Pacific Northwest National Laboratory as part of a research and development effort initiated in the early 1990s.[3]

Graphite-moderated reactors were primarily used in the early weapons programs of the NPT nuclear weapon states. In contrast, heavy-water reactors played more prominent roles in more recent weapons programs (Israel, India, and Pakistan), and they were also part of some other nuclear programs with ambiguous objectives, for example in Sweden and Argentina.[4] Iran's new Arak reactor is another project that has raised international concerns. In this context, it appears critical to explore the potential of nuclear-archaeological methods for heavy-water reactors.[5]

REACTOR MODEL

To examine the applicability and robustness of nuclear archaeology for heavy-water reactors, an adequate test reactor is needed. For this purpose, we have selected Canada's NRX (Figure 1),[6] which was a heavy-water-moderated and light-water-cooled reactor, originally developed for civilian purposes, and, according to the reactor description, optimized for plutonium production.[7]

The original NRX reactor went critical in July 1947 with a thermal power level of 20 MW, later increased to 40–42 MW,[8] and was permanently shut down in 1993.[9] As part of the *Atoms for Peace* Program, India obtained a copy of the NRX reactor, now CIRUS,[10] which went critical in 1960 and became fully operational in 1963. Plutonium produced with this reactor was used for India's nuclear test in 1974. Reportedly, Pakistan's 50 MW Khushab-I reactor is also similar to NRX.[11]

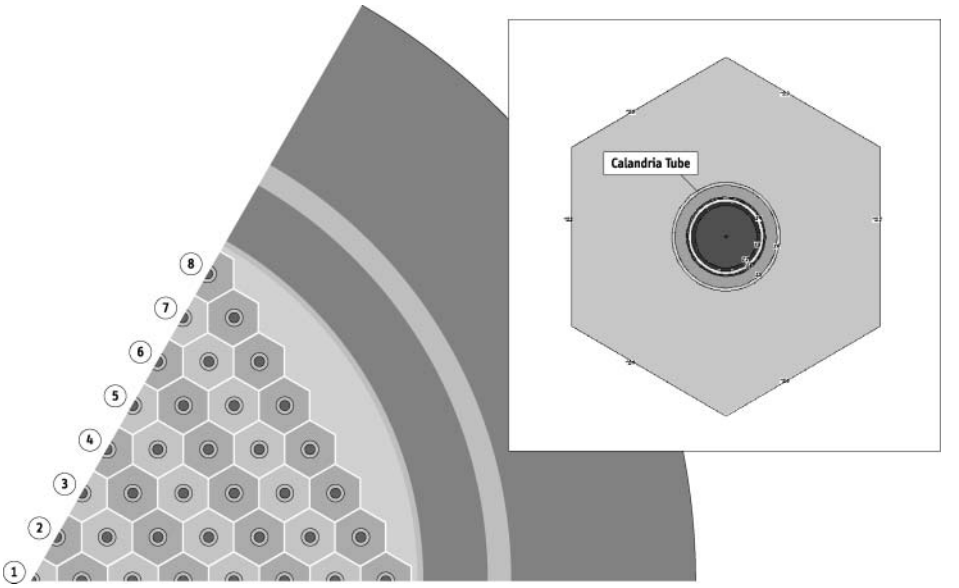

Figure 1: 60-degree core segment and unit cell of the NRX test reactor (MCNP model).

We have carried out extensive infinite-lattice and full-core reactor burnup simulations for NRX/CIRUS in order to determine neutron flux levels, effective (spectrum-averaged) cross sections, and plutonium buildup in the fuel. Results are used to characterize the perturbation of isotope ratios and to develop a sampling strategy that could be tested in the field. All calculations have been carried out with MCODE,[12] which uses the Monte Carlo particle transport code MCNP5 and the general point-depletion code ORIGEN2.[13] MCODE regularly updates the fuel composition in MCNP, which is used to determine spectrum-averaged one-group cross-section data for all relevant nuclides so that ORIGEN2 libraries do not have to be used.

