Immobilization of Excess Weapons Plutonium in Russia

L. J. Jardine
Lawrence Livermore National Laboratory

G. B. Borisov and O. A. Mansourov
State Research Center of Russian Federation
A. A. Bochvar All Russia Research Institute of Inorganic Materials (VNIINM), Moscow, Russia

This paper was prepared for submittal to the Waste Management 1999 Conference (WM'99)
February 28–March 4, 1999
Tucson, Arizona

January 25, 1999

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.
DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.
IMMOBILIZATION OF EXCESS WEAPONS PLUTONIUM IN RUSSIA

L. J. JARDINE
University of California
Lawrence Livermore National Laboratory
P. O. Box 808, L-195
Livermore, CA 94551

G. B. BORISOV AND O. A. MANSOUROV
State Research Center of Russian Federation
A. A. Bochvar All Russia Research Institute of
Inorganic Materials (VNIINM)
Moscow, Russia

ABSTRACT

In this paper, we examine the logic and framework for the development of a capability to
immobilize excess Russian weapons plutonium by the year 2004. The initial activities underway
in Russia, summarized here, include engineering feasibility studies of the immobilization of
plutonium-containing materials at the Krasnoyarsk and Mayak industrial sites. In addition,
research and development (R&D) studies are underway at Russian institutes to develop glass and
ceramic forms suitable for the immobilization of plutonium-containing materials, residues, and
wastes and for their geologic disposal.

INTRODUCTION

During the Cold War, the United States and Russia conducted various activities and operations
associated with the production of plutonium for nuclear weapons. Intermediate products and
wastes containing plutonium were generated as a result of these operations. However, the United
States and Russia followed very different approaches regarding the recovery and purification of
plutonium in various intermediate products and wastes during the Cold War period. Approaches
were based, in part, on the estimated values of plutonium, which were not always the same for
the U.S. and Russia. To understand possible disposition pathways or options in Russia for
immobilization of excess weapons plutonium, it is useful to understand the foundations of the
different approaches and the Russian perspectives on the value of the plutonium contained in
residues and waste streams.

This paper provides a short background that highlights these differences in practices and
perspectives, followed by a summary of the strategy and assessments used to examine the
potential merits of plutonium immobilization in Russia at the three industrial plutonium
production sites, Krasnoyarsk (Zheleznogorsk or K-26), Mayak (Ozersk or Chelyabinsk-65),
and Tomsk (Seversk or Tomsk-7).

BACKGROUND

During the Cold War and until about 1994, U.S. policy employed a type of economic discard
limit (EDL) to establish the concentration of plutonium in residues and wastes below which the
production of new plutonium was estimated to be more economical than recovery (1,2). An EDL
 depended upon many factors, including plutonium concentrations and matrix forms; it was not a single numerical value but a variable that was dependent upon the types of residues and waste. Determinations whether to discard plutonium-containing material, residue, or waste; package for storage; or process to recover the plutonium were based on an EDL value. A discard determination for a specific item implied storage because the United States had no operating geologic repository for disposal. As a result, the United States has substantial inventories of impure Pu-containing materials, residues, and wastes of many types and forms that are still are in storage, and are being actively studied to establish a final disposition procedure in the United States (3,4,5).

Russia basically followed an EDL-type of approach. However, Russia required the recovery of plutonium in all residues and wastes at much lower concentration levels than did the U.S., where they were simply discarded and placed in storage. In addition, a single numerical EDL value was used for all plutonium-containing materials, residues, and wastes, regardless of Pu concentration and matrix form. The Russian policy was that all plutonium would be recovered from Pu-containing materials, residues, and wastes until the concentration of plutonium was below 200 mg of Pu /kg of solids (200 ppm or 0.02 wt%) in any discharged waste stream. Discharged waste streams were then solidified with cement and sent for near surface disposal. This 200-ppm EDL value used in Russia is orders of magnitude lower than the typical U.S. EDL, and did not depend upon the solid matrix form of the plutonium. As a result, in the 1960s, Russia developed and installed extensive plutonium recovery processes in the plutonium production sites at Mayak, Tomsk, and Krasnoyarsk, and did not discard, package, and store Pu-containing materials, residues, and wastes as did the United States. These recovery processes are in operation even today at the industrial sites under the current Russian policy of plutonium recovery. As a result, Russia does not have substantial inventories of impure plutonium to consider for immobilization and geologic disposal. Thus, it is the immobilization of future plutonium-containing materials, residues, and wastes in Russia that must be considered because current inventories are minimal.

