UCRL-JC-133279
PREPRINT

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This paper was prepared for submittal to the
Waste Management '99 Symposium
Tucson, Arizona
February 28 - March 4, 1999

February 12, 1999

Lawrence Livermore National Laboratory

The Blending Strategy for the Plutonium Immobilization Program

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ABSTRACT

The Department of Energy (DOE) has declared approximately 38.2 tonnes of weapons-grade plutonium to be excess to the needs of national security, 14.3 tonnes of fuel- and reactor-grade plutonium excess to DOE needs, and anticipates an additional 7 tonnes to be declared excess to national security needs. Of this 59.5 tonnes, DOE anticipates that ~7.5 tonnes will be dispositioned as spent fuel at the Geologic Repository and ~ 2 tonnes will be declared below the safeguards termination limit and be discarded as TRU waste at WIPP. The remaining 50 tonnes of excess plutonium exists in many forms and locations around the country, and is under the control of several DOE Offices. In addition to the plutonium, the feed stock also contains about 17 tonnes of depleted uranium, about 600 kg of highly enriched uranium, and many kilograms of neptunium and thorium and about 8 to 10 tonnes of tramp impurities. The Materials Disposition Program (MD) will be receiving materials packaged by these other Programs to disposition in a manor that meets the "spent fuel standard."

To minimize the cost of characterization of the feedstock and to minimize purification processes, a blending strategy will be followed. The levelization of the impurities, the plutonium isotopics, and the actinide impurities will also provide some benefits in the area of proliferation resistance. The overall strategy will be outlined and the benefits of following a blending instead of a purification program will be discussed.

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

INTRODUCTION

Significant quantities of weapons-usable plutonium and highly enriched uranium have become excess to national defense needs in both the United States and Russia as a result of the end of the Cold War [1-7]. The U. S. National Academy of Sciences (NAS) has characterized these excess materials as constituting a clear and present danger to national and international security [1-2]. [The U. S. Department of Energy (DOE) has defined weapons-usable plutonium to include all plutonium except that present in spent nuclear fuel and plutonium which contains greater than 10% ^{238}Pu.] DOE, through the National Laboratories and site contractors, is actively working to support the nation's response to that threat. The recommendation of the NAS [1-2] was that options for long-term disposition of the excess plutonium should seek to meet the "spent fuel standard." The NAS definition of the "Spent fuel standard" is to "make the weapons-usable plutonium roughly as unattractive and as inaccessible for retrieval and weapons reuse as the residual and growing stockpile of plutonium in spent fuel from commercial reactors." [1-2] To dispose of the excess fissile materials in a fashion that meets the "spent fuel standard" DOE, on behalf of President Clinton, has announced the United States will pursue, in parallel, a dual-track disposition strategy [8] that allows for:

- Immobilization of surplus plutonium in ceramic forms embedded in high level waste glass; and
- Burning of surplus plutonium as mixed oxide fuel in existing reactors.

Some of the plutonium declared excess consists of "pits" or triggers from dismantled weapons. This material meets the weapons specifications for impurities. With the possible exception of gallium, it should be relatively easy to convert this material to plutonium oxide suitable for use in reactors as MOX fuel. The remainder of the surplus plutonium consists of:

A. Residues from weapons manufacture;
B. Materials left in process when the weapons complex was shut down at the end of the Cold War;
C. Experimental reactor fuels that were prepared but not irradiated; and
D. Other miscellaneous materials.

Preparing these for use in MOX fuel would be very costly and result in the absorption of large doses of radiation.

DOE has directed the Lawrence Livermore National Laboratory to develop the processes to immobilize this material in a titanate-based ceramic form and then embed the ceramic forms in high level waste glass. The preferred site for this immobilization operation is the Savannah River Site in South Carolina.