CANDIDATE ISOTOPES

One critical design feature of the NRX, and heavy-water moderated reactors in general, is the so-called "calandria," i.e., a vessel filled with heavy water. Channel tubes penetrate this vessel, hold the fuel rods, and separate coolant from moderator (Figure 1). The calandria is an excellent candidate for sampling because it is in close proximity to the fuel and allows sampling at many different positions in the core.[14] Typical calandria materials for non-power reactors, such as the NRX, are aluminum alloys. Table 1 shows the mass-spectrometric analysis of a historic nuclear-grade aluminum sample,[15] which confirms that many candidate trace-materials can be expected even in high-purity material;

Table 1: Aluminum analysis for a sample of cladding material from the Manhattan College Reactor (MCZPR, US-0119)[15]

Z Element	Content	Z Element	Content	Z Element	Content
5 Boron	2 ppm	23 Vanadium	<100 ppm	29 Copper	1400 ppm
12 Magnesium	<100 ppm	24 Chromium	<100 ppm	30 Zinc	200 ppm
14 Silicon	1900 ppm	26 Iron	5100 ppm	40 Zirconium	<100 ppm
22 Titanium	200 ppm	28 Nickel	<100 ppm	82 Lead	<100 ppm

these are either residual impurities or materials that have been added to improve irradiation behavior and corrosion resistance.

For the present analysis, we consider selected isotope ratios of boron, lithium, chlorine, calcium, titanium, chromium, iron, nickel, zirconium, and lead. This list reflects our pre-experimental understanding, based on inspection of the effective cross sections of relevant nuclides and their likely prevalence in candidate materials. Using results from MCNP simulations of our test reactor, we can solve the set of differential equations for the concentrations of the isotopes in any given isotope system.[16] As an example, Figure 2 (top) shows the perturbation of the natural isotope ratios for the chlorine system over the life of the reactor.

Different types of errors need to be considered when translating a measured isotope ratio α into a fluence value Φ.[17] Most importantly, the relative concentrations of the parent and daughter isotopes determine the error of the fluence estimate. Measurement errors are high initially when the relative concentration of the daughter isotope is small and high again when the isotope ratio is close to an extremum. As a result, every isotope system will have a characteristic fluence range with optimum performance. The critical parameter defining the useful fluence range of a particular isotope system is given by the ratio of the error for the fluence measurement normalized to the magnitude of the fluence measurement itself.

$$\eta_\Phi = \left| \frac{\partial \Phi}{\partial \alpha} \right| \eta_\alpha \rightsquigarrow \eta_{\mathrm{rel}} = \frac{\eta_\Phi}{\Phi} \tag{1}$$

We assume that an isotope ratio should no longer be used as a measurement tool when this relative error is greater than 0.01. Figure 2 (bottom) shows the expected relative errors for various isotope ratios as a function of the neutron fluence in the structural material. The most promising ratios are summarized along with their optimum fluence ranges in Table 2. The focus of this analysis is on aluminum alloys, and typical impurities in these alloys, but the method can also be applied to other types of cladding and reactor-component materials (e.g., zircalloy or stainless steel).