In summary, these decades of different industrial-scale practices in Russia and the United States have led to fundamental differences in philosophies and approaches to the disposition of excess plutonium. Basically, Russia regards as valuable and recovers for reuse ‘all’ plutonium-containing materials to levels of 200 ppm Pu, and still operates industrial site plants today for this purpose. Therefore, it is the inventories of plutonium residues and wastes generated in the future that can be immobilized in Russia, with the single exception of some existing inventories of sludges at the production sites from past Pu reprocessing operations that contain mixtures of weapons plutonium and fission products. For these reasons, Russia does not consider immobilization of concentrated impure Pu-containing materials (>50 wt%) to be a credible disposition option although the United States is actively examining its potential (5). However, as part of the Joint U.S.-Russia plutonium disposition activities, Russia is examining the options and the merits of immobilizing some of the less concentrated (as high as approx. 1–13 wt%) Pu-containing materials, residues, or wastes at the Krasnoyarsk, Mayak, and Tomsk plutonium processing sites. The specific ranges of plutonium concentrations and inventories are now being established in various studies by Russian organizations.
STRATEGY FOR PLUTONIUM IMMOBILIZATION IN RUSSIA

As part of the Joint U.S.-Russian plutonium disposition activities initiated in January 1995, a strategy for the stabilization and immobilization of excess Russian weapons origin plutonium has been developed (6) and is being implemented. The objective is to develop the capability for industrial scale stabilization and immobilization of Russian impure Pu-containing materials, residues, and wastes at one or more of the three plutonium processing sites in Russia by the year 2004.

The strategy is composed of three distinct types of activities, scheduled in two separate phases. Phase 1 is underway now in Russia. Phase 1 activities assess the specific industrial sites and their sources of impure Pu-containing material that could be immobilized. Phase 1 ends with recommendations of specific plutonium immobilization forms and processes, including the merits of implementing such immobilization processes at an industrial site compared to the site’s current processes and practices of recovery and purification of plutonium for reuse. Phase 2 starts if a decision is made to continue the development and implementation of a plutonium immobilization process at a specific site. Phase 2 ends when radioactive operations with plutonium for immobilization processes are started.

The three strategic activities are: (1) research and development; (2) engineering, design, and construction (EDC); and (3) large tests and demonstrations:

• R&D activities are focused on supporting plutonium immobilization requirements at the various industrial sites. These activities include the development and characterization of plutonium glass and ceramic immobilization forms and processes.

• The EDC activities are focused on establishing plutonium immobilization processes and facilities at the plutonium processing industrial sites of Mayak, Krasnoyarsk, and Tomsk. The initial or Phase 1 EDC activities include system analyses and engineering feasibility studies to identify and select plutonium immobilization processes based on the site-specific sources of impure plutonium. Once a specific immobilization process is selected, determined to be feasible, and approved for implementation at an industrial site, the various phases of design will be conducted so that the immobilization facilities can be constructed and started up. Existing technologies, equipment, and facilities are to be utilized to the maximum extent practical.

• The third type of activity, large tests and demonstrations, are on a scale beyond that of R&D. They are focused on generating the engineering data needed by the EDC activities.

Based on the Phase 1 strategy, Russian organizations are studying the options for changing from recovery of impure Pu for reuse—their current practice—to direct immobilization of future impure Pu-containing materials into solids with Pu concentrations higher than 200 ppm. The impacts of discarding Pu-containing materials with concentrations greater than 200 ppm (0.02 wt%) to as high as 1–13 wt% are being assessed. Impacts are being measured by comparisons of economics, schedules, worker radiation exposures, and environmental impacts, including the need for geologic disposal.
ASSESSMENTS OF THE MERITS OF PLUTONIUM RECOVERY FROM RESIDUES AND WASTES IN RUSSIA IN PROGRESS AT INDUSTRIAL SITES

