Report

Establishing Confident Accounting for Russian Weapons Plutonium

THOMAS W. WOOD, BRUCE D. REID, JOHN L. SMOOT, AND JAMES L. FULLER

Thomas W. Wood is a Senior Program Manager in the Environmental Technology Division at Pacific Northwest National Laboratory (PNNL) and Manager of Hanford Site Planning and Integration Services. Bruce Reid is a Program Manager at PNNL in nonproliferation technologies and international reactor safety initiatives. John Smoot is a Program Manager at PNNL for activities under the Warhead Safety and Security Exchange Agreement between the United States and Russia. James L. Fuller is the Director of Defense Nuclear Nonproliferation Programs at PNNL.

Ten years after the end of Cold War and the dissolution of the Soviet Union, at a time when U.S.-Russian relations verge on unprecedented levels of cooperation, significant uncertainty remains about Russian weapons-grade plutonium production. During the last decade, cooperative U.S.-Russian programs have made considerable progress in strengthening the site-level security and accounting systems at Russian nuclear facilities; providing secure centralized storage for nuclear materials and warheads; and planning bilateral programs for the ultimate disposition of weapons plutonium by incorporating it into power reactor fuel. The agreements signed at the May 2002 Moscow summit secure a continuing commitment to this agenda. Yet the fundamental question of how much weapons-grade plutonium was produced in Russia remains shrouded. Although the Russian government officially maintains that it has adequate certainty regarding Russian plutonium production, the total production remains a state secret. In informal conversations, however, Russian officials acknowledge a lack of adequate system-wide physical accounting for plutonium stocks. Ultimately, confident management and disposition of this material hinges on the development of some sort of "comprehensive transparency regime" along the lines advocated by Steve Fetter.[1]

An essential element of any meaningful transparency regime would be the knowledge that all plutonium production has been accounted for and remains in the weapons themselves or in safe storage, or has been rendered useless for weapons by burning it in reactors or other means. As Fetter points out, uncertainties about warhead numbers or "the amount of fissile material available to make new warheads would loom much larger" as the number of deployed warheads decreases beyond Strategic Arms Reduction Treaty II (START II) levels. The terms of the May 2002 Treaty of Moscow, which stipulate the reduction of operationally deployed warheads to no more than 1,700 per side by the end of 2012, increase the incentive to ensure that all fissile material is accounted for. Adequately accounting for this material, of course, will require some reasonably precise concept of "all." Despite (or perhaps because of) the controversy

regarding the reversibility of the reductions called for in the Treaty of Moscow, the time is now ripe for increased transparency measures on both sides of the U.S.-Russian strategic relationship. Any means to create transparency with regard to stocks of fissile materials should help cement the progress represented by the treaty.

The September 11, 2001, terrorist attacks and new evidence confirming the nuclear aspirations of terrorist groups underscore the threat of fissile material being stolen from inadequately secured stockpiles. This threat is real, current, and independent of the balance of deployed nuclear weapons in the United States and Russia. While U.S. assistance programs and increased Russian emphasis on site-level Materials Protection, Control, and Accountability (MPC&A) measures have clearly increased overall security of Russian fissile materials, the distribution of these materials across many sites and the apparent lack of a "top-down" accounting system for Russian weapons plutonium is not reassuring. In simple terms, it is what we don't know that is of concern, and "we don't know what we don't know."

The United States recognized the need for this high-level accounting of plutonium production several years ago in *Plutonium: The First 50 Years*.[2] This unilateral declaration of U.S. production states that total U.S. weapons-grade production was 90.5 metric tons (MT).[3] Although no statistical accuracy figure is quoted for this estimate, the report does acknowledge sources of uncertainty and gives a figure of 2.8 MT for "material unaccounted for."[4]

The level of uncertainty about Russian production is much higher. While official U.S. estimates of Russian production remain classified, several unofficial estimates by both U.S. and Russian authors in the open literature in the past few years reveal a significant spread. Anatoli Diakov, in a paper presented at the Fifth International Conference on Radioactive Management and Environmental Remediation, gave an estimate of 125 MT for Russian weapons plutonium production.[5] A few weeks later, at the Global 95 meeting in Versailles, Diakov presented a similar paper that estimated total production at 150 MT. Diakov now stands by the earlier number as the accurate one.

Several similar analyses have also been conducted and published. In the absence of an exhaustive reactor-by-reactor comparison of estimates in the literature, it should be noted that any such estimate of production is constructed from a set of assumptions about reactor operation dates, power levels, and fuel cycle parameters. In this context, it is also important to realize that all Russian and U.S. plutonium production reactors were upgraded in several stages, so final power levels were typically several times the initial design levels. The uncertainty associated with this history of upgrades, plus the uncertainly in plutonium yields associated with different fuel designs and fuel cycles, means that any honest estimate based on this approach will have a margin of error of tens of MT.