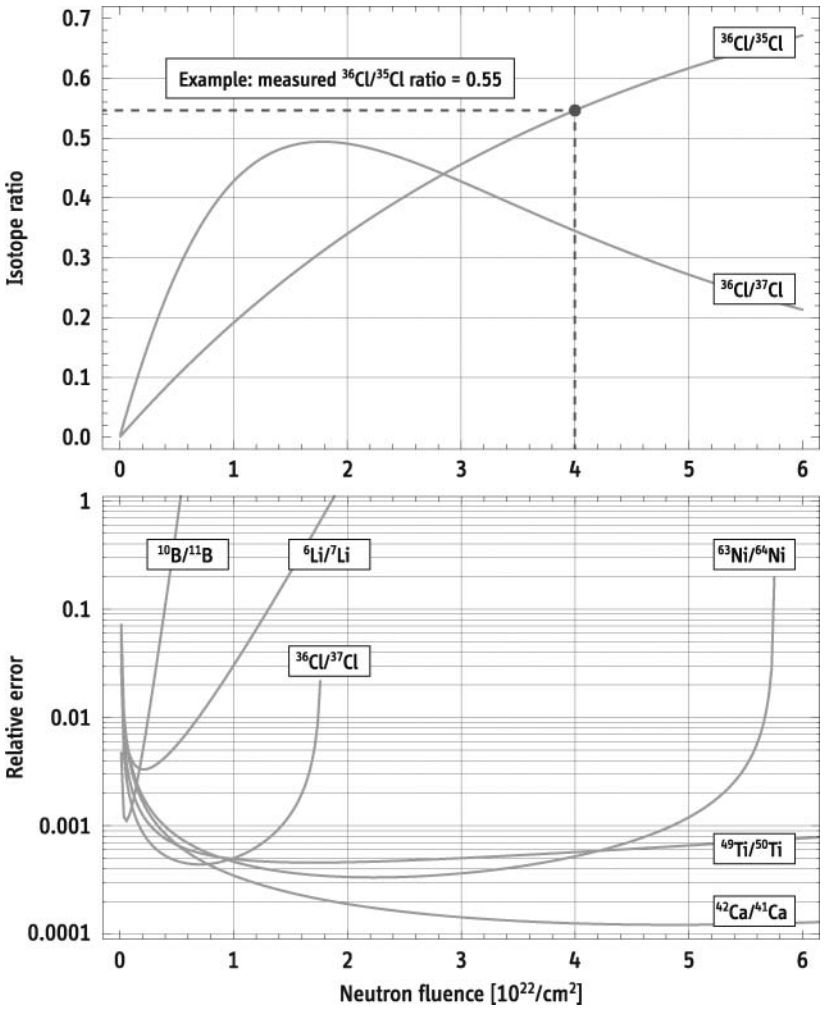

Figure 2: Chlorine isotope ratios as a function of neutron fluence over 40 effective full-power years. Simulated numerically in MATLAB using effective cross sections derived from MCNP simulations (top). Relative neutron fluence errors from mass-spectrometry measurement errors for low- and medium-fluence indicators (bottom). In the example highlighted above, a $^{36}Cl/^{35}Cl$ ratio of 0.55 indicates a neutron fluence of 4.0×10^{22}cm/cm^3.

SAMPLING STRATEGY

We have now established the relationship between isotope ratios and neutron fluence, but still need to develop an understanding of how to go from local fluence (and local plutonium production) to a global plutonium estimate, i.e., the total amount of plutonium produced in the fuel over the lifetime of the reactor. Here, we propose a two-step sampling strategy for obtaining such a plutonium production estimate, minimizing both sampling effort and error. This process

Table 2: Key characteristics of selected isotope ratios for various elements.

		Fluence range	Remarks
Lithium	$^{6}Li/^{7}Li$	Low–Medium	Production of ^{7}Li from ^{10}B
Boron	$^{10}B/^{11}B$	Low	—
Chlorine	$^{36}Cl/^{37}Cl$	Medium	Poor cross-section data for ^{36}Cl
Calcium	$^{42}Ca/^{41}Ca$	Medium–High	Poor cross-section data for ^{41}Ca
Titanium A	$^{48}Ti/^{49}Ti$	High–Super high	—
Titanium B	$^{49}Ti/^{50}Ti$	Medium–High	—
Chromium	$^{54}Cr/^{53}Cr$	High–Super high	Scarcity of ^{54}Cr
Iron	$^{57}Fe/^{56}Fe$	High	—
Nickel A	$^{61}Ni/^{62}Ni$	High	—
Nickel B	$^{63}Ni/^{64}Ni$	Medium–High	Poor cross-section data for ^{63}Ni
Zirconium	$^{91}Zr/^{92}Zr$	High–Super high	Fluence range very high, $> 5 \times 10^{22}$ cm^{-2}
Lead	$^{207}Pb/^{206}Pb$	High–Super high	Fluence range very high, beyond $> 5 \times 10^{22}$ cm^{-2}