The material to be immobilized exists in many forms and locations around the country, and is under the control of several DOE program offices. It is assumed that the Materials Disposition Program (MD) within the Office of Fissile Materials Disposition will be receiving excess fissile materials packaged by the Offices of Defense Programs (DP), Environmental Management (EM), and Nuclear Energy (NE) facilities. It is assumed that the EM facilities will either prepare the plutonium as necessary to meet nonproliferation concerns and transportation requirements for transfer to the Waste Isolation Pilot Plant (WIPP) or stabilize the material for transfer to MD. It is also assumed that DP and NE will package their excess materials as necessary and transfer them to MD.

It is an objective of the plutonium immobilization project to:

A. Accommodate the full range of plutonium feedstocks while minimizing feed characterization costs;
B. Minimize handling/processing of the material and therefore minimize the radiation dose to operators;
C. Minimize disruption to the EM (94-1) Stabilization Program; and
D. Minimize the final immobilization product variations and formulation space.

The planned immobilization facilities will have only limited capabilities to remove impurities prior to preparing the plutonium feedstocks for immobilization. Instead, blending will be done to levelize the impurities, the plutonium isotopics, and the uranium content.

Technical specifications have been published which allow potential feedstocks to be categorized as either acceptable for transfer into the immobilization processes, or unacceptable without additional processing prior to transfer into the immobilization processes. These feedstock requirements should allow cost-benefit analyses to be preformed to determine whether a specific material should be:

1. Transferred to MD for immobilization;
2. Transferred to MD for conversion or other preparation prior to Immobilization;
3. Pre-processed on site to meet immobilization specification prior to transfer to MD;
4. Transferred to another site which can do the required processing prior to transfer to MD; or
5. Prepared for shipment to WIPP for geologic storage.

Preliminary analyses of the full 50 tonnes suggest that:

1) Approximately 45 tonnes of the excess plutonium have tramp impurity concentrations much lower than the plutonium immobilization acceptance specifications. These materials can be used to blend down the tramp impurities in the remainder of the material.

2) Approximately 3 tonnes can easily be blended with the high purity feeds to meet the plutonium immobilization specifications and another tonne or so can be processed in the immobilization plutonium conversion area to yield materials that can be blended to provide acceptable feed for immobilization.
3) The remaining 3 tonne must be excluded in their present form.

However, approximately 2 tonnes of the excluded materials could be processed in existing DOE facilities to make them acceptable to the immobilization process. Examples of residues that could easily be process through Savannah River prior to shutting down the 221-F Canyon Facilities include materials such as:

- Sand, slag, and crucible residues;
- Scrub alloy; and
- Plutonium fluoride residues.

This leaves approximately a tonne of plutonium as low assay materials that probably should be declared waste and shipped to WIPP.

However, if the MOX option is chosen, approximately 33 tonnes of the high purity materials will be transferred to the MOX option. These acceptance specifications were written assuming that the 33 tonnes would go to MOX and not immobilization.

EXCESS MATERIALS

There have been a number of publications that have provided estimates of the quantities of excess nuclear material. The latest estimates are given in Table 1. Associating these quantities with a specific cans of material is, however, more difficult to do. The assumption of the immobilization project is that the final set of feeds will be specified as follows:

A. The plutonium isotopics vary from about 3% ^{240}Pu to approximately 40% ^{240}Pu.
B. The plutonium assay in the candidate materials varies from ~10% to >99 wt%.
C. The last date of purification of these materials varies from the early 1960's to the late 1990's, therefore the ^{241}Am content varies from as little as 200 ppm to as much as 15 wt%.
D. The uranium content varies from trace depleted uranium in the plutonium to trace plutonium in fully enriched uranium.
E. The impurities in the existing feed stocks are dominated by the following elements: aluminum, carbon, calcium, chlorine, chromium, iron, fluorine, gallium, potassium, magnesium, molybdenum, sodium, silicon, tantalum, and zinc.