To put this level of uncertainty into context, 25 MT represents more weapons-grade plutonium than is held in the British, French, and Israeli inventories combined. In terms of warheads, it is roughly equal to the START II inventory allowed to the United States and Russia—3,500 warheads. It is also more than double the agreed-upon target of 1,700 warheads per side set in the 2002 Treaty of Moscow. It is several hundred to a few thousand times as large as the potential plutonium inventory held by North Korea, for which the United States and its allies have offered to exchange light water reactors at a cost of several billion dollars.

This level of uncertainty, while no longer relevant in terms of current Russian strategic capability, poses some problems for the future. First, "system-level" accounting for all fissile material is fundamental to a continuing, sustainable MPC&A program in Russia. The issue of overall plutonium inventories is a basic boundary condition in any such program and, left unresolved, limits the ultimate value of this program.

In addition to declaring its production, the United States recognized soon after the Cold War that verifying fissile materials production was both important and challenging. In 1991, the Federation of American Scientists (FAS) published *Ending the Production of Fissile Materials for Weapons—Verifying the Dismantlement of Nuclear Warheads*,[7] which examined the technical feasibility of verification measures for both fissile materials production and warhead dismantlement. This report first called for "cooperative research (Nuclear Archeology) on the physical evidence that could be used to check and refine these production records."

Shortly after this report was published, the U.S. Department of Energy (DOE) initiated a program at Pacific Northwest National Laboratory (PNNL) to evaluate and develop the technical basis for nuclear archeological meth-

ods. This program, funded by the DOE and the Arms Control and Disarmament Agency (ACDA), was active from 1992 to 1998. PNNL collaborated with several other laboratories on the program, including Oak Ridge National Laboratory, Los Alamos National Laboratory, Massachusetts Institute of Technology, University of Missouri, and Berkeley Laboratories in the United Kingdom.[8] The program developed several concepts for verification measures but focused most intensely on developing a method to estimate plutonium production in graphite reactors—the Graphite Isotopic Ratio Method (GIRM).

NUCLEAR ARCHEOLOGY PROGRAM AND DEVELOPMENT OF GIRM

The Nuclear Archeology Program was quite broad in concept and included validation or verification measures for all fissile materials, including highly enriched uranium (HEU), and methods, including environmental sampling, epidemiology, and economic estimation, in addition to evaluation of product and plant materials—in short, the entire spectrum of physical evidence suggested by the FAS report. By the second year of this program, however, the majority of its resources were devoted to the detailed evaluation and development of GIRM, based on the promise it showed in theoretical studies and its clear relevance to the Russian plutonium production question. In fact, all Russian weapons plutonium production occurred in graphite-moderated reactors (see Table 1).

The basic principle of GIRM is that trace impurities in nuclear graphite, via neutron capture reactions that alter their natural isotopic ratios, can serve as accurate indicators of cumulative neutron flux (fluence) and thus lifetime plutonium production. This principle depends on the existence of useful impurities in the graphite, the physical stability (lack of mobility) of the impurities within the graphite matrix, the permanence of the moderator graphite for the life of the reactor, the ability to measure impurities in isotopics accurately, and knowledge of reactor physics and fuel cycle parameters that correlate fluence with plutonium production in the fuel. A summary description of the method has been published by PNNL.[9]

Graphite is manufactured from petroleum coke or other organic materials that contain a variety of elemental impurities. To support a sustained fission reaction using natural uranium fuel, the combined neutron absorption cross-section of graphite impurities must be below about 2 parts per million (ppm) equivalent boron concentration. Thus, nuclear graphites are purified extensively using a variety of chemical methods but still retain measurable quantities of many impurities.

Table 2 presents data on selected elemental impurities in several grades of Hanford reactor graphite. Similar analyses of British, French, and Russian reactor graphite, while differing in the details of impurity concentrations, show measurable quantities of potentially useful indicator elements. Early feasibility work focused on laboratory measurements of isotopic ratios for impurity elements in irradiated nuclear-grade graphite using archival samples from the Hanford C-Reactor and the French G-2 reactor. The work addressed impurity concentrations, sample preparation (machining, ashing, digestion, and ion exchange chemistry), and mass spectrometric analysis, which were important early technical hurdles to the feasibility of GIRM.

It was hoped initially that isotopic ratio measurements could be made directly from unprocessed graphite samples using glow discharge mass spectrometry (GDMS). This approach proved infeasible.[10] PNNL developed a sample preparation concept in which graphite (or its ash) was digested and used to prepare elemental separates for spectrometric analysis. These procedures were adapted from standard analytical methods and combined to yield a series of elemental separates from each sample. To avoid a complex separations scheme, initial work focused on calcium, titanium, strontium, and barium. Each of these elements was a reasonably good indicator in the relevant fluence range and was easily separable. These elements were present in the Hanford and French graphites in typical concentrations of 100 parts per billion (ppb) to a few ppm, with the exception of calcium, which was often a few hundred ppm.