varies significantly from other approaches, which typically envision a series of measurements throughout the core, building a global fluence profile, and generating *one* global plutonium estimate therefrom. Here, we will separate these steps and *obtain a plutonium estimate for every fuel channel sampled,* leading to a process that is more robust and possibly easier to implement. This two-step approach is described in more detail below.

From Local Isotope Ratios to Channel-Fluence Estimates

In a first step, we envision acquisition and analysis of samples taken along one fuel channel in order to determine an average fluence estimate $\bar{\Phi}$ for this particular channel. If the number of samples is limited, a basis of cosine functions can be used to fit the data with a minimum of computational difficulty while avoiding artifacts from using arbitrary basis functions.[18] This axial fluence profile can then be integrated to determine the average fluence $\bar{\Phi}$ for the entire fuel channel. Based on infinite-lattice burnup calculations, and using information about the average discharge burnup of the fuel as illustrated in Figure 3 below, this fluence estimate can be used to determine the lifetime plutonium production in this particular fuel channel.[19]

From Channel-Fluence Estimates to Global Plutonium Estimates

In a second step, full-core reactor simulations are used to determine the expected neutron fluence $\bar{\Phi}$ for every channel of the core throughout the life of the reactor (Figure 4). Ideally, these calculations should be based on additional information available on the operating history of the reactor to improve initial production estimates obtained with this method. Once a first neutron fluence

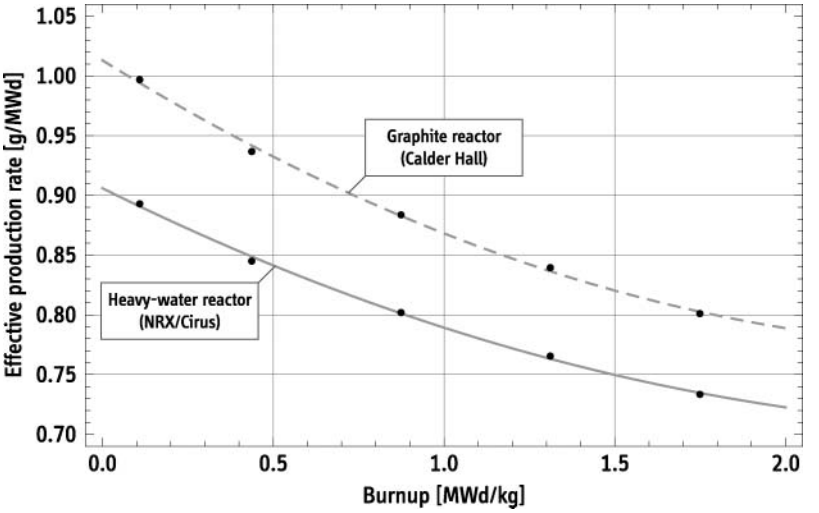

Figure 3: The effective plutonium production rate decreases as the discharge burnup of the fuel decreases. These data can be used to correlate local neutron fluence and plutonium production. Weapon-grade plutonium (93–94% Pu-239) corresponds to a burnup of about 1.3 MWd/kg. MCODE simulations using MCNP5/MCNPX and ORIGEN2.

