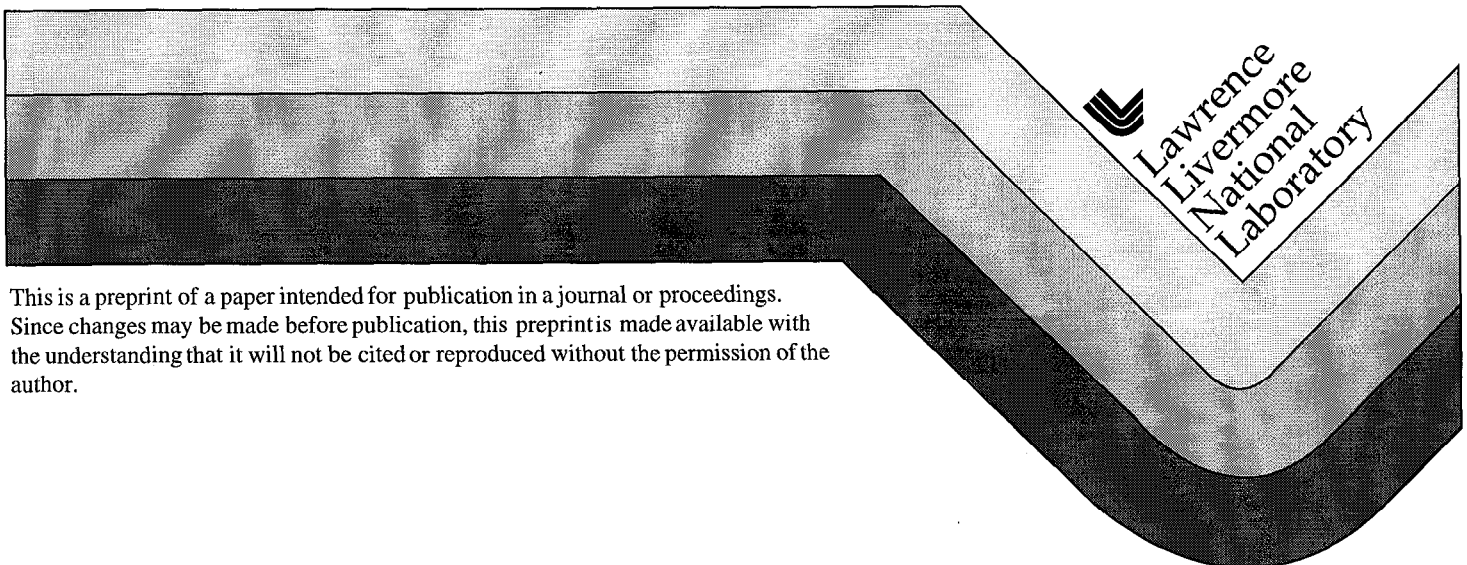


Materials Disposition Plutonium Acceptance Specifications for the Immobilization Project

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**MATERIALS DISPOSITION PLUTONIUM ACCEPTANCE
SPECIFICATIONS FOR THE IMMOBILIZATION PROJECT**

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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. 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."

For disposition by immobilization, the planned facilities will have only limited capabilities to remove impurities prior to blending the plutonium feedstocks to prepare feed for the plutonium immobilization ceramic formation process. Technical specifications are described here that allow potential feedstocks to be categorized as either acceptable for transfer into the MD Immobilization Process, or unacceptable without additional processing prior to transfer to MD. Understanding the requirements should allow cost benefit analyses to be performed to determine if a specific material should be processed sufficiently

to transfer to MD or should be prepared for shipment to WIPP.

Preliminary analyses suggest that about 45 tonnes of this material have impurity concentrations much lower than the immobilization acceptance specifications. In addition, approximately another 3 tonnes can easily be blended with the higher purity feeds to meet the immobilization specifications. Another 1 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. The remaining 3 tonnes must be excluded in their present form. However, approximately 2 tonnes of this remaining material could be processed in existing DOE facilities to make them acceptable to the immobilization process. This leaves about a tonne that probably should be declared waste and shipped to WIPP.