The Ministry of the Russian Federation for Atomic Energy (Minatom) and the Russian Institutes understand that the current practice of Pu recovery is labor-intensive in terms of operations manpower and the maintenance of process equipment. However, it does allow the Russian industrial sites to use on-site surface burial of cemented “low-level” waste streams and to avoid accumulating stored residues or Pu-containing materials that would later require final disposition decisions. The plutonium recovery operations also generate many secondary wastes and perhaps increase worker radiation exposures, both of which are undesirable factors. Changing to different industrial site operations that cease plutonium recovery and instead directly immobilize Pu-containing wastes with much greater than 200 ppm of plutonium may reduce the Russian plutonium recovery operational costs and other undesirable factors. However, these new practices would require that interim storage of immobilized forms be performed until geologic disposal of the immobilized waste forms can be implemented at Russian sites. Surface burial of such Pu-containing immobilized forms with >200 ppm of plutonium would not be acceptable as it is for cemented waste forms with <200 ppm of Pu-containing materials. In addition, Russian sites without immobilization technologies would also need to install such technologies and retrain their work forces as they shut down current plutonium recovery operations. The decision to change, or not change, the decades of plutonium processing practices at industrial sites in Russia is complex.

Engineering Feasibility Studies

Engineering feasibility studies, funded in part by the U.S. Department of Energy (DOE), are in various stages of progress by Russian design organizations, industrial sites, and institutes to systematically evaluate and compare the current plutonium recovery processes with various alternative future immobilization processes for Krasnoyarsk and Mayak. The studies address the Russian site-specific needs, requirements, capabilities, and existing and future inventories of Pu-containing materials, residues, and wastes. For example, future plutonium disposition operations all generate some Pu-containing materials, residues, and waste streams containing >200 ppm Pu. These future operations of interest include:
- Weapon component plutonium melting (for making ingots for storage and conversion)
- Plutonium metal conversion to oxide (for making nuclear fuel pellets)
- MOX fuel fabrication (for using plutonium from excess weapons components in reactors).

Current Phase 1 engineering feasibility studies are underway to assess whether such low plutonium concentrations might be better immobilized for geologic disposal rather than processed to recover plutonium with extensive aqueous processing.

These Russian Phase 1 studies will be used for future decisions of how, and if, to change the current Pu processing operations and recovery practices at the industrial sites, and how to assess
the current Minatom and Russian policy position that “all” plutonium is worth recovering for its energy value. Basically, the word “all” is being examined today in Russia by the Phase 1 engineering feasibility studies. These studies also address interim storage and geologic disposal requirements for immobilized forms, as well as job force reductions, staff retraining issues, and special transportation and packaging needs required to implement immobilization of Pu-containing wastes at the Russian industrial sites. The Phase 1 feasibility studies are also identifying the Russian Federation, regional, and local laws that would require changes for implementation of immobilization options.

The initial engineering feasibility studies are regarded as noncommittal, Phase 1 activities of the Russian strategy that are summarized in Ref. 6. However, they are key elements in establishing a technical baseline of how Russia could change over to immobilizing some types of Russian Pu-containing wastes containing more than 200 ppm of plutonium. Tomsk is more reluctant to agree that a Phase 1 engineering feasibility study is meaningful than are Krasnoyarsk and Mayak. This is, in part, because Tomsk lacks experience with radioactive glass immobilization technology, and is not considering geologic disposal of solid wastes.

**PLUTONIUM IMMOBILIZATION APPROACH FOR INDUSTRIAL SITES AT MAYAK AND TOMSK**

Figure 1 below illustrates schematically an approach for plutonium immobilization that could be applied to the two major plutonium processing sites at Mayak and Tomsk, which still perform strategic and other plutonium processing operations. Notice the fact that any possible plutonium disposition option in Russia requires a geologic repository (end box, Fig. 1). In this figure, we

- Compare plutonium recovery vs. immobilization for current and future Pu-containing wastes
- Recognize sensitive transparency issues
- Recognize that a Russian geologic repository is required for any disposition options.