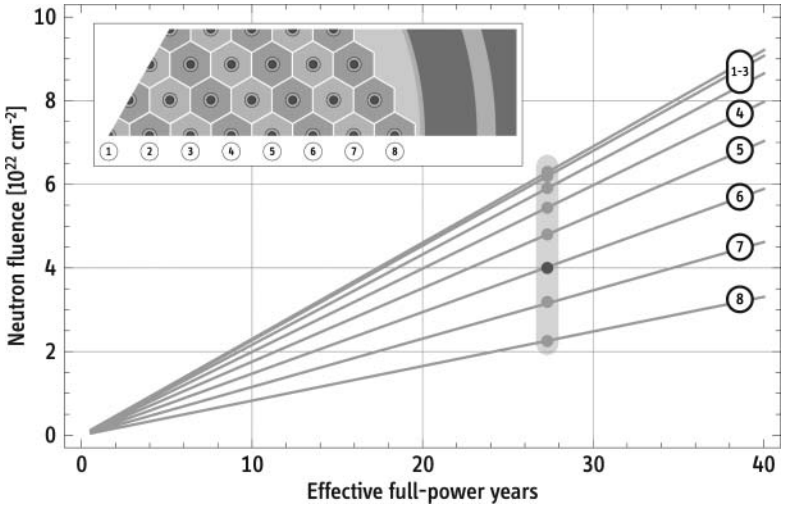

Figure 4: Channel-fluence cross-correlation. The fluence in any one fuel channel is related to the fluence in every other individual channel. These dependencies can be calculated with full-core reactor simulations and later refined when additional measurements become available. As an example, assume that a fluence of $4.0 \times 10^{22}/\text{cm}^2$ is measured in Ring 6; the chart can then be used to estimate the fluence values in all other fuel-channel rings, which range from about 2.2 to $6.4 \times 10^{22}\text{cm}/\text{cm}^3$ in these calculations. MATLAB results using data from MCODE simulations (MCNP5/MCNPX and ORIGEN2).

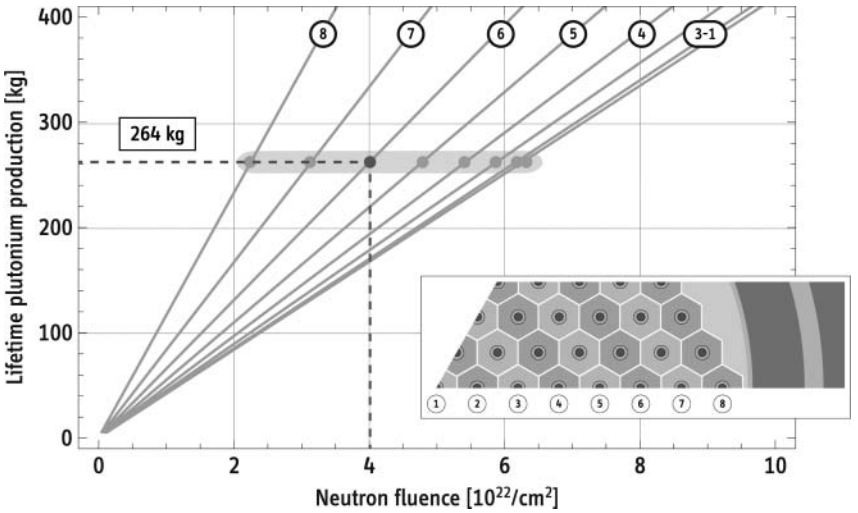

Figure 5: Global plutonium production estimates. The fluence in any individual fuel channel can be used to arrive at a global plutonium estimate. Global estimates from different channels can then be combined to obtain a final estimate with a smaller statistical error. MATLAB results using data from MCODE simulations (MCNP5/MCNPX and ORIGEN2). In this example, the average neutron fluence of $4.0 \times 10^{22}/\text{cm}^2$ calculated for Ring 6 translates into a lifetime production of 264 kg of plutonium in the reactor.

value $\bar{\Phi}$ has been determined based on the analysis of samples from one particular fuel channel, fluence estimates are then also available for every other individual channel in the reactor.