Thermal ionization mass spectrometry (TIMS) allowed for accurate measurements of isotopic ratios from (elemental separate) samples of mass as low as 100 nanogram (ng; 10E-7 gr). Analytical precision (both within run and replicability) was on the order of a few tenths of a percent to one percent. This level of error is insignificant in the context of the other uncertainties in the problem.

Based on these results, the methods developed for sample preparation and analysis met the desired performance objectives, and the initial mass spectrometric analyses were judged successful. The overall conclusion

Table 1. U.S. and Russian Plutonium Production Reactors

Country/Site	Reactor	Moderator	Startup	Shutdown
U.S./Hanford	B	graphite	1944	1968
	D	graphite	1944	1967
	F	graphite	1945	1965
	H	graphite	1949	1965
	DR	graphite	1950	1964
	C	graphite	1952	1969
	KE	graphite	1955	1971
	KW	graphite	1954	1970
	N	graphite	1964	1987
U.S./Savannah River	R	heavy water	1953	1964
	P	heavy water	1954	1988
	L	heavy water	1954	1988
	K	heavy water	1954	1988[a]
	C	heavy water	1955	1985
Russia/Ozersk	A	graphite	1948	1987
	IR-AI	graphite	1951	1987
	AV-1	graphite	1950	1989
	AV-2	graphite	1951	1990
	AV-3	graphite	1952	1990
Russia/Seversk	I-1	graphite	1955	1990
	I-2	graphite	1958	1990
	ADE-3	graphite	1961	1990
	ADE-4	graphite	1964	operational[b]
	ADE-5	graphite	1965	operational[b]
Russia/Zheleznogorsk	AD	graphite	1958	1992
	ADE-1	graphite	1961	1992
	ADE-2	graphite	1963	operational[b]

(a) K reactor at Savannah River site was refurbished between 1988 and 1992, tested in 1992, and placed in cold standby until 1996, when it was shut down permanently.
(b) These reactors remain operational as of July 2002. A U.S.-Russian project to replace the heat and power generated by these reactors with conventionally-fuelled power plants is under way, with completion (and reactor shutdown) scheduled in Seversk for the end of 2005 and in Zheleznogorsk for the end of 2006.

of this work in 1993 and 1994 was that the GIRM process demonstrated significant promise for determining the plutonium production of graphite-moderated production reactors.[11]

To address the other uncertainty factors confronting a practical application of GIRM, a full-scale reactor demonstration of the process was performed from 1995 through 1997 using the Trawsfynydd Unit II reactor in Wales.[12] This is a commercial, gas-cooled, graphite-moderated reactor that was starting decommissioning activities when the demonstration began. Ninety samples were acquired from that reactor. Nuclear Electric, the plant operator, provided the operating history (fuel management), operating conditions, and the reactor and fuel design and materials to assist in determining the cumulative plutonium production. The actual plutonium production records for Trawsfynydd Unit II reactor were not provided at the beginning of the demonstration to ensure that the test was a blind one.

Based largely on TIMS results for titanium isotopes from these samples, the cumulative plutonium production for the Trawsfynydd Unit II reactor over its operating life was estimated as 3.63 MT. This estimate was calculated to have an accuracy (relative standard error) of 5.2 percent based on the information known about the reactor during the study and error propagation calculations using the reactor physics model. The Nuclear Electric plutonium production records indicated that this estimate was in fact

Table 2. Concentrations (ppm) of Selected Impurities in Hanford Reactor Graphite

Element	Detection Limit[a] (ppm)	Grade of Graphite[b]					
		KSO	KCF	CSO	CSF	GBF	SGBF
Al	0.007	0.59	0.06	0.36	0.09	0.15	0.83
B	0.005	3.0	0.08	2.8	0.13	0.04	0.10
Ba	0.0005	2.6	0.02	2.6	0.03	0.04	0.007
Ca	0.002	210.0	0.13	135.0	0.27	0.59	0.22
Cr	0.003	1.1	BDL	0.34	BDL	0.005	BDL
Cu	0.001	0.68	0.15	0.19	0.28	0.06	0.68
Fe	0.001	5.6	0.33	2.8	0.28	3.1	0.19
Li	0.001	0.37	0.002	0.21	0.003	0.003	0.003
Ni	0.005	0.31	0.02	2.5	0.06	0.20	0.05
S	0.02	31.0	0.04	33.0	0.07	0.07	0.05
Si	0.005	1.3	0.67	6.0	1.3	0.07	1.25
Sr	0.0005	4.0	0.003	3.1	0.002	0.008	BDL
Ti	0.001	7.5	0.001	8.2	0.01	0.02	0.001
V	0.001	11.0	BDL	12.0	0.004	0.12	0.015
Zn	0.001	5.4	0.06	160.0	0.16	0.08	0.43

[a]) These detection limits are circa 1980 and are conservative.