These specifications are written primarily for large lots of material, for example, 100 kg or more of plutonium in the lot. Small lots of material, such as is common for Central Scrap Management Office (CSMO) materials, will have to be handled on a case by case basis.

INTRODUCTION

The Materials Disposition Program (MD) within the Office of Fissile Materials Disposition will be receiving fissile materials packaged by facilities operated for the Offices of Defense Programs (DP), Environmental Management (EM), and

Nuclear Energy (NE) to disposition them in a fashion that meets the "spent fuel standard," i.e., "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." It is assumed that EM will either prepare the plutonium as necessary to meet non-proliferation and transportation requirement concerns for transfer to WIPP, or stabilize the materials as necessary for their transfer to MD. It is assumed that DP and NE will package their excess materials as necessary and transfer them to MD. All dispositioned material will ultimately reside in a geologic repository.

An objective of the immobilization project is to accommodate the range of plutonium feed stocks, with the least possible cost for feed characterization, the least possible handling of the material, and the least possible disruption to the EM (94-1) Stabilization Program. It is imperative that a cooperative program between MD and EM be laid out such that no orphan materials remain within the DOE Complex at the end of the disposition program.

The head-end of the plutonium immobilization facilities will have the capability:

- to convert metals to oxide,
- to dejacket unirradiated fuel elements,
- to grind materials,
- to calcine materials, and
- to leach soluble salts from materials.

This capacity can be increased if it is determined by MD and EM that increasing the capacity to partially process incoming feed stock within the conversion facility is preferable to partial processing within existing DOE facilities.

The first stage of immobilization will convert plutonium oxide with its accompanying uranium oxide and impurity oxides into a multiphase crystalline ceramic. For planning, two cases have been studied: a 50 tonnes case in which all of the excess plutonium is immobilized, and a hybrid case, in which approximately 32 tonnes of plutonium are dispositioned as MOX fuel and about 18 tonnes are immobilized. The specifications for materials going to first stage immobilization have been set assuming the hybrid case. This is the most conservative case because the high purity of the 32 tonnes intended for MOX would greatly aid blending down the impurities within

the 18 tonnes of impure plutonium. There will be about 17 tonnes of depleted uranium, about 600 kg of full enriched uranium and can be up to about 8 to 10 tonnes of impurities.

The baseline second stage of immobilization will provide the radiation deterrent portion of the "spent fuel standard" by encapsulation of the cans of immobilized plutonium within high level waste glass containing ¹³⁷Cs.

The composition, forms, and storage packages of plutonium materials throughout the complex are not well defined. The majority of the separated plutonium that is not in pit form is housed in the plants -- Rocky Flats, Hanford, and Savannah River -- in conditions that are not acceptable for long term storage. These materials will require repackaging and some will require stabilization or minimal processing to allow safe storage until disposition is complete. Until 1994, complex-wide directives that applied to plutonium storage, including safety and safeguards orders, were general in nature. Therefore, current plutonium storage practices vary among sites. These directives do not provide much detail on how plutonium should be prepared for storage, in what sort of containers it should be placed, and how the containers should be monitored.

To support either long-term storage or disposition of these excess materials, data packages must be developed for the repackaged materials. Source and processing information that provide insight into the impurities and forms of the plutonium materials must be included to facilitate future disposition of these materials. (Some of this information is currently available at the facilities, but could be lost during facility decommissioning activities.) Common data formats are proposed to enhance interpretation by future materials handlers, simplify inter-site transfers of materials, and decrease the possibility of inter-site differences due to communication errors.