Using these reference data, one can then obtain global plutonium estimates independently from each channel the inspector has access to. This situation is illustrated in Figure 5, which can be used to "look up" the global plutonium estimate corresponding to the local fluence estimate in any channel (or ring of fuel channels, as shown in Figure 1) sampled. Individual estimates can be reconciled to reduce the estimated error of the global estimate significantly. *Instead of generating one global plutonium estimate from one global fluence field, independent global plutonium estimates are now generated from each channel-averaged fluence estimate.* This approach reduces the complexity of the estimation procedure and reduces the number of samples necessary to gather a global plutonium production estimate. The accuracy of this new method would have to be demonstrated in a field test. Previous efforts carried out for graphite-moderated reactors estimated the uncertainty of the standard sampling method to ± 3–7%.[20]

THE WAY FORWARD

Recent new interest in nuclear disarmament is likely to revive research and development on verification technologies for arms control. Nuclear

archaeology, which has been explored off and on since the 1990s, has led to the development of the graphite isotope-ratio method to estimate historic plutonium production at selected sites. The method is best suited for closed down reactors, but can in principle also be applied to operational reactors during temporary shutdowns and fuel reloads to obtain "snapshots" of cumulative production up to that point.

In this article, we have proposed an extension of the method to heavy-water moderated reactors, providing a first overview of potential candidate isotopes that could be used in aluminum structures, which are most relevant for this particular type of reactor. Furthermore, the expected fluence ranges in heavy-water reactors can be significantly higher than those encountered in graphite-moderated reactors, which requires use of isotope ratios different from those used for GIRM; in particular, the classic boron ratio ($^{10}B/^{11}B$) is a much less useful indicator because ^{10}B is rapidly consumed, while selected chlorine, calcium, titanium, and nickel ratios become more valuable. On a conceptual level, we propose a variation of the isotope-ratio method that provides global plutonium production estimates for every fuel channel sampled. This approach could make the method more robust and also offer advantages with regard to implementation in the field.

As a next step, the robustness and applicability of the proposed method ought to be tested in a full-scale exercise. Such an experiment could be used to benchmark the method, quantify errors, and validate optimum sampling strategies. Any exercise will involve on-site sampling combined with computer simulations, which would be carried out in preparation for the exercise itself and again once mass-spectrometric results are reported. Bilateral projects between weapon states could be carried out on a site-by-site basis to establish confidence in the method and the process. Participation of nuclear non-weapon states, which too have many adequate reactors that could be used as testbeds for joint archaeology exercises, would further increase confidence in the method.

NOTES AND REFERENCES

1. The United States and the United Kingdom have made detailed declarations of their fissile material and nuclear weapon inventories. Similarly, France and China have made official or unofficial statements about their nuclear weapon stockpiles. For a detailed discussion of declarations, see Chapter 3, "Declarations of Fissile Material Stocks and Production," in *Global Fissile Material Report 2009,* International Panel on Fissile Materials, (October 2009) 32 (www.ipfmlibrary.org/gfmr09.pdf).

2. S. Fetter, "Nuclear Archaeology: Verifying Declarations of Fissile-Material Production," *Science & Global Security,* 3 (1993), 237–259 (www.ipfmlibrary.org/fet93.pdf).

3. T. W. Wood, B. D. Reid, J. L. Smoot, and J. L. Fuller, "Establishing Confident Accounting for Russian Weapons Plutonium," *Nonproliferation Review,* (Summer 2002) 126–137.

4. Due to the higher power density achievable in their cores, a heavy water reactor is smaller than a graphite-moderated plant of comparable power level and plutonium production rate. Heavy-water reactors are also more easily adapted to produce tritium for second-generation fission weapons.

5. This work is based on: A. Gasner, *Beyond GIRM: Nuclear Archaeology for Heavy Water-Moderated Plutonium Production Reactors,* Senior Thesis, Department of Mechanical and Aerospace Engineering, Princeton University (April 2010).

6. E. A. G. Larson, *A General Description of the NRX Reactor,* CRIO-1043 (AECL-1377), Chalk River, Ontario (July 1961) (www.ipfmlibrary.org/lar61.pdf).