[b] These grades refer to coke sources, plant locations, and purification methods.

accurate to within 0.3 percent of declared production.[13] Clearly, the Trawsfynydd reactor work demonstrated that GIRM is a viable means of estimating plutonium production in a graphite-moderated reactor, provided sufficient samples are obtained in the active core region.[14]

In addition to the U.S. work in the PNNL Nuclear Archeology Project, some Russian work has been relevant to GIRM feasibility in the last several years.[15] Between 1996 and April 2000, the Moscow State Engineering Physics Institute (MEPhI) conducted work under an International Science and Technology Center (ISTC) grant (ISTC Project 561) to investigate the radioactive contamination of the graphite moderator in Russian plutonium production reactors. PNNL was one of several international collaborators on this project. The results of this work will be used to plan safe and cost-effective disassembly of the reactors and subsequent storage of the radioactive material. Lack of knowledge of the characteristics of the graphite, the influence of irradiation, and the penetration of contaminants into the graphite (resulting from operating events such as seal failure of the fuel elements or cooling water leaks) created the need for an in situ graphite sampling program. During the initial work by MEPhI, three reactors were investigated at Seversk. The results of this work are valuable for possible GIRM application. They demonstrated the technical adequacy of sampling methods and gave some assurance that the Russian graphite contains useful indicator elements as well as an archive of irradiated graphite for additional characterization.

ACCURACY OF GIRM

The utility of GIRM or any nuclear archeological method requires a reasonable degree of accuracy. In this context, it is important to remember that we are seeking a measure of the weapons-useful plutonium that was in fact produced and separated. From the Hanford experience, the uncertainty associated with reprocessing losses is on the order of 1 to 2 percent of total (in-reactor) production; that is, reprocessing yield might vary roughly from 95 to 99 percent as a function of the process used and the specifics of the reprocessing plant operation for a given dissolver batch. Because this uncertainty is extremely difficult to resolve, it constitutes an information barrier and sets a practical limit on the certainty for which an estimation technique like GIRM should reasonably strive. This is illustrated in Table 3, which gives resulting plutonium inventory accuracies as a function of the combined accuracy of GIRM and the reprocessing loss fraction estimate.

In 1994 through 1997, work was performed to quantify the accuracy that might be expected with GIRM in estimating the cumulative plutonium production in a reactor. The uncertainties addressed included: (1) measurements of isotopic ratios; (2) translating uncertainties in reactor parameters to uncertainties in local plutonium production; and (3) integrating the local plutonium production estimates into an estimate of the cumulative plutonium production for the entire reactor. The analytical accuracies assumed in this work were those from the previously described TIMS studies of Hanford and French graphites. The reactor physics models used were based on the British WIMS computer code and were applicable to water-cooled, graphite-moderated reactors (including the Russian designs).

This work estimated that, for a single generic production reactor with about 30 appropriately distributed graphite samples, the GIRM process could determine the cumulative plutonium production within a seven percent relative standard error. This level of accuracy was projected under conditions in which very little information existed about the reactor operating conditions or fuel management scheme (e.g., reactor moderator temperature is

Table 3. GIRM and Net Plutonium Inventory Accuracies[(a)]

GIRM (percent)	**Reprocessing** (percent)	**Net Inventory** (percent)
10	2	10.40
5	2	5.38
2	2	2.83
1	2	2.24
0	2	2
10	1	10.05
5	1	5.1
2	1	2.24
1	1	1.41
0	1	1

(a) Accuracy of estimates expressed as percent standard error.

assumed to be known only within a 400 degree Centigrade range), and only one indicator element is used in the analysis. These conditions tend to degrade GIRM's accuracy.[16]

However, a recent recalculation of the uncertainties for a single reactor, for which less conservative assumptions were made about knowledge of the operating parameters, shows that substantially better accuracy is reasonably possible. This calculation, presented in Table 4, gives an estimated relative standard error of 2.55 percent. Overall, the calculation is believed to be realistic in terms of the ability to estimate reactor operational conditions at the Russian production sites.[17]

This is a rough calculation using a simple error model. Against the calculated result of an error of between two and three percent for each reactor, one should consider potential complicating factors that are present in Russian production reactors. These include the potential for contamination of the moderator graphite from "wet" (loss of coolant) accidents or, in a few cases, fuel-melt accidents. Another complication is the fact that Russian reactors have in many cases had the graphite lining of fuel tubes replaced. These complications are serious but are believed to be surmountable in an application of GIRM. These conditions are described in more detail in the next section, which considers the potential for application of GIRM in Russia.

Perhaps offsetting these special factors in the Russian production reactors are the prospects for using multiple indicator elements and the fact that the inventory we are concerned about is the sum of several reactor production totals. A program that applied GIRM to multiple reactors would benefit from the aggregation of these estimates. To the extent that these estimates were statistically independent, their sum would be considerably more accurate than the average individual estimate. For example, a set of 13 independent GIRM estimates (one for each Russian production reactor), each accurate to five percent relative standard error, would yield a total production estimate accurate to about 1.4 percent standard error. This is as accurate as necessary, given the "information barrier" of the reprocessing losses.

