Vitrification of Excess Plutonium (U)

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VITRIFICATION FOR DISPOSITION OF EXCESS PLUTONIUM

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ABSTRACT

As a result of nuclear disarmament activities, many thousands of nuclear weapons are being retired in the U.S. and Russia, producing a surplus of about 50 MT of weapons grade plutonium (Pu) in each country. In addition, the Department of Energy (DOE) has more than 20 MT of Pu scrap, residue, etc., and Russia is also believed to have at least as much of this type of material. The entire surplus Pu inventories in the U.S. and Russia present a clear and immediate danger to national and international security. It is important that a solution be found to secure and manage this material effectively and that such an effort be implemented as quickly as possible.

One option under consideration is vitrification of Pu into a relatively safe, durable, accountable, proliferation-resistant form. As a result of decades of experience within the DOE community involving vitrification of a variety of hazardous and radioactive wastes, this existing technology can now be expanded to include immobilization of large amounts of Pu. This technology can then be implemented rapidly using the many existing resources currently available. A strategy to vitrify many different types of Pu will be discussed. In this strategy, the arsenal of vitrification tools, procedures and techniques already developed throughout the waste management community can be used in a staged Pu vitrification effort. This approach uses the flexible vitrification technology already available and can even be made portable so that it may be brought to the source and ultimately, used to produce a common, borosilicate glass form for the vitrified Pu. The final composition of this product can be made similar to nationally and internationally accepted HLW glasses.

BACKGROUND

The Committee on International Security and Arms Control (CISAC) of the National Academy of Sciences (NAS) was commissioned by General Scowcroft, National Security Advisor to President Bush, to conduct a study on disposition alternatives for management of excess plutonium resulting from disarmament activities. This charter was later confirmed by the Clinton Administration. After receiving input from many sources and evaluating the options, NAS recently issued the results of this study in a report entitled "Management and Disposition of Excess Pu" [1] in January of 1994. The treatise covered many important aspects of the Pu disposition question and with respect to long-term management of the excess Pu stated the following:

"Then two most promising alternatives for achieving these aims (long-term Pu disposition) are:

• fabrication and use as fuel, without reprocessing, in existing or modified nuclear reactors; or
• vitrification in combination with high-level radioactive waste."
A third option, which has not been studied in as much detail, burial of Pu in deep boreholes, was also mentioned as a possible consideration.

As a result of the NAS study, the vitrification option was elevated to the same level of importance as a reactor option and listed as one of two leading preferences for ultimate disposition of weapons grade Pu.

The Savannah River Site (SRS) has been involved in vitrification of high level waste for several decades and also associated with vitrification of a variety of other types of radioactive and non-radioactive materials [2,3]. As a result of over 20 years of vitrification experience and about 40 years of Pu handling and processing experience, the site has been requested to provide input into this subject by a variety of groups and agencies addressing the Pu disposition problem [4-7]. The SRS efforts address not only management of weapons grade Pu, but also Pu scrap, residues, etc. that exist within the DOE complex. An early summary of vitrification options is documented elsewhere [8].

WHY VITRIFICATION

There are many potential advantages associated with the vitrification option for long-term management of plutonium. These include the following:

Immediacy of Implementation:

Many experts in the field believe it is important to act quickly to immobilize Pu for security, safeguards, safety and environmental reasons. Because of the advanced state of the art of vitrification and as a result of the existing capabilities and experience in this area, there is no other Pu option that can be implemented as rapidly.

Flexibility:

The vitrification option provides a common technology for treatment of almost all forms of Pu. This includes not only weapons grade Pu, but also significant quantities of more complex Pu scrap and residue compositions, currently existing within the DOE community and posing additional problems.

Technology Availability:

As a result of the High Level Waste program and associated waste management efforts, there have been decades of research on development of techniques and procedures for vitrifying radioactive and hazardous components, equipment and facilities for these tasks, and in specifying waste form qualifications to assure product quality. These efforts are directly applicable to Pu vitrification and could be piggy-backed upon. A very extensive and capable vitrification infrastructure exists containing experienced and dedicated experts throughout federal and national laboratories, academia, and industry throughout the United States as well as in other countries.