Figure 1, which assumes the Mayak site, illustrates the potential disposition pathways for excess plutonium in Russia and the role that immobilization can provide for the industrial site and for the future plutonium conversion and MOX facilities assumed be located there. Russia has not yet made site selection decisions for plutonium disposition so the figure is intended for illustration only. Similar variations can be drawn in a figure for Krasnoyarsk or Tomsk.

The pathways differ significantly from the U.S. immobilization disposition pathways because of different Russian policies and practices in plutonium processing. The major differences stem from the Russian practice, which was, and remains today, to recover plutonium from all residues and wastes until the Pu concentration is <200 ppm in the waste stream. Then it is made into a cement solid form for near surface burial. This plutonium recovery practice is illustrated by the dashed lines labeled “Pu wastes” on the right side of Fig. 1. As a result, Russia has no substantial inventories of impure plutonium in concentrations greater than ~1% that can be considered for immobilization.
Figure 1. Diagram of options for the immobilization of Pu-containing materials at a single Russian industrial site compared to plutonium recovery.

Some inventories of impure weapons origin plutonium could be immobilized; these exist in sludges at low plutonium concentrations and together with products from past Pu reprocessing operations at Mayak, Krasnoyarsk, and Tomsk. In addition, future plutonium disposition operations in Russia (e.g., plutonium weapons component melting, plutonium conversion, and MOX fuel fabrication for utilization of excess Russian plutonium in reactors) will generate Pu-containing materials, residues, and wastes that could be immobilized rather than sent to plutonium recovery operations. Future operations associated with maintenance of the Russian strategic weapons at the Chemical Metallurgical Plants (CMP) at Mayak and Tomsk will also generate Pu-containing materials residues and wastes that could be immobilized rather than processed for recovery. Thus, as illustrated in Fig. 1, a single immobilization facility could be considered for an industrial site to immobilize multiple sources of Pu-containing materials—including current plutonium HLW sludges, future residues and wastes from the main plutonium melting, Pu metal conversion, and MOX fuel fabrication, as well as from future CMP plutonium operations.
Weapons Plutonium Classification Issues

The plutonium weapons component melting operations illustrated in Fig. 1 are used to prepare metal ingots suitable for long-term storage and conversion and to remove the Russian sensitive classified information regarding weapon component shapes and masses. Shapes and masses of specific weapon components are two of three major types of sensitive classified design information (in both the U.S. and Russia). Classification requirements prevent full disclosure of these plutonium attributes between both sides. Some small amounts of Pu-containing wastes and residues are generated in these melting operations. Any new plutonium metal conversion facilities and new MOX fuel fabrication facilities will also generate Pu-containing residues and wastes requiring either recovery or immobilization. Figure 1 illustrates that a single immobilization facility for these Pu-containing residues and wastes and the HLW sludges at, for example, Mayak could be used to produce stabilized solid waste forms suitable for storage until a geologic repository becomes available in Russia. If the Pu-containing residues and wastes and the HLW sludges were immobilized, then it would not be necessary to recover the plutonium if the current Russian policy toward recovery were changed.

As indicated in Fig. 1, the mixing of Pu-containing HLW sludges with residues and wastes from the future Russian plutonium disposition operations would likely remove the Russian classification sensitivity of the weapons plutonium isotopes. Russia currently regards the isotopics of weapons origin plutonium as sensitive and classified while the U.S. does not. This difference leads to some currently unresolved transparency issues between the U.S. and Russia that are being actively discussed. It can be noted in Fig. 1 that the Pu isotope Russian classification issue will remain a serious unresolved transparency issue throughout conversion, MOX fuel fabrication, and until the MOX fuel is actually irradiated in a reactor. Immobilization using the existing Pu-containing HLW sludges at a site like Mayak offers an early opportunity to possibly remove the Russian classified plutonium isotopic information.

Geologic Repository Requirement

Figure 1 also illustrates that the irradiated MOX spent fuel requires storage until a future decision is made whether to reprocess the spent MOX fuel or to implement a direct geologic disposal option. The Russian policy is storage until the reprocessing and plutonium recovery for recycle option is implemented. It is important to note that a geologic repository is required no matter which plutonium disposition path is followed for spent MOX fuel (i.e., either for immobilized HLW in a closed fuel cycle or for spent MOX fuel in an open fuel cycle). The geologic repository is, of course, required for plutonium immobilized forms.