7. Larson, *op. cit.,* 1.

8. Reactor power was first increased to 40 MW, after modifications in 1950 and 1952, and then again to 42 MW in 1961; Larson, *op. cit.*, p. 1. Note that NRX had an accident with a partial core meltdown in December 1952. For additional details on the operating history of the reactor, see also D. G. Hurst, *Canada Enters the Nuclear Age: A Technical History of Atomic Energy of Canada Limited,* (Montreal, McGill-Queen's University Press, 1997).

9. IAEA Research Reactor Database (www.iaea.org/worldatom/rrdb).

10. R. D. Sage, D. D. Stewart, H. B. Prasad, and H. N. Sethna, *Canada-India Reactor,* Papers presented by Canada-India to the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland, September 1–13, 1958, AECL-729, May 1959 (www.ipfmlibrary.org/sag59.pdf).

11. Khushab-I has been operating since 1998. Pakistan is currently building two additional plutonium production 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 NRX/CIRUS design. Recent satellite imagery suggests that Khushab-II is now operational.

12. Z. Xu, *Design Strategies for Optimizing High Burnup Fuel in Pressurized Water Reactors,* Ph.D. thesis, Massachusetts Institute of Technology, January 2003; and Z. Xu, P. Hejzlar, M. J. Driscoll, and M. S. Kazimi, "An Improved MCNP-ORIGEN Depletion Program (MCODE) and its Verification for High Burnup Applications," PHYSOR, Seoul, October 7–10, 2002. MCODE is used with the kind permission of its author.

13. Monte Carlo Team, Los Alamos National Laboratory, *MCNP—A General Monte Carlo N-Particle Transport Code, Version 5,* LA-UR-03-1987, X-5 (April 2003) revised March 2005; A. G. Croff, *A User's Manual for the ORIGEN2 Computer Code,* ORNL/TM7175, Oak Ridge National Laboratory (July 1980); and S. Ludwig, Revision to ORIGEN2, Version 2.2, Transmittal Memo, May 23, 2002.

14. The location of the source material is a major concern for applications of the isotoperatio method. For a heavy-water moderated reactor with a calandria or for a graphite-moderated reactor, source material can be sampled throughout the reactor core. If there is no source material close to the fuel, one may be forced to take samples from the pressure vessel or other secondary structural components. In this case, the error from uncertainties in modeling the reactor would increase.

15. Sample material courtesy of James E. Matos, Argonne National Laboratory; Sample analysis: Activation Laboratories, www.actlabs.com.

16. These are simplified sets of differential equations, typically for 2–4 nuclides. In fact, it is an essential condition for the use of a given element as an indicator that the initial concentrations of adjacent elements that can produce the indicator element be small compared to the initial concentration of the tracking element, i.e., complex production

and decay chains are avoided on purpose. See Chapter 4 ("Forward Modeling of Isotopes in Calandria Aluminum") in Gasner, *op. cit.,* for a detailed discussion.

17. These errors include errors from uncertainties in the cross-section libraries, errors from uncertainties in the initial concentration of the element in multi-isotope systems, errors from production in adjacent elements, and errors from geological variations in the isotopics of the element. For a more detailed discussion, see A. Gasner, *op. cit.*

18. The solution of the diffusion equation yields a cosine dependency in the axial direction for both the cubical and the cylindrical reactor. The presence of control rods may result in more complex axial flux and fluence profiles.

19. Multiple isotope ratios from different elements can be used to improve the estimate for global plutonium production in much the same way the sampling along extra fuel rods improves the results because each pair of isotope ratios provides an independent global plutonium production estimate. Moreover, using channel-averaged fluence estimates to generate plutonium estimates enables the use of different isotopes in different regions of the core and the integration of independent estimates quickly and easily without compromising the accuracy of the method or introducing computational difficulty, as it would if one 3D fluence field were used to generate a plutonium estimate.

20. Wood et al., *op. cit.,* 131–133.