Waste Glass Performance:

HLW glasses have been demonstrated to have excellent chemical durability, mechanical integrity, radiation and thermal stability. Chemical durability is considered to be the most important technical performance property of a waste glass form. It is important to note that actinide bearing glasses exhibit excellent chemical durability. The leaching of
actinides is generally 10-100x better (lower leach rates) than modifiers or alkali cations contained in HLW waste glass systems.

**Processing Considerations:**

The ability to vitrify radioactive materials is not only well developed, but also well demonstrated. In the case of HLW, actual production facilities are in operation worldwide. These include the French process in Marcoule and also La Hague, the German vitrification operation in Mol Belgium, the Sellafield facility in England and others. Construction of the first HLW vitrification facility in the United States, the Defense Waste Processing Facility (DWPF) at SRS, has recently been completed and is scheduled to be in production in about one year. Vitrification of Pu represents an extension of this already available vitrification technology and its many components.

**Waste Minimization:**

Due to the type of equipment and buildings needed to process radioactive materials, and as a result of the vitrification facilities and equipment used to treat other types of radioactive wastes, existing contaminated and non-contaminated facilities could be modified to perform Pu vitrification. These facilities include buildings and equipment designed to receive Pu, store the material, process the Pu, and vitrify it into acceptable products. These facilities currently exist within the DOE complex and although they would require various degrees of modification, their existence would eliminate the need to build and later D&D additional buildings.

**Ability to Immobilize Pu in Glass:**

Immobilization of Pu into borosilicate glass has already been demonstrated in HLW programs. This early work involved immobilization of 7 wt. % of plutonium oxide into a glass matrix. Higher Pu loading is probable. Hence, there is no question whether Pu can be vitrified - the maximum amount immobilized remains to be defined and will most likely not be determined by solubility limits but by criticality considerations.

**Inherent Criticality Control:**

Due to the composition of borosilicate glasses, boron as well as lithium, which are normally present in HLW forms, can act as poisons to assist in criticality control. This is especially important during handling and processing operations. Other poisons such as gadolinium and rare-earths would be added to further this effect, which would be especially relevant for long-term repository storage scenarios due to their insolubility. Additional work would be necessary to be better assess the effects of potential poisons for criticality control in some of the final disposal options under consideration and for increasing the difficulty in reclaiming Pu from the glass matrix. Criticality control represents the most important consideration in all stages of any disposal option.

**Acceptability/ Waste Form Qualifications:**

The only waste form which has achieved a degree of national and international acceptance for immobilizing HLW is borosilicate glass. HLW glasses already contains Pu, although in very small amounts. It took approximately ten years and thirty million dollars to qualify the SRS HLW waste glass composition. This important and necessary effort could be piggy-backed upon for the Pu vitrification option.
Proliferation Resistance:

There are many ways in which Pu can be immobilized into glass to produce durable, safe, proliferation-resistant forms. These options depend on the degree of proliferation resistance required and are directly proportional to the cost and complexity of the operation. For example, simply immobilizing Pu into glass can be achieved rapidly and most easily and provides the highest degree of flexibility. The Pu-glass product produced would be more proliferation resistant that Pu in its weapons form, but could be reclaimed fairly easily by those reasonably familiar with this field. The degree of proliferation resistance could be increased significantly, however, by either initially mixing the Pu directly or by re-melting Pu-only glass, with fission products or existing HLW. The radiation field associated with the radioactive additives would considerably increase the difficulty in obtaining or handling this material and in subsequent transportation and reprocessing operations to reclaim Pu. Proliferation resistance can be further enhanced by the potential size and weight of the product and most importantly, by the safeguards that would be necessary for any undertaking of this type.