ASSUMPTIONS

1. Separated plutonium that is declared excess will either be disposed of in accordance with the "Spent Fuel Standard" at the Federal Repository or will be diluted below the Safeguards Termination Limit and disposed at WIPP. DOE irradiated fuels will be disposi-

- tioned at the Federal Repository by EM as part of the spent fuel program.
2. The MD Immobilization Facility will begin operations in the 2005 - 2006 timeframe. It is assumed that this facility will be at the Savannah River Site.
 3. The head-end of the plutonium immobilization facilities will have the capability:
 - to convert metals and alloys to oxide through the hydride-oxidation technique,
 - to deject unirradiator fuel elements,
 - to grind materials to proper sizes,
 - to calcine materials, and
 - to leach soluble salts from specific materials (a limited capacity).
 4. Blending will be used to minimize other processing and characterization costs and improve product quality and reproducibility of the immobilized form.
 5. The first stage of immobilization will convert plutonium oxide (along with its accompanying uranium oxide and tramp impurity oxides) to a monolithic, multiphase, crystalline ceramic, predominantly the mineral pyrochlore. The product form will also contain varying amounts of zirconolite, brannerite, rutile and potentially other phases. The ceramic immobilized plutonium will be sealed into cans.
 6. The second stage of immobilization will encapsulate canned ceramic forms in HLW glass containing ^{137}Cs at the DWPF. The HLW glass will provide the radiation barrier required to meet the "spent fuel standard".
 7. The Immobilization Facility will operate as an unclassified facility with either IAEA or other international inspections anticipated. Therefore, no classified materials will be accepted into the immobilization facility.
 8. All plutonium declared excess (materials not part of the strategic reserve or scheduled to be transferred to WIPP) will be available to the immobilization facility on demand.

BLENDING

The isotopic composition of the 50 tonnes of excess plutonium feed stock vary from 3% ^{240}Pu to ~40% ^{240}Pu . The plutonium assay in the candidate materials varies from <10 wt% to >99 wt%. The last date of purification of these materials varies from the early 1960s to the late 1990s; therefore the ^{241}Am content varies from as little as 200 ppm to as much as 15 wt%. The

uranium content varies from trace depleted uranium in the plutonium to trace plutonium in fully enriched (93% ^{235}U) uranium. In general, the impurities in the existing feed stock are dominated by the following elements: Al, C, Ca, Cl, Cr, Fe, F, Ga, K, Mg, Mo, Na, Si, Ta, U, W, and Zn. It is obvious that large scale blending will be necessary to obtain a reasonably consistent plutonium feed stream.

The blending strategy is similar to the old metal blending strategy used to manufacture nuclear weapons. Metal was divided into four grades depending upon the level of impurities -- categories I, II, and III, and War Reserve (WR) metal. WR grade met the impurity specifications for the metal to be used in weapons. Category I metal was higher purity material than required to meet WR specifications. Category II metal had impurities that could easily be blended with Category I to give WR metal. 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 WR specifications without careful planning of the blend mixture. The rule of thumb was "always ask Rocky to consider the blending route first before you considered re-purification."

Chemical data for the excess feedstock vary in completeness but approximations are as follows:

- Group I materials: Materials with purity far exceeding what is required for immobilization: ~45 tonnes of the 50 tonnes case. (Approximately 13 tonnes for the 18 tonne case)
- Group IIa materials: Materials with impurities that can be blended into acceptable feed stocks for immobilization: ~ 3 tonnes.
- Group II-b: Materials with impurities the Immobilization Conversion Facility can accommodate: ~ 1 tonnes. (This includes the "chloride oxides" at Rocky Flats and at Hanford.)
- Group III-a: Materials previously identified by internal DOE studies as requiring processing in the SRS canyon (aqueous dissolution and re-precipitation): ~1 tonne. These materials includes 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 Sa-

vannah River, these Iii materials will move into Group I.)

- Group IIIb. Salt residues from molten salt processing. These have been previously identified as needing removal of the chloride salts for stabilization purposes: ~1 tonne. (After removal of about 75% of the 16 tonnes of spent chloride salts, this material would meet the description of Group IIa.)
- Group IIIc. There is also a group of materials that, if calcined to remove carbon, could come to the 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%.