Of course, these individual reactor estimates would not be completely independent because the reactor designs are common or similar among reactors and sites, and the

Table 4. Recalculation of Uncertainties[18]

	dPu/dP[(a)]	**Parameter Span Assumed**		**Resulting Relative Standard Deviation (percent)**	
Parameter	**(percent)**	**(in ref.)**	**(realistic case)**	**(in ref.)**	**(realistic case)**
Moderator Temperature	2.6/100°C	400°C	40°C	3.0	0.3
Fuel Temperature	0.49/100°C	200°C	40°C	0.3	0.06
Coolant Temperature	2.06/100°C	250°C	50°C	1.48	0.29
Fuel Enrichment	0.85/0.1 percent	0.189 percent	0.05 percent	1.6	0.42
Goal Exposure	2.2/100 MWD/T	350 MWD/T	100 MWD/T	2.2	0.62
Cross-Sections	1.6/1 percents	NA	NA	4.8	2.4
Resulting Total Uncertainty in Plutonium Estimate				6.46	2.55

Key: MWD = megawatt days

(a) Maximum partial of production to parameter over variant range.

isotopic measurements will be conducted using similar methods. However, the actual isotopic ratios in the graphite are in fact independent, and these drive the estimates to a considerable degree.

Current work at PNNL includes developing and applying sophisticated multivariate simulation models for a richer assessment of GIRM accuracy and errors. Initial work with these models confirms that the simple error model for which results are given here is conservative. While these models will be useful in setting application requirements driven by error "budgets," the bottom line on GIRM accuracy is that it cannot be known precisely until the actual sampling and analysis campaigns are conducted. More valuable than demonstrating that the prospective accuracy per reactor will be three percent rather than six percent is the observation that even relatively crude results aggregated over several reactors would be a dramatic improvement over the current level of uncertainty.

The method of determining plutonium production from samples in graphite-moderated reactors has been proven. The calculated accuracy of this method is good enough to increase our certainty about Russia production substantially, and the method performed far better than this theoretical accuracy in its one full-scale test.

POTENTIAL FOR APPLICATION OF GIRM IN RUSSIA

Factors that bear on the applicability of GIRM to the Russian production reactors include design features, operational histories, current conditions, and ability to sample and analyze samples. Our evaluation is based on published sources as well as information gained in collaborations on ISTC and DOE lab-to-lab projects in Russia.

All of the Russian weapons plutonium production reactors were graphite-moderated, water-cooled reactors. In principle, this fact allows the GIRM technique to supply a global weapons-grade plutonium production estimate for Russia. All but three reactors have been shut down and are in some stage of deactivation or decommissioning. All of the shutdown reactors have been defueled, and much of the cooling systems, control rods and drives, and instrument and other support systems have been removed. The graphite cores are intact, with ready access to fuel channels from which samples could be taken. In one case, a concrete layer has been poured on the upper surface of the moderator block to limit contamination during extended safe storage. This concrete poses an obstacle to accessing the fuel tubes, but it could be circumvented.

Three Russian production reactors remain in operation at Seversk and Zheleznogorsk. In 1993, Presidents Clinton and Yeltsin agreed in principle that these reactors would be shut down, and in 1996 the Gore-Chernomyrdyn Commission endorsed a proposal to convert the cores and fuel design of these reactors to an alternative design that would no longer produce weapons plutonium. Since then, safety issues with the new design have resulted in a decision to abandon this concept, and the current preferred outcome is replacement of these reactors with fossil fuel power plants. In the meantime, the reactors continue to operate, providing steam heat and electrical power for the surrounding communities (and creating about 1.2 MT per year of weapons plutonium). The current program to replace these reactors would result in the shutdown of the reactors in Seversk by the end of 2005 and in Zheleznogorsk by the end of 2006. Although operational, these reactors could be sampled for GIRM analysis. The samples are small enough in volume that reactor safety is not degraded. Both the Hanford production reactors and the U.K. Magnox reactors were sampled several hundred times during their operational lives without any adverse effect on reactor operability or safety.

Russia has the capability to sample all of their production reactors. Sampling was a routine procedure at all three production sites and the opportunity remains for further sampling. During the ISTC study conducted at Seversk by MEPhI, hundreds of samples were obtained from the three reactors over several months. The specifications for these samples were reviewed by PNNL, and the samples are adequate for use with the GIRM technique.

Russian production reactors used replaceable graphite fuel channel liners (sleeves) to maintain the dimensional stability of the fuel tubes during prolonged irradiation of the cores. This poses an issue for GIRM application because graphite from these liners would have been exposed to only a fraction of the life-cycle neutron fluence in these reactors. Care would need to be taken in sampling to avoid contamination between these two sources of graphite. Because the liners are typically about one centimeter (cm) thick and GIRM samples taken from the moderator block are several cm in length, only a remote possibility exists for outright substitution of sleeve samples for block samples.