OVERVIEW OF A POTENTIAL Pu STRATEGY

Vitrification provides an important option to immobilize and dispose of not only weapons grade Pu, but also many other forms of Pu of concern within weapons producing countries. The many potential advantages of this option were discussed earlier and in Figure 1, a simplified flowsheet summarizing an overall vitrification strategy is depicted.
The major steps in the Pu vitrification strategy shown above can be summarized as follows:

Pu Handling and Glass Preparation

I. Preparation (receipt, pre-storage, and pre-treatment)
   II. Conversion to melter feed
   III. Vitrification into intermediate or final Pu-glass products
Interim and Final Product Storage

IV. Interim product storage
V. Final product disposition

Following is a brief discussion of each of these important areas:

Preparation:

The vitrification strategy applies to Pu in metal form currently contained in weapons as well as Pu in other forms such as oxides, buttons, scrap, solutions, ash, salts, residues, etc. The other forms of Pu are contained at DOE sites including Rocky Flats, Los Alamos National Laboratory, Hanford, Argonne National Laboratory, Idaho National Engineering Laboratory, and the Savannah River Site. Preparation of these materials will involve receipt, disassembly/separations in some cases, assaying, interim storage and pretreatment prior to immobilization.

Conversion:

There are two major options that can be used for conversion of Pu from a metallic form to a suitable melter feed. These include (a) oxidation—burning it to produce plutonium oxide powder and (b) dissolution—dissolving it to yield a plutonium acid solution. Additional preparation involving Pu scrap and residues could be performed, depending on composition and subsequent melting characteristics. Vitrification can handle either liquid or oxide feeds.

Vitrification:

A vast array of electric melting techniques have been developed for vitrification of radioactive wastes over the years and include indirect heating, joule heating, plasma and microwave vitrification. Many of these techniques and their tested glass melters could be used to vitrify Pu. The proposed strategy considers two main options for vitrification:

- In Option A, shown in Figure 1, the treated Pu is melted directly with fission products such as Cs-137 or HLW to produce a highly radioactive Pu glass product. This could be accomplished in several ways and provides the highest degree of proliferation resistance possible. Because of the highly radioactive wastes to be mixed with the Pu, specially contained and shielded facilities and equipment would be necessary.
- In Option B, an interim Pu-only glass is first produced as an interim product. This option has the advantages of being able to use a wide range of site-specific vitrification technologies already located at sites containing the waste, can be performed by the use of relatively simple portable equipment and gloveboxes, and can be accomplished most rapidly and easily due to the relatively low levels of radioactivity involved. While the resulting Pu-only glass does not produce the most proliferation resistant glass product, it does provide the most flexibility because it can be transported to other locations that have more highly radioactive wastes, such as HLW or selected fission products, and later be re-melted with this waste to produce more proliferation resistant forms. Since specially contained facilities and equipment are necessary for this part of the operation, current facilities with other missions could be used by meshing into existing programs and schedules to produce duel missions and optimum use of existing resources.
An important concept in this strategy is that a common final waste glass form can be produced with a similar composition to HLW glass \[9\], which has undergone a very time consuming and expensive process to be certified and made acceptable. A resulting Pu-HLW glass compositions would be anticipated to be as good, if not better, than the already acceptable HLW glass compositions.

**Interim Product Storage:**

A need to store Pu bearing glasses temporarily is important to the vitrification option. The design of the interim storage facility would depend on factors such as the composition and radioactive content of the products and the intended duration of storage before ultimate disposal. Among the most important considerations for this facility are worker radiation exposure, public and environmental protection from radiologic hazards, and material safeguards and accountability.

**Final Product Disposition:**

The reference concept for ultimate disposition of HLW glass and spent fuel is to dispose of this material by deep burial in carefully selected geologic repositories. This is also being considered for Pu bearing glasses. A very significant challenge for any repository scenario is to demonstrate safe and effective performance of products out to very long time periods (1000 to 10,000 years, and longer). This challenge would be expected to be even more formidable for any immobilization alternative containing large amounts of Pu, due to criticality considerations. The waste form and waste package will be designed to prevent criticality from occurring but this must be demonstrated to a very high degree in a very complex environment. While this would not be expected to be as significant an undertaking for a "Retrievable Surface Storage Facility or RSSF", it would be expected to take much more effort to demonstrate in a geologic repository.

The final form and composition of Pu glass would be tailored for technical and political considerations, involving possible reuse or non-reuse by the weapons producer and for optimizing proliferation resistance, especially towards potential non-friendly nations.

**Vitrification of Pu Using Existing Resources**

As discussed earlier, there exists an infrastructure knowledgeable on vitrification of radioactive and hazardous wastes that could be used for vitrification of Pu. Along with the developed technology, experience and experts, are existing buildings, equipment and supporting hardware and software for such an effort. While buildings and equipment exist throughout the DOE complex, the following discussion will emphasize vitrification options and facilities associated with the Savannah River Site (SRS). These facilities are shown along with the accompanying main vitrification steps discussed earlier in Figure 2.
At least six separate vitrification options have been identified using SRS facilities. The processing options involve three potential Pu products; a Pu-only glass (interim product), a Pu/Cs-137 glass (final product) or a Pu/HLW glass (final product).

Among the main existing facilities which could be modified and are critical to these vitrification options are the following:

**Plutonium Storage Facility (PSF):**

The mission of the PSF was to receive Pu materials from offsite that would later be processed. The building contains a fully safeguarded vault for automated transport and stacking, gloveboxes for opening drums and removing and inspecting contents, and instruments for non-destructive assay and computer accountability. The facility also contains a delivery systems for later processing of materials in an adjacent facility, the New Special Recovery (NSR).

**New Special Recovery (NSR):**

The mission of the NSR was to process plutonium from throughout the DOE complex. This includes dissolving Pu scrap, oxide or metal from various sources to produce purified Pu metal buttons or oxide powder. The facility contains state of the art glovebox trains for feed preparation, waste handling, dissolution processes, samples and analysis, along with a supporting remote control room. This facility also contains extra room which could be outfitted with glass melters to pursue one of the vitrification options under consideration.

**F Canyon (Including the Multi-Purpose Processing Facility or MPPF):**

221-F Canyon was the world's first PUREX production plant used to dissolve natural and depleted uranium targets and to recover the uranium and plutonium. It is a large, heavily shielded facility operated by remote means. The building has the potential of
being refitted with a melter and used for vitrification in support of another vitrification option of immobilizing Pu directly with HLW or selected fission products. The MPPF is located within the facility and contains eight modules that are now being used for vitrification of Cm and Am using a bushing melter, and will also be used for vitrification and subsequent clean up of Pu scrap on site.

**Defense Waste Processing Facility (DWPF):**

The mission of the DWPF is to immobilize the 34 million gallons of HLW currently being stored at SRS into borosilicate glass. This represents the first waste vitrification facility constructed in the United States and is scheduled to be in production in about one year. All of the operations necessary to process, vitrify, containerize, seal and decontaminate HLW glass units are present, including supporting capabilities to control the process and verify product quality. This facility and its equipment could be modified to vitrify larger quantities of Pu than presently contained within HLW.

Important for DWPF options is not to adversely effect the current, important mission of the facility. This could most easily be achieved by the staged approach mentioned earlier. There are two primary ways that Pu vitrification could be conducted using the DWPF. First, during the scheduled melter change out of the DWPF, the facility and equipment could be modified and Pu bearing feed introduced either directly as a Pu feed or as a Pu bearing glass frit, to produce Pu-HLW glass forms in 304L stainless steel containers. These units would be similar to HLW glasses in canisters, 2-ft. in diameter and almost 10-ft. high. An important potential advantage of this option is that waste loading to the glass could be increased by 1-2%, which would still result in a highly durable form which could immobilize 50 MT of Pu as part of the HLW program, and without increasing the number of waste canisters produced.

The second main option involves placing the interim product, Pu-bearing glasses, into HLW canisters and pouring HLW over them in the current HLW vitrification campaign. While this option does not produce as integral a product as the Pu-HLW glass, it does produce a form with similar radiation characteristics (proliferation resistance) and is considerably easier and less expensive to implement into the current facility and schedule.

**SUMMARY**

Vitrification is a technically viable option to immobilize and manage Pu resulting from disarmament activities as well as a wide range of existing Pu scrap and residue compositions. A vitrification infrastructure exists from waste management programs. This includes expertise and experience, personnel, buildings, equipment and supporting capabilities, which could be used to implement vitrification of Pu in a time expedient and cost effective manner.

**REFERENCES**