There are a number of ways to be creative and blend plutonium-bearing materials on a large scale. The baseline for immobilization is to blend on the 40 to 50 kg Pu scale. One approach is with a sample splitter which partitions the oxides into 10 to 12 receiver container. As each receiver container can be nuclearly isolated, there will never be more than 4.5 kg of plutonium in any one place at any one time. Furthermore, neither water nor other hydrogeneous materials will be allowed into this segment of the processing line. Because the material in each of the receivers will be layered, each receivers 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.

After blending, if the batch meets specifications for processing, it will be fed to the ceramic fabrication line.

If the batch is only slightly out-of-spec (within perhaps 10%), then additional uranium and ceramic precursor may be added (the uranium plus plutonium will be kept constant to assure the right mineral assemblage) to the batch fed downstream to the ceramic fabrication line. The nominal concentration goal is for a plutonium content of 10.5 wt% and a uranium content of 21 wt% of the ceramic weight. Typically, the plutonium plus uranium content will be kept constant at

31.5 wt%, but the plutonium will be allowed to vary slightly to accommodate outlying impurity concentrations.

If the batch is substantially out-of-spec, the entire 40 to 50 kg of plutonium feed will be returned to the working vault and re-blended with new material. This time the blending will be done using the more accurate data provided by the characterization of the 40 to 50 kg blend batch.. Computer modeling suggests that about 20% of the feed batches will have to be blended more than once (for the 18 tonnes case) because of the lack of good elemental data for some of the incoming feed material.

The amount of enriched uranium that will come to the Immobilization Program is uncertain. There is at least 600 kg of fully enriched uranium that is contaminated with plutonium. The easiest way to handle this enriched uranium is to assume that it is equivalent to plutonium and blend accordingly. If all of the enriched uranium came into the Immobilization Program, treating it as if it were 600 kg of plutonium would increase the number of canister filled with fissionable material by 20. Since only 10% of the glass is displaced from the canister by the addition of the ceramic immobilized plutonium (enriched uranium), the number of additional canisters sent to the repository would be only 2.

The estimate of the number of re-blends will be dependent upon the quality of the data available going into the blend operation. The better the data that are available, the more accurate will be the blend. We fully realize that average data for large lots of material are much more reliable than for individual containers of material. We also fully realize that there will be surprises -- these will be handled through the limited capacity aqueous leaching/processing line or by re-blending.

POST BLENDING SPECIFICATIONS

Just as in nature, the minerals formed during the fabrication of the ceramic immobilization product depend upon the ratio of the individual elements that are present in the feed materials. Also, just as in nature, the minerals can accommodate a fairly wide range of elements so long as substitution is possible. These substitution rules are

based upon atomic charge and ion size. The substitution rules, which represent specifications for post-blended feed to ceramic fabrication, are given in Table I.

The hybrid (18 tonne) case contains about 75,000 gram-atoms of plutonium. The best data to date suggest that the ceramic product can absorb at least 1.75 moles of impurity per gram-atom of plutonium; this is equivalent to about 130,000 moles of impurities. If one assumes a weighted average molecular weight of the impurities of about 60 to 80, then the ceramic formulation has the capacity to absorb about 8 to 10 tonnes of impurities, depending upon what the impurities are.

It should be noted that in the specifications (Table 1), the term volatiles refers to compounds that vaporize at 1350°C, not at 800 to 950°C as normally thought of for Loss on Ignition (LOI) analyses.