One factor that needs to be assessed in more detail is the availability of engineering data on reactor operation. As illustrated by the previous accuracy calculation, the specifics of fuel enrichment and burnup, graphite operating temperature, and other factors bear substantially on interpreting isotopic measurement in terms of plutonium production. Records at some Russian sites have been destroyed or lost, but we understand that those remaining are being preserved.

Measurements of radioactive isotopes often lose their value for samples that have decayed for several years since irradiation. Some fuel isotopic measurements are particularly sensitive in this respect. An advantage of GIRM is that it uses stable or very long-lived isotopes, for which decay is negligible. Thus, the isotopic "signal" that ultimately determines the production estimate is permanently encoded in the impurities within the reactor core. It is important, however, to know the core location of GIRM samples. The statistical aggregation of sample information over the reactor core depends on this information to fit typical flux shapes to the data. These fluence (and associated plutonium production) fields are then integrated to obtain a reactor production estimate. Thus, a complete disassembly of the reactor core, while preserving the isotopic information, would pose a much more difficult nuclear archeological problem. The good news is that the decommissioning plans call for these cores to be kept intact in definite "safestore" conditions.

Several of the Russian reactors have suffered repeated "wet" accidents in which cooling water or steam was released in the graphite core. In some cases, fuel has melted into the moderator structures. These accidents are a concern for GIRM application from at least two perspectives. First, cooling water could introduce additional impurities into the graphite. This could, in effect, "reset the meter" of isotopic ratios because these impurities would have natural isotopic ratios at the time of their introduction. This problem might be a particular concern relative to chlorine. In addition, fuel-release accidents in a region of the core would rule out using the actinides as indicators and could significantly alter the neutronics of nearby regions.

Because the graphite impurities of interest are typically present (in U.S., French, and U.K. graphites) in the form of small oxide clusters (several microns to tens of microns), there is some hope that introduced impurities could be preferentially separated prior to analysis. This would allow GIRM samples to be taken from the regions affected by accidents. Another approach would be to avoid these zones in taking samples.

The final but critical factor in the applicability of GIRM is the impurity composition of the moderator graphite. We have some published information based on MEPhI studies of the Seversk reactors for decommissioning purposes. These studies[19] included evaluations of elemental concentrations of chlorine, iron, cobalt, zinc, arsenic, silver, cesium, tungsten, gold, mercury, thorium, and uranium. These elements were selected for their ability to form radioactive isotopes that are a concern in decommissioning, while the potential GIRM indicator elements form stable isotopes. Even so, there is some overlap. Chlorine, cobalt, thorium, and uranium are useful indicators, and all are present in easily measurable concentrations. Counting studies were also done for specific nuclides, including hydrogen-3, carbon-14, chlorine-36, nickel-63, strontium-90, barium-133, and europium-152, -154, and -155. Results indicate that chlorine, nickel, strontium, barium, and europium isotopes were commonly present.

Although this was not included in the published results, informal discussions with the Russians suggest that other possible indicators, including titanium and calcium, were commonly present in production reactor graphite. This should be confirmed with analytical results as one of the first steps in a cooperative program to further assess GIRM for application to Russian reactors.

ALTERNATIVE METHODS FOR ESTIMATING PLUTONIUM PRODUCTION

GIRM is not the only method that can increase certainty about past Russian plutonium production. Other records- or materials-based methods could be valuable. A comprehensive review of records could at least confirm the extent and value of production history. Current indications are that the only comprehensive plutonium accounting scheme in Russia are the financial records documenting transfers of final plutonium product from the Ministry of Atomic Energy (Minatom) to the Ministry of Defense.

Among materials-based methods, assays of the front or back ends of the defense fuel cycle (fuel fabrication or waste processing) could conceivably be used to estimate production. Evaluation of Hanford fuel fabrication and waste processing byproducts in the early stages of the nuclear archeology program showed only weak cor-

relation to cumulative production. A similar outcome could be expected in Russia.

Methods based on environmental sampling have also been evaluated by both the United States and Russia and were discussed briefly at an all-institute meeting held in May 2001 (see next section). Some of these offer promise as cumulative indicators but have not been systematically evaluated. In summary, although GIRM is not the only possible method on which an independent estimate of Russian weapons-grade plutonium production could be based, it is the most direct (in terms of theoretical correlation with production) and the most rigorously tested.

What we now know about the Russian production reactors suggests that GIRM should be technically applicable and has the prospect of substantially increasing confidence that Russian weapons plutonium is adequately accounted for. Russia must take the next step—a more detailed assessment of the applicability of GIRM. The process could enhance the confidence of Russian officials and scientists in GIRM as a reliable tool to achieve better accounting, and would also establish their technical ability to conduct the necessary analyses.