Table 1. Composition of United States Surplus Plutonium by Form and Grade

Form	Weapon-Grade[1]	Fuel-Grade[2]	Total
Metal[3]	27.8	1.0	28.8
Oxide[4]	3.1	1.2	4.3
Reactor Fuel[5]	0.2	4.2	4.4
Irradiated Fuel[6]	0.6	6.1	6.7
Other Forms[7]	6.4	0.7	7.1
Totals	38.2	13.2	51.3

1) Weapon-grade plutonium contains less than 7% ^{240}Pu.
2) Fuel-grade plutonium contains from 7% to <19% ^{240}Pu.
3) Metal refers to plutonium in weapon components, ingots and buttons.
4) Oxide refers to plutonium oxide powder
5) Reactor fuel refers to fabricated mixed-oxide fuel and metal alloy fuel elements, pellets and mixed-oxide powder.
6) Irradiated fuel refers to mixed-oxide fuel and metal alloy fuel.
7) Other forms refer to enriched uranium/plutonium oxides and residual process materials from the fabrication of weapons components.

BLENDING STRATEGY

The blending strategy is similar to the traditional metal blending strategy used to manufacture nuclear weapons. The plutonium metal blending strategy divided metal into four grades depending upon the level of impurities: war reserve metal and categories I, II, and III:

- The war reserve grade met the impurity specifications for the metal to be used in weapons.
- Category I metal was higher purity material than required to meet war reserve specifications.
- Category II metal had impurities that could easily be blended with Category I to meet the war reserve metal specifications. This allowed a larger fraction of the available metal to proceed to the weapons foundry than would have otherwise been possible.
- Category III generally meant that impurities were too great to allow blending to war reserve specifications without re-purification or very careful planning of the blend mixture. The rule of thumb was "always ask Rocky Flats to consider the blending route first before you considered repurification."

Chemical Breakdown. Based upon the limited chemical data available, the feedstock can be grouped as follows:

- Group I Materials. Materials with purity far exceeding what is required for immobilization: approximately 45 tonnes of the 50 tonne case, or approximately 12 tonnes for the 18 tonne case.
- Group IIa Materials. Materials with impurities that can be blended into acceptable feed stocks for immobilization: approximately 3 tonnes of plutonium.
- Group IIb Materials. Materials with impurities the Immobilization Conversion Facility can accommodate: approximately 1 tonne of plutonium. This includes the "chloride oxides" at Rocky Flats and at Hanford.
- Group IIIa Materials. Materials previously identified by internal DOE studies as requiring processing in the Savannah River Site (SRS) canyon (aqueous dissolution and reprecipitation): approximately 1 tonne of plutonium. These materials include fluoride materials and scrub alloy at Rocky Flats as well as sand, slag, and crucible materials at both Rocky Flats and Hanford. After processing at Savannah River, these materials will move into Group I.
- Group IIIb Materials. Salt residues from molten salt processing. These have been previously identified as needing removal of the chloride salts for stabilization purposes: approximately 1 tonne of plutonium. After removal of about 75% of the 16 tonnes of spent chloride salts, this material would meet the description of Group IIb.
- Group IIIc Materials. There is also a group of materials that, if calcined to remove carbon, could come to the plutonium immobilization program. These materials are under study to determine just how much can be absorbed within the immobilized product. Some of these materials have plutonium contents as low as 5 to 10 wt%.

Blending. There are a number of ways to be creative and blend plutonium-bearing materials on a large scale. The method we conceive of blending the feeds is by using a distributor. The distributor looks something like an octopus. It has a single feed hopper that will hold the contents of only one plutonium storage can (< 4.5 kg of plutonium oxide powder). Distributed around but below the hopper a number of chutes or legs. Under each chute there is a receiving can. Each of the chutes and its related receiving cans are nuclearly isolated from the others. The way the blender works is that as one can of plutonium powder is added, it splits the contents between each of the chutes. When the first can has been divided amongst the chutes, a second feed can is added. This is continued until the total amount of plutonium has been added to the receiving cans. At the end of the process, each of the receiving cans contains a layer from each of the input cans. Each of the receivers will then be homogenized to give the final blend. Assuming that the distributor distributes the same amount of powder to each of the chutes and hence the receivers, each of the receivers will contain the same blend of materials.