Mayak and Tomsk CMPs

Finally, it is worth commenting on the CMPs at Mayak and Tomsk. These plants currently operate Pu recovery operations and follow the current Russian policy of recovering plutonium in waste streams and residues until discarded waste streams are below 200 ppm. The two plants
generate different types of wastes and residues and use different plutonium recovery operations. Mayak generates Pu-containing wastes and residues containing 1–13% plutonium while the Tomsk site generates Pu-containing wastes and residues containing 1–5% plutonium. Mayak uses a high-temperature (800°C) NaOH fusion process to destroy the chemical matrix of their hard-to-dissolve residues with 1–13% plutonium. The hard-to-dissolve residues are those that are not recovered in the first or second acid leach recovery cycles. The fused residues are then leached in acid, the plutonium purified and recovered by solvent extractions. The remaining leached fused residues contain <200 ppm plutonium, which are then cemented and discarded.

Tomsk does not use the high-temperature NaOH fusion process but instead uses multiple acid and alkaline leach solution cycles of their hard-to-dissolve Pu-containing residues with 1–5% plutonium. These plutonium recovery operations at Mayak or Tomsk generate large amounts of secondary wastes, induce some additional worker radiation exposures, and require additional equipment process and maintenance operations. Discussions are in progress on the merits of performing Phase 1 engineering technical feasibility studies to systematically quantify and compare the costs and benefits of plutonium recovery with a new option of direct immobilization of the CMP Pu-containing wastes and residues. However, no Phase 1 studies have yet been initiated as there are no operational problems with their recovery processes to date.

PLUTONIUM IMMOBILIZATION APPROACH FOR THE INDUSTRIAL SITE AT KRASNOYARSK

An engineering feasibility study for the Krasnoyarsk site for immobilization of Pu-containing sludges from past plutonium reprocessing operations is underway. The site has no CMP so a different scheme is being examined than was discussed for Mayak and Tomsk. The Krasnoyarsk (K-26) site has 6000 m³ of sludges containing an estimated 600 kg of weapons plutonium. The study compares recovery and purification of the 600 kg of plutonium as required by current Russian policy with non-recovery and direct immobilization by vitrification of the sludges using two different glass formulations, namely, a phosphate and a borosilicate composition. R&D activities are being conducted to develop a glass or glass-like composition for immobilizing the sludges using a microwave melting method at two Russian institutes, the A. A. Bochvar Research Institute of Inorganic Materials and the V. G. Khlopin Radium Institute. The one-year initial Phase 1 feasibility study will be completed in June 1999. Figure 2 illustrates the three options being developed and compared in the ongoing engineering feasibility study. The comparisons are being made in terms of costs, secondary wastes, radiation exposures, and environmental impacts. Note that a geologic repository is needed for any of these immobilization options for solid wastes.
Figure 2. Three options are being evaluated for the Krasnoyarsk site in an ongoing Phase 1 engineering feasibility study.

CONCLUSIONS

Immobilization is being examined as a viable option in Russia for some excess weapons origin Pu-containing materials at Krasnoyarsk, Mayak, and perhaps Tomsk.

It is not reasonable to expect the immobilization of excess weapons plutonium in Russia to follow the same pathway as the one that the United States is actively examining. Significant
differences in the approaches to plutonium processing were followed for decades by the United States and Russia during the Cold War and are still in place today in Russia.

Designing and constructing a single plutonium immobilization process at a Russian industrial site for disposition of Pu-containing HLW sludges generated from past plutonium production operations and Pu-containing materials, residues, and wastes from future and current plutonium operations appears to offer many benefits.

Phase 1 engineering feasibility studies for Mayak and Tomsk are being discussed that could establish the potential benefits of immobilizing Pu-containing materials, residues, and wastes rather than continuing the current plutonium recovery operations where any discharged waste stream contains <200 ppm of plutonium. The Phase 1 study of plutonium immobilization is well underway at Krasnoyarsk and others are anticipated to follow.

ACKNOWLEDGMENTS

Work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

REFERENCES