COOPERATIVE EVALUATION OF GIRM

The first steps in a cooperative evaluation of GIRM were taken in FY 2001 in a DOE-Minatom "lab-to-lab" project conducted by PNNL and the Russian Federal Nuclear Center-All Russian Scientific Research Institute for Experimental Physics (RCNF-VNIIEF). This project focused on exchanging basic methodological documents for GIRM, followed by a technical working group meeting organized by VNIIEF for several Russian institutes in Moscow in May 2001.

Seven Russian institutes attended this meeting. Collectively they represent expertise in graphite reactor design and operation, plutonium production history, sampling and analytical chemistry, and reactor physics and statistics. There were 47 attendees representing the Kurchatov Institute, the Afrikantov Experimental Machine Building Design Bureau (OKBM), the Institute of Physics and Power Engineering (IPPE), the All-Russian Scientific Research Institute of Technical Physics (VNIITF), the Mining and Chemical Combine (Zheleznogorsk), MEPhI, and Minatom in addition to the PNNL and VNIIEF personnel.

The meeting included presentations on the basic nuclear archeology concept and DOE project; an extensive description of GIRM and the proof-of-principle experiments conducted to date; GIRM accuracy and uncertainties; the impurity composition of the Russian graphite; and a preliminary evaluation of the applicability of GIRM in Russia. The conclusion was that GIRM shows substantial technical merit as a tool to validate plutonium production in graphite reactors. The Russians commented that using stable isotope ratios via mass spectrometric analysis was "an elegant solution" to the general problem of fluence estimation in reactor materials.

While the evaluation of the technical working group was generally positive, the group recognized that considerable work would be required to fully assess an application of GIRM in the Russian production reactors. There was considerable enthusiasm for such a program, including a proposal by IPPE to conduct an evaluation in the AM reactor at Obninsk. This and other possible paths are described in the next section.

PATH FORWARD

With the basic method of GIRM developed and a fair amount of experimental data in hand, there is now an opportunity to support Russian experts in their own evaluation of this tool. To realistically visualize how this process might proceed, we must consider the politics of plutonium accountability in Russia. Because the official Russian position on plutonium production is that reliable accounting now exists but totals remain state secrets, it would be premature to advocate the immediate application of GIRM at Russian plutonium production reactors. Certainly, Russian officials want and deserve a high degree of confidence that the method would not give misleading results, and they want to be in control of any application at their reactors. Thus, the next step for possible application of GIRM in Russia involves supporting confirmatory analysis by Russian institutes in a context that does not threaten the security of their plutonium production records.

In principle, this work could be conducted on a reciprocal basis if desired, using U.S. graphite samples and a program of parallel planning for application of GIRM at the Hanford reactors to mirror the assessment of applicability in Russia. Whether this is desirable is not yet clear. The assessment would include several main tasks, briefly described below.

Graphite Elemental Composition Analysis

Available data suggest that the requisite elemental impurities are present in the Russian graphite, but this needs to

be confirmed in systematic comparative studies for a wide range of graphite grades and vintages. This work could use unirradiated graphite, revealing no information about reactor operation, and is well within the technical capability of several Russian institutes. The work would confirm the existence of useful indicators and suggest an optimal indicator suite and separations scheme.

Sample Preparation and Isotopic Composition Studies

Once indicators are identified, Russian institutes could begin testing and applying elemental separations and mass spectrometric methods to demonstrate or build capability for the basic analytical methods that allow for GIRM estimation. This work might require some transfer of technology from the United States to Russia, possibly including instruments for TIMS or other mass spectrometric analysis, but would ultimately give confidence that the measurements were sufficiently accurate. It could be initiated using unirradiated graphite and later graphite irradiated in research reactors. The final step in this task would be to demonstrate that the isotopic indicators can be confidently correlated with neutron fluence estimated by other means.

Full Scale Proof-of-Principle

The ultimate proof of GIRM must be a test in which the method is used to predict plutonium production in an actual Russian reactor. Only this type of demonstration allows for all possible sources of error to be realized and to interact; thus, it gives the only compelling proof of accuracy in practice. This phase of work could (technically) be conducted at any of several reactors. The AM reactor at Obninsk is worthy of consideration; it has the advantage that its fuel assemblies (not just the moderator block) contain graphite, allowing for direct correlation of isotopics in graphite and fuel. An alternative test site would be the F reactor at the Kurchatov Institute; this reactor has several advantages: (1) it has a very low and well-known power density and well-characterized flux profile; (2) it is the first operational graphite reactor in Russia and thus is likely to present a robust impurity composition characteristic of early nuclear graphite manufacture; and (3) it contains the original natural uranium fuel, allowing for the possibility of a lifecycle correlation between graphite impurity and fuel isotopics. As far as we know, this feature is unique among graphite reactors worldwide and would constitute the definitive test of GIRM accuracy.

CONCLUSIONS

The need to provide a comprehensive accounting method for Russian weapons plutonium is real and pressing. The United States has an opportunity to provide Russia with an important technical tool in support of this objective. Even without an agreement to apply this tool in a bilateral context, GIRM would allow Russian experts to confirm any questionable reactor-level accounting within their system on a confidential basis and help prioritize MPC&A and other measures. These goals can be accomplished in a cooperative program for which a technical consensus already exists, with a degree of U.S. technical support that is comfortable for Russia, and in a very cost-effective manner, given the risks involved.