SOURCES OF PLUTONIUM

The composition, forms, and storage packages of plutonium materials throughout the complex vary considerably. The materials come from a wide variety of sources: direct conversion of plutonium pits; unirradiated plutonium/uranium fuels; and a variety of metallic and chemical compounds and forms remaining from weapons component recycle processes. The DOE shut down most plutonium facilities in the late 1980s. All plutonium operations at Rocky Flats were stopped in 1989. The Plutonium Finishing Plant at Hanford and the F Area facilities at Savannah River suspended operations about the same time. These facilities are not expected to resume operations except on a limited basis for material stabilization and facility decommissioning. (It is assumed that Savannah River and Los Alamos will stabilize materials not only from their own site but some materials from other sites as well.) The Los Alamos National Laboratory (LANL) plutonium facilities have continued to operate; pit recycling and production will be conducted at LANL. Lawrence Livermore National Laboratory (LLNL) has a small processing facility that supports research and development. The facilities at LLNL could be used for partial purification of spent chloride salt residues if EM and MD determine this was more desirable

than placing spent chloride salts into WIPP after waving the Safeguards Termination Limits.

The majority of the separated plutonium that is not in pit form is presently housed at the plants that were shut-down in the late 1980's-- Rocky Flats, Hanford, and Savannah River. The general consensus of the Defense Nuclear Facilities Safety Board (DNFSB) is that some of this material is stored under conditions that are not acceptable for long term storage.

DISCUSSION OF SPECIFICATIONS

For planning purposes, the 50 tonnes of surplus plutonium to be used as disposition feed material can be separated into eight categories, which represent the physical and chemical nature of the plutonium. Two of the categories — clean metal (including pits) and clean oxide — could either be fabricated into MOX fuel, if the hybrid approach is selected, or immobilized. The remaining six categories of material — impure metals, plutonium alloys, impure oxide, uranium/plutonium oxides, alloy reactor fuel, and oxide reactor fuel — would be immobilized.

Known Acceptable materials

Based upon the present state of knowledge of the plutonium residues, and the ceramic immobilization form impurity experiments so far completed and analyzed, some materials appear to be sufficiently characterized by process history that they can be blended into acceptable immobilization feed. These include:

1. **Clean Metal.** Pure Pu metal generally with < 100 ppm of any given chemical impurity. The Pu can be weapons-grade, fuel-grade, or reactor grade. The only major chemical impurities are Ga and radioactive decay products such as Am, Np, and U. Examples of pure metal items include unalloyed "buttons" of Pu metal, billets, ingots, castings or rough machined items, finished machine weapons components, and other miscellaneous metal pieces and parts. If classified, these must be declassified prior to being received into the immobilization facil-

- ity. This totals about 31.8 tonnes of plutonium.
2. **Clean Oxide.** Plutonium oxides (Pu $\geq 85\%$) with less than 3 wt% impurities. This totals about 1.7 tonnes of plutonium, all of it nominally 6% ^{240}Pu .
 3. **Alloy Reactor Fuel and Oxide Reactor Fuel.** Plutonium-containing reactor fuel that has been manufactured, but not irradiated in a reactor. The fuel can be either alloy reactor fuel or reactor fuel containing plutonium oxide mixed with uranium oxide. The majority of alloy fuel is fuel elements for the Zero Power Plutonium (now referred to as Physics) Reactor (ZPPR) fuel elements which consists of both plates and rods (irradiated to about 50 watt-days/MT). There is a total of more than 40,000 items containing about 3.8 tonnes of plutonium. The plates (totaling about 3.5 tonnes of Pu alloy) typically contain plutonium isotopics of 4.5%, 8.6%, 11.5%, 22%, or 27% ^{240}Pu . The majority of mixed oxide fuel is fuel elements, and partially fabricated fuel (rods, pellets, reject pellets, and loose blended powders) from either ZPPR or the Fast Flux Test Facility (FFTF). The ZPPR rods (totaling about 0.3 tonnes of mixed Pu-U oxides) typically contain plutonium isotopics of 11.6% or 26% ^{240}Pu . This FFTF mixed oxide totals about 1 tonne of plutonium. The primary plutonium isotopic is about 11.5% ^{240}Pu .
 4. **Enriched Uranium/Plutonium Oxide.** Plutonium oxides mixed with enriched uranium oxides. This category varies from trace uranium in the plutonium to trace plutonium in the uranium. This category contains about 0.9 tonne of plutonium and about 0.6 tonne of enriched uranium.
NOTE: If there is no plutonium in the uranium the material should go the enriched uranium disposition program, not the plutonium disposition program.
 5. **Impure Metal.** Impure plutonium metal of uncertain composition in which the primary impurities are < 50 wt% of the item weight. The impure metal category contains about 3.4 tonnes of plutonium. A portion of this can be fed to the blending step after conversion to oxide (see below).
 6. **Plutonium Alloys.** Plutonium-containing alloys with impurities that are less than 50 wt%.

7. **Impure Oxide.** Plutonium oxides with >3 to 50 % by weight of impurities. The impure plutonium oxide category contains about 6.4 tonnes of material. A portion of this can be fed directly to the blending step. The so called "chloride oxides," (containing ~ 700 kg of plutonium and ~ 300 kg of NaCl/KCl), which are part of this category, can be processed in the conversion head-end of immobilization to yield an acceptable feed.

The first four items above total more than 38 tonnes of plutonium (about 6 tonnes for the hybrid case). All of this material meets or exceeds the purity level needed for immobilization. The next three items total about 10.7 tonnes, of which about 70% is at or above the purity level needed for immobilization. Therefore, about 45 tonnes of the plutonium feedstock can be accepted directly into the immobilization program without regard to impurity blending. The primary reason to blend this 45 tonnes of material is to levelize the uranium composition and plutonium isotopics across all of the material.

The remaining 30% of the above material (~ 3.2 tonnes) can either be blended directly with the cleaner materials, or can be sufficiently purified by planned conversion facility, which has the capabilities to make them acceptable for blending.

Excluded Materials

The Immobilization front-end facility is not being designed to handle the following 3 tonnes of plutonium materials. These materials must be either pre-processed in other facilities to yield a product acceptable for transfer to MD Immobilization or prepared for shipment to WIPP.

1. **Dissimilar Materials.** Unlike materials from different points of origin (glovebox lines, MBAs, IDCs, facilities, ANSI codes, etc.). Dissimilar materials from different origins shall not be mixed together. [Alternatively these materials could be fully (chemically and physically) re-characterized prior to transfer to MD. It should be noted that there is no budget to support such characterization; in effect, this mixing would generate orphan materials.] Mixing of some very similar materials could be toler-

ated without causing additional cost and exposure. Materials in the clean oxide category, i.e. >85 wt% Pu, could be mixed without risk. Mixing of materials from the same glove box (IDC, etc.) which have approximately the same plutonium and impurity content could be combined without risk. Blending of floor sweeping from two different gloveboxes, e.g., the oxide cascade dissolver glove box and the hydrofluorinator glovebox, that are not like materials, would add risk

Reason: : Indiscriminate mixing results in the loss of process knowledge, which is very important in determining the probable range of expected impurities. Blending at the site level of dissimilar materials could result in additional analysis, additional re-blends, require larger working vault space, etc., at the Immobilization Site. This loss of knowledge results in additional cost and additional exposure to personnel. The mixing of a few cans at the site level might save a few thousand dollars but result in having to increase the number of canisters going to the federal repository. As each canister will cost approximately \$500,000 to place into the Federal Repository, the few thousands of dollars saved at the site could result in an increased cost to DOE of half a million dollars or more.

2. Classified materials.

Reason: The MD Immobilization facility is expected to be under either IAEA or other bilateral international inspection.

MD is constructing a pit conversion facility to deal with classified materials and convert them into unclassified materials. There are also two National Laboratories which can deal with classified materials. Savannah River will also have some capacity to declassify materials. A combination of the four sites should be able to handle all declassification matters.

3. Other Actinides. Plutonium materials with any of the following radioisotopic contents:

- with ^{238}Pu content greater than 5 atomic percent ^{238}Pu ,

- with ^{241}Am content greater than 15 wt% ^{241}Am ,
- with ^{237}Np content greater than 5 wt% ^{237}Np ,
- with ^{233}U content greater than 0.5 wt%.

However, higher isotopic materials can be considered for acceptance on a case by case bases. Batch sizes of materials determined to be acceptable materials may have to be limited to handle the higher radiation dose.

Reasons: The immobilization program can accept much higher contents of ^{238}Pu , ^{241}Am , and ^{237}Np than normally expected because of the way the materials will be handled. There are reasonably small amounts of these materials, therefore they can be handled on an intermittent basis. Most of these materials are oxides and can be taken directly to the plutonium blending station. Downstream of the plutonium blender, they will be diluted by at least a factor of ten; downstream of the ceramic precursor blender they will be diluted by another factor of at least 10. The additional self-shielding will reduce the dose to the operators even further.

Reasons: The Immobilization Plant will be automated for remote operations, to the extent practical but with contact maintenance.

Reason: The present concept of the vaults provide no provisions for water cooling.

4. Fluoride Materials. Un-reacted PuF_3 or PuF_4 , failed runs, misfires, or floor-sweepings from the glove-boxes between fluoride precipitation or fluorination and bomb reduction.

Reason: The ceramic immobilization form has a limited capacity to absorb the fluoride ion. Processing the material known to be very high in fluoride through the SRS canyons, as proposed, reserves the limited fluoride capacity for those items with lower fluoride contamination.

5. Molten salt solvent residues:

- Alkali and alkaline earth halide salts used as solvents for pyrochemical processing: Direct Oxide Reduction (DOR), Molten Salt Extraction (MSE), and Electrorefining (ER),

- Calcium fluoride solvent salt from bomb reduction, usually called sand, slag, and crucible (SS&C) residues.

Reasons: Volatiles include any compounds that vaporize from the ceramic during sintering at 1350°C, not at 800 to 950°C as normally thought of for Loss on Ignition (LOI) analyses. The volatiles create an internal back pressure in the pores during sintering that can lead to poor densification. In addition to limiting the extent of reaction, which may result in the wrong product phases, the high porosity may be interconnected, thus allowing penetration of water and a possible significant increase in the dissolution rate in the repository.

The Immobilization facility will have a limited capacity to remove soluble salts from the incoming feed stocks. The amount of volatiles that can be absorbed by the ceramic is about 45,000 moles. The chloride salts contain about 200,000 moles of volatile material. The fluoride materials contain about another 1000 moles of volatiles. The chloride oxides contain another 4,500 moles of volatiles. Other materials such as sand, slag, and crucible residues also contain considerable volatile materials. As a rough estimate, the volatile material in the major groups of materials is about 5 to 6 times what the ceramic can absorb. Processing of these materials, in some fashion other than blending, will be necessary.

6. **Added Stable Elements.** Plutonium materials with excessive amounts of stable elements added during the stabilization processing.

Reason: As stated above, the ceramic hybrid case has the capacity to absorb about 133,000 moles of impurities. Stable elements added to the stabilization process count against that 133,000 moles along with the impurities already in the plutonium feed stocks. When decisions are made as to the stabilization process to use, those that result in less impurities added to the plutonium should be chosen over those that add more impurities. For example, one method of stabilizing pyrochemical salts that has been studied is oxidation by either MoO_3 , Sb_2O_3

or V_2O_5 followed by salt distillation. However, the oxidants, with the exception of Sb_2O_3 , are nonvolatile under the conditions of the distillation process and remain in the plutonium heel. Although only one sample result is available, the Pu content was 45.7 wt% and the Mo content was 36.5 wt%. This is equivalent to a Mo to Pu mole ratio of about 2. If all of the salts were treated by this method, about 5% of the plutonium coming to immobilization would be from the salts but the salts would bring about 8 to 10% of the impurities that can be absorbed by the ceramic product. Clearly, a stabilization reagent that leaves behind fewer moles of non-volatile oxides would be preferable from an immobilization standpoint.

Vanadium is particularly troublesome in that excessive vanadium lowers the melting point of the ceramic below 1350°C, which is the operating temperature of the sintering furnace.

Calcium carbonate is another compound that has been suggested to pyro-oxidize the salts. Calcium is a major constituent of the desirable mineral pyrochlore. However, excess calcium will stabilize unwanted perovskite and insufficient calcium will stabilize brannerite. In-between, calcium will react with the brannerite to form pyrochlore. Having brannerite in the formulation is acceptable, however, we do not want excessive amounts of brannerite in the final product. Obviously, calcium is one element that has to be closely controlled. With the amounts of calcium already in the plutonium residues, the ability of the ceramic formulation to absorb calcium added in the stabilization processes is limited. How to handle the calcium problem is under study and the actual amount of excess calcium that can be accepted will be determined by this study.

In short, it is better to try another method than to generate an orphan material.

7. Spent fuels that have been irradiated to more than a few hundred watt-days per metric ton

Reasons: The Immobilization Plant is being built as a contact handling facility. There is an existing path for disposing of

spent fuel. Irradiated materials should be handled along with other categories of spent fuel or the plutonium must be sufficiently purified to come into the immobilization program.

8. **Some Plutonium Alloys.** Plutonium alloys in which the non-actinide content is greater than 27 atomic percent, i.e., scrub alloy from MSE salt residue scrubbing with aluminum.

Reasons: The Immobilization Plant, as planned, does not have the capability to convert higher concentration alloys to oxide.

9. **RCRA-listed materials.**

Reason: The ROD for the Storage and Disposition of Weapons-usable Fissile Materials Final PEIS states: No RCRA wastes would be immobilized unless the immobilization would constitute adequate treatment under RCRA. The immobilization product must be consistent with the repository's waste acceptance criteria.

Documentation

Data Base. An electronic data base shall be maintained to serve as a source of relevant information about stored materials and packages. This data base may consist of several files (which, in themselves, may be data bases), some of which may be classified. For completeness, MC&A documentation shall be coordinated with the data base.

Data base content requirements. The data base shall include, as a minimum, identification of the following material characteristics:

1. Chemical and physical form
 2. Best available elemental and isotopic composition including all actinides.
 3. Quantity (mass) of material.
 4. Stabilization test results and conditions of tests.
 5. Source of stored material (e.g., site, facility and MBA that generated the material, and IDC), if available.
 6. Last known date when Americium content was effectively zero.
7. Specific processing conditions to include processing date, process duration, processing equipment, stabilization temperature, and stabilization atmosphere.
 8. Other information relative to the contents such as expected major impurities with source of impurities data (e.g., process knowledge, destructive analysis, and X-ray analysis)
 9. The data base shall include, as a minimum, identification of the following package characteristics:
 - Nominal fill gas composition of each container on sealing (e.g., air, He, or Ar)
 - Leak test data record for each container
 - Package configuration - quantity and type of containers in package.
 - Date of packaging for each container.
 - Initial radiation field [gamma and neutron at contact and 300 mm (12 in.)] including how measured and correction factor if available.
 - Baseline package gross weight, dimensions, and tare weight.
 - The unique identification number and TID number, if any, associated with each container.
 - The manufacturer lot identification number for each container.
 10. The data base shall include, as a minimum, the following records for surveillance and inspections:
 - Surveillance results
 - Records of tests performed
 - Dates of inspections
 - Names of individuals performing inspections
 11. The data base shall include, as a minimum, specific locations of stored materials.

Other Requirements

1. The majority of the plutonium to be immobilized is currently at sites other than the Savannah River Site. Therefore all packages must meet all government shipping regulations.
2. Detailed chemical characterization exists for only a small portion of the material to be immobilized. However, much information is available from process knowledge. For each area of the

process, it is generally known what the primary elemental impurities are or can be. It is therefore imperative that this knowledge be preserved