One of the homogenized receivers will be characterized to determine if that particular feed batch meets the specifications to be feed downstream to the ceramic fabrication line. If the feed batch indeed meets the specifications, then it will be transferred downstream to the ceramic fabrication line. If the feed batch does not meet specifications, then the batch will be returned to the vault for re-blending. The blend schedule will be set-up with the best available data.

The baseline for plutonium immobilization is to blend on the 40- to 60-kg plutonium scale. One approach is a sample splitter that partitions the oxides into 10 to 12 receiver containers. As each receiving container can be individually shielded to produce an isolated environment with respect to criticality, there will never be more than 4.5 kg of plutonium oxide in any one place at any one time. Furthermore, neither water nor other hydrogenous materials will be allowed into this segment of the processing line. Because the material in each of the receivers will be layered, each receiver will be shaken or tumbled to homogenize the feed materials. One of the receivers of blend stock will be sampled and analyzed. This will be the first complete characterization of the plutonium feed prior to the ceramic fabrication steps. The scheduling of feed batches for blending will be done using a computer model with the best available data. However, full chemical characterization of most of the plutonium materials was never made. This material was stored for re-cycle through a purification line and only the data required to determine which line it should be processed through was maintained. To hold down the cost of characterization, process knowledge will be used to the maximum extent possible to determine the blend schedule.

In principal, an upper limit of the number of blending operations to achieve perfectly homogeneous feed to the immobilization plant can be estimated. For example, if approximately 10,000 cans of plutonium feed material were to be fed to the immobilization plant and if these were blended 10 at a time, 1000 batches would be generated. Each of these batches would be split into 10 cans. Each of these cans would contain 10% of the material that was in each of the 10 cans that went into the batch. Thus, the original material would have been diluted by a factor of ten. If ten lots of these 10 cans were blended in like fashion, each of these cans would contain 1% of the original material that was in each of the 100 cans that went into that batch. If this blending and splitting procedure were repeated 4 times, then each of the scan in the final stage would contain $1/10{,}000^{th}$ of the material in each of the original cans, and would be essentially identical to all other cans generated in the final stage. In all, 4,000 blending operations (batches) would be required, and storage for 10,000 cans would be needed. This would require 80,000 can movements between the vault and the blending station.

Whereas the second blend could begin after more than 10% of the material had been processed through conversion, the fourth and final blend could not begin until after greater than 91% of the material had been processed through conversion. Total blending would therefore take about 12 to 15 years to complete. This is not within the spirit of an acceptable answer to a "clear and present danger." Acceptable feed can be generated by much less blending.

If an acceptable feed material could be generated with one stage of blending, then three fourths of the operational costs and most of the time delay associated with the ideal case described above could be saved and much less in-line storage capacity would be needed. However, the available feed stream impurity data indicates that the average impurity level of many of the feed streams do not meet the requirements of the ceramic feed specifications. Thus, different feed streams must be mixed into each batch. In addition, due to can-to-can variations within a feed stream, an unacceptable batch might be generated from a feed stream that, on average, has acceptable properties but the particular can used in the batch was outside the acceptance specifications. There is a need to mix cans from different feed streams to dilute high levels of impurities in different feed streams and in different cans within a feed stream.

Simulation Model. A discrete-event simulation model was developed to evaluate different blending strategies. After each batch is blended, the batch is characterized. Batches that do not meet requirements must be returned to storage and reblended with other feeds to reduce the impurity content to acceptable levels.

Some general assumptions that were used in developing the simulation model were:

- All materials are available for blending on demand—the blending problem is decoupled from the storage problem.
- The blending strategy will be aggressive.
- A can of feed material cannot be used in more than one batch.
- The statistical properties (mean and standard deviation) of each feed stream are known, but the properties of individual cans are not well known.
- The total amount of plutonium in each stream is known.

The simulation model picks one or more cans from various feed streams and sends them to the blending operation. Cans are fed to the blending portion of the model where the amounts of plutonium and contaminants are accumulated in the blender. The cans are combined until the blender limit would be exceeded by the addition of the next can in the queue. At this point, the properties of the material are recorded, and the blender is emptied.

The discrete event simulation model was built using a commercially available simulation modeling system (Extend®). It includes approximately 1500 nodes (e.g., queues and splitter).

RESULTS-DISCUSSION

The model was exercised assuming different amount of plutonium in the blend batch. The number of reblends necessary dropped sharply until a batch size of approximately 48-kg of plutonium was used. Above 48 kg, the curve flattened out with the number of reblends decreasing very slowly. This leads to the conclusion that the blend batch size must be at least 48-kg of plutonium to minimize the amount of in-line storage necessary to storage the cans of plutonium waiting reblending.

Savannah River has also completed a preliminary criticality analysis. This analysis assumes that the receivers are double batched or totally filled with high assay plutonium powders. Therefore, to maintain criticality control, the size of the receivers must limited so as to maintain less than about 10 kg of plutonium when filled with high assay plutonium oxide.

Since the feed stock overall contains approximately as much uranium as it does plutonium, if the receivers are assumed to accept approximately 4 kg of plutonium, they will also be receiving approximately 4 kg of uranium. The amount of tramp impurities that the batch can accept would mean that approximately 2 kg of tramp impurity would also be present in each receiver. Therefore, the receiver must be sized to receive approximately 10 kg of plutonium, uranium, tramp impurity mixture. As all of these are assumed to be oxides, the total mass will be about 12 to 13 kg.

A more detailed criticality study must be completed for the system that is actually designed. At this time, this analysis leads directly to the conclusion that a minimum of 12 receivers are necessary to maintain criticality control and carry out the necessary blending to meet the requirements of the ceramic specifications.

The simulation model currently incorporates relatively simple logic for blending feed streams and does not rely upon knowledge of the impurity content of individual plutonium storage cans prior to blending. So long as the blend batch is kept at 48-kg or larger, it appears as if less than 20% of the blend batches would have to be reblended. Currently, some of the feed streams are very poorly characterized; relying on sparsely documented engineering judgement. Some of the feed streams are so poorly characterized that only the plutonium content is known. As more information become available about the feed stream characteristics, the inputs to the simulation model can be refined.

CONCLUSIONS

Blending and not costly chemical pre-characterization of the individual cans of plutonium in the feed stream will be sufficient to produce a feed that is acceptable for fabrication of the ceramic immobilization pellets. To accomplish this, however, a minimum blend batch size of about 48-kg of plutonium must be used. By being creative, this blend batch size can be carried out within criticality control limits by using twelve chutes and receivers; these chutes and receivers must be nuclearly isolated from each other.

The individual storage cans of plutonium oxide will contain less than 4.5-kg of powder at the end of the 94-1 Stabilization Program. Only those cans that contain >85% plutonium will the plutonium approach 4.0-kg. Each of the receivers, however, will contain about 4-kg of plutonium, about 4-kg of uranium, and up to 2-kg of tramp impurities for a total of about 10 to 12-kg total mass (when oxygen is accounted for); the twelve receivers will contain about 120 to 145-kg of total mass. By characterizing the batch after the blending, less than 5% of the number of samples will be generated than would have been required to characterize the feed cans as part of the 94-1 Stabilization Program.

The batches that fail to meet the specifications will be recycled with much better characterization data available than is currently available for many of the present storage cans of plutonium materials.

The best estimate at present is that the in-line storage vault must be sized to accommodate a recycle of about 20% of the blend batches.

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