Such a program would support MPC&A, reactor shutdown agreements, and other elements of the current U.S.-Russian cooperative agenda. It would also remove an important barrier to establishing system-wide accounting for weapons plutonium in Russia. This program, if successful, would also increase the value of and incentives for a fissile material cutoff treaty (FMCT). While the FMCT is important and valuable independent of certainty about past production, the two objectives are complementary.

Use of GIRM in Russia would also provide an important precedent for other applications. Several other countries have used graphite-moderated reactors, and the technology will always be attractive based on the ability to produce plutonium from natural (unenriched) uranium. This precedent would be valuable from both a policy perspective and in terms of establishing an accepted international technical basis for GIRM.

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[1] Steve Fetter, "A Comprehensive Transparency Regime for Warheads and Fissile Materials," *Arms Control Today* 29, (January 1999).

[2] U.S. Department of Energy, *Plutonium: The First 50 Years*, DOE/DP-0317 (Washington, DC: U.S. Department of Energy, 1996).

[3] This figure excludes 1.7 MT of civil plutonium, 12.9 MT of fuel grade plutonium, 0.6 MT created in research reactors, and 5.7 MT acquired from other countries for an inventory 111.4 MT.

[4] This is consistent with estimates by cleanup contractors at Hanford, that slightly less than 2 MT of plutonium remain in the high-level and transuranic wastes or in the soil under the fuel processing and waste treatment facilities. Thus, while this material is unaccounted for in the MPC&A sense, we estimate that much of it remains in waste at Hanford.

[5] Anatoli Daikov, "Utilization of Already Separated Plutonium in Russia and International Security Problems: Consideration of Short and Long-Term Options," *Proceedings of Global 95*, Versailles, France, 1995.

[6] Production of weapons-grade plutonium continues in three reactors at two

sites in Russia because the steam heat and electrical power from these reactors is vital to the nearby communities. This adds about 1.2 MT per year to the total production.

[7] Federation of American Scientists, *Ending the Production of Fissile Materials for Weapons—Verifying the Dismantlement of Nuclear Warheads* (Washington, DC: Federation of American Scientists, 1991).

[8] Follow-on work under DOE NN-40 and its successor organizations has continued methods development and involved several Russian Institutes, as described in the section on Cooperative Evaluation of GIRM.

[9] James P. McNeece, B. D. Reid, and T. W. Wood, *The Graphite Isotope Ratio Method (GIRM): A Plutonium Production Verification Tool*, PNNL-12095 (Richland, WA: Pacific Northwest National Laboratory, 1999).

[10] More recent unpublished results suggest that direct measurement of some isotopes is possible using secondary ion mass spectrometry (SIMS).

[11] Thomas W. Wood, D. C. Gerlach, B. D. Reid, and W. C. Morgan, *Feasibility of Isotope Measurements: Graphite Isotopic Ratio Method*, PNNL 13488 (Richland, WA: Pacific Northwest National Laboratory, 2001).

[12] Studies showed that sampling a shutdown Hanford reactor would be feasible but costly. The safety and radiation protection requirements, and the need to remove the metal fuel channel liners from these reactors, would have cost nearly $1,000,000 in 1995, while the Trawsfynydd samples were obtained at a cost of £90,500, ($146,000).

[13] Achieving this level of accuracy, however, required special statistical techniques that accounted for bias in isotopic measurements due to a very low abundance of the indicator element (titanium).

[14] Bruce D. Reid, D. C. Gerlach, P. G. Heasler, and J. V. Livingston, *Trawsfynydd Plutonium Estimate* (Richland, WA: Pacific Northwest National Laboratory, September 1997).

[15] This is by no means an exhaustive account; no general Russian language review has been done.

[16] Robert J. Talbert, B. D. Reid, P. G. Heasler, D. C. Gerlach, C. M. Heeb, and K. A. Pauly, *GIRM Accuracy Calculations*, PNL-RTC-0693 Rev. 1 (Richland, WA: Pacific Northwest National Laboratory, 1995).

[17] Other factors also introduce conservatism into this calculation. All the partial derivatives used are taken at their maximum values in the original operating parameter ranges.

[18] Pacific Northwest National Laboratory, "Accuracy of Plutonium Production Estimates from Isotopic Ratios in Graphite Reactors," PNL-RTC-0693 Rev. 1, DEL, declassified May 1998, Table 3.11.

[19] Anatoli Bushuev, V. N. Zubarev, and I. M. Proshin, "Composition and Content of Impurities in Graphite of Russian Reactors," *Proceedings of Technical Working Group Meeting on the Graphite Isotope Ratio Method*, RFNC-VNIIEF 205976-A-K4 (Sarov, Russia:RFNC-VNIIEF, 2001).