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DISCLAIMER

DISPOSITION OF SURPLUS FISSILE MATERIALS VIA IMMOBILIZATION

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Abstract

In the Cold War aftermath, the U. S. and Russia have agreed to large reductions in nuclear weapons. To aid in the selection of long-term management options, the U.S. DOE has undertaken a multifaceted study to select options for storage and disposition of surplus plutonium (Pu). One disposition alternative being considered is immobilization.

Immobilization is a process in which surplus Pu would be embedded in a suitable material to produce an appropriate form for ultimate disposal. To arrive at an appropriate form, we first reviewed published information on HLW immobilization technologies to identify forms to be prescreened. Surviving forms were screened using multi-attribute utility analysis to determine promising technologies for Pu immobilization. We further evaluated the most promising immobilization families to identify and seek solutions for chemical, chemical engineering, environmental, safety, and health problems; these problems remain to be solved before we can make technical decisions about the viability of using the forms for long-term disposition of Pu. All data, analyses, and reports are being provided to the DOE Office of Fissile Materials Disposition to support the Record of Decision that is anticipated in Summer of 1996.

Introduction

Background

The end of the Cold War has brought the arms and nuclear materials production race to a close and, as a result, significant quantities of Pu and HEU have become surplus to defense needs in both the U. S. and Russia. With the collapse of the Soviet Union and the economic and social challenges faced by newly formed states, these growing stockpiles are a serious risk for nuclear proliferation. The National Academy of Sciences[1] (NAS) report on the management and disposition of excess weapons Pu characterized this as a "clear and present danger." This nuclear danger is in many ways more diffuse, harder to manage, and stronger than the nuclear tensions of the Cold War era. The dangers exist not only in the potential proliferation of nuclear weapons but in the potential for environmental, safety, and health consequences if these fissile materials are not properly managed.

In response to the President's nonproliferation policy, DOE Secretary O'Leary created a Department-wide project for control and disposition of surplus fissile materials on January 24, 1994, which later that year became the Office of Fissile Materials Disposition (MD). MD was charged with coordinating efforts for the long-term storage and disposition of surplus weapons-usable fissile materials within the Department. MD,

through the various task teams, used a screening process that considered all potential long-term storage and disposition options, evaluated them against screening criteria, and identified alternatives reasonable for evaluation in the Storage and Disposition PEIS. The screening criteria, developed with input from the public during the PEIS scoping process, reflect the President's Nonproliferation and Export Control Policy of September 1993 and the January 1994 Joint Statement by the President of the Russian Federation and the President of the United States of America on Nonproliferation of Weapons of Mass Destruction and the Means of Their Delivery. The analytical framework established by the NAS study also contributed to the basis for the development of the screening criteria.

The "Spent Fuel Standard"

The primary goal of disposition is to render weapons-usable fissile materials inaccessible and unattractive for weapons fabrication while protecting human health and the environment. The NAS report recommended that Pu disposition strategies work to attain a "spent fuel standard." This standard would require a disposition form from which extraction and weapons use of any residual Pu and other fissile materials are at least as difficult or unattractive as the recovery of residual Pu from spent commercial nuclear fuel. When a disposition strategy achieves this standard, the proliferation risk associated with the residual surplus material is neither unique nor greater than that associated with the much greater inventory of residual Pu in spent fuel. Achieving the spent fuel standard implies that any residual Pu is embedded in a highly radioactive matrix, mixed with a variety of other chemical species on a substantially homogeneous basis, and packaged in a large, bulky, integral form which provides barriers to reuse.

Immobilization with Radionuclides

Immobilization Task Team Members

To support the PEIS for Disposition of Pu, MD chose Lawrence Livermore National Laboratory (LLNL) as Lead Laboratory to study and recommend methods for transforming Pu into long-term immobilized forms; to provide appropriate input to other Disposition Tasks Teams so as to assess the technical feasibility of immobilization as a long-term disposition option; and to describe infrastructures required to dispose of Pu. Support laboratories include Westinghouse Savannah River Technology Center, Argonne National Laboratory, Oak Ridge National Laboratory, and Pacific Northwest Laboratory. At the direction of MD, LLNL further secured the aid and help of a number of university and industrial partners; these include MIT, U. C. Berkeley, Clemson University, COGEMA, British Nuclear Fuel Ltd., and the Australian Nuclear Science and Technology Organisation.

Technology Selection

Under the immoblization alternative, surplus Pu would be immobilized in an acceptable matrix to create a chemically stable form for disposal in a high-level waste repository (or other alternative disposal system). The fissile material would be mixed with high-level wastes or other radioactive isotopes and immobilized to create a radiation field that could serve as a proliferation deterrent comparable to commercial spent nuclear fuel. To be economically viable, the Pu concentration must be in the range 0.4 to 10 wt%. To arrive at suitable forms, a literature search identified 72 waste forms[2] (72 unique names, only 45 unique forms) that have been considered for immobilizing radioactive wastes. A screening process was implemented to select the best waste forms for the Pu disposition application. A more detailed discussion of the screening process is

given elsewhere;[3] a summary of the process is given below.

A two-stage approach based on decision analysis techniques, was adopted for screening. This allowed more rigorous selection techniques to be used as options became more closely matched. Stage 1 applied a small set of criteria (laws and regulations) to a large number of forms; this procedure quickly eliminated forms that were clearly inappropriate for Pu immobilization. After prescreening, candidate forms were further screened using multi-attribute utility analysis to determine the most promising technologies for Pu immobilization. Promising immobilization families[4–11]—glass, ceramics, and metals—were then evaluated to identify inherent chemical, engineering, environmental, safety, and health problems and to seek solutions for them before making technical decisions about the viability of any of the families for long-term disposition of Pu.

Processing Options

Seven immobilization options comprising these three immobilization forms are being evaluated in the PEIS/Record of decision process:

A. Vitrification
   - Internal radiation barrier
     New vitrification facilities
     Defense Waste Processing Facility (DWPF) adjunct melter
   - External radiation barrier
     DWPF Can-in-canister
   - *Glass Material Oxidation and Dissolution System (GMODS) variant*

B. Ceramics
   - *Internal radiation barrier*
   - *External radiation barrier*

C. *Electrometallurgical treatment*

Three candidate sites are being considered for immobilization facilities: Hanford, INEL, and SRS.

National Academy of Sciences Objectives for Safeguarding and Monitoring

In their study,[1] the NAS suggested that the immobilization program must be designed to meet three key security objectives:

1. Minimize the risk that weapons materials could be obtained by unauthorized parties.
2. Minimize the risk that weapons materials could be reintroduced into the arsenals from which they came.
3. Strengthen the national and international control mechanism and incentives designed to ensure continued arms reduction and prevent the spread of nuclear weapons.

It follows from these objectives that security, rather than energy consideration or cost, may be the primary criterion in choosing disposition technologies. That is, options and technologies may be chosen so that safeguarding and monitoring procedures can be effectively implemented throughout the overall disposition process.

In particular, the objectives for safeguarding (materials control and accountancy, containment and surveillance, and physical protection) and monitoring (bilateral or international through the IAEA) will probably include the following:

- Preventing theft and unauthorized use.
- Demonstrating that the control, accounting, and security for separated excess Pu are comparable to those for nuclear weapons.
- Detecting any reuse for weapons by the host country.

Preventing Theft and Unauthorized Use

The first step of proliferation resistance is provided by the form of the material, e.g., bulk, weight, and radiation field. The package being planned for immobilization is similar to the standard package planned for the immobilization of HLW by vitrification at the DWPF. This container is about 0.6 m in

diameter and about 3 m long; the nominal mass is about 2200 kg. The combination of size and weight, physical protection measures (such as gates, guns, and guards), and a radiation barrier forms significant impediments. Although bulk and weight need little comment, we further discuss radiation field below.

In most schemes being studied, the radiation field is assumed to be supplied either by HLW or by ^{137}Cs from the CsCl capsules now stored at Hanford). The strength of the radiation field has not yet been decided. The "self protecting" standard for spent fuel is usually taken to be 1 Sv/hr (100R/h) at 1 m. Unless other physical security measures are taken, 1 Sv/h is not considered sufficient for immobilized Pu. For a lethal dose (LD_{50}, a 50% chance of death), an exposure of about 4.5 Sv (450 rads) is required. The dose rate at one meter from the edge of an LWR fuel assembly along the perpendicular bisector to the axis, which is the direction of maximum dose, is about 60 Sv/h (6000R/h) for a 5-year cooled fuel of 33 GWd/MT burn-up. Vitrified HLW canisters from DWPF are anticipated to generate a dose rate of up to 9 Sv/hr (900 R/h). The Immobilization Task Team is using a tentative planning radiation field of 10 Sv/h (1000 R/h). Dose rates of about 10 Sv/h (1000 R/h) would be considered "self-protecting."

By itself, achieving the spent fuel standard does not afford sufficient proliferation resistance; the material must still be properly located and/or properly safeguarded. Furthermore, it is possible that sufficient proliferation resistance can be achieved without embedding the immobilization form in a highly radioactive matrix, such as emplacement in a deep borehole, where geologic location imposes a barrier significant against clandestine retrieval of the material. After the Pu has been sealed in a geologic repository or borehole, the radiation barrier is augmented and replaced by lack of accessibility, i.e., an isolation barrier.

Demonstrating Separated Pu Control, Accounting, and Security

The Disposition Program encompasses all isotopes of Pu, except ^{238}Pu, in weapons-usable quantities in various forms such as pits, metals, oxides, scrap, compounds, and residues, as well as ^{233}U and other minor actinides (if needed). Physical protection, accountancy and control measures for repository disposal of Pu are necessary whether the Pu is embedded with HLW components or ^{137}Cs isotopes in glass, crystalline ceramics metals, or other materials. The measures that must be imposed on geological-disposed Pu will, until the repository is permanently sealed, be similar to those required for surface storage of separated Pu. Inventorying would likely be supplemented by containment and surveillance methods such as portal-perimeter monitoring. Sensors might include motion sensors on the perimeter and gamma and/or neutron detectors at the portals.

The materials control and accountancy (MC&A) for immobilized Pu should not present any unusual difficulties. It amounts to ensuring that containers of immobilized Pu are accounted for and have not been tampered with. Tags or permanent bar codes on the immobilization containers would facilitate inventorying; with the use of robots this possibly could be done remotely. Constant visual surveillance using television combined with weight, motion, and radiation sensors could be used to detect the removal or tampering with any container.

This type of inventorying and surveillance of individual immobilization containers would begin at the facility where the Pu was immobilized. Inventorying is important to ensure that no Pu is diverted prior to immobilization, as well as to evaluate the security and criticality risks. Methods employed to safeguard mixed-oxide fuel fabrication facilities, such as used in Japan, and large reprocessing facilities, such as in

France and England, could be employed in an immobilization facility.

Detecting Reuse by Host Country

Pu has a unique chemistry. It can exist in solution in four oxidation states. The plus four oxidation state (Pu[IV]) is also unique in that in a nitrate medium it is one of the few elements that form an anion—$Pu(NO_3)_6^{++}$. A few elements also form neutral complexes with nitrate. By manipulating the oxidation state, especially in nitrate medium, it is possible to separate Pu from all other elements relatively easily—actually the more difficult elements to remove from Pu are U, Th, Np, Au, Pt, Ir, and Pd. Therefore one pass through an anion exchange column is sufficient to purify Pu sufficiently to be used in a crude weapon even if each of these elements has a concentration 10 times that of the Pu. Very high recovery efficiencies can be obtained with simple equipment, even with poor technique. Solvent extraction and anion exchange are the most common Pu separation methods used worldwide, but a number of pyrochemical techniques could also be used. The deterrent that the immobilization form can offer to Pu reuse in weapons is the time necessary to dissolve the immobilization medium. Ceramic forms are the most difficult form to dissolve and hence offer the most deterrent to the host nation to reuse the Pu.

Regardless, this only adds time and cost to recover, purify, and reuse the Pu in any fashion which the host nation saw fit. As a result, only adversarial verification and monitoring can detect the possibility that the host nation is attempting to reuse the Pu.

Summary

An International Team was assembled for the express purpose of selecting suitable immobilization forms and processing technologies for the Surplus Pu Disposition Program Fissile Materials Disposition Program Office. The Task Team used the NAS Study as a reference point for starting the evaluation but was not limited to the recommendations of the NAS. Three basic immobilization forms have been selected and the processing options to provide those three forms have been defined. Environmental Data has been supplied to the DOE contractor preparing the PEIS for the Disposition Program. The Task Team is now developing cost data for the Record of Decision which is anticipated in the Summer of 1996. The Task Team is also evaluating these options to determine whether they indeed meet the security standard and goals set up by the NAS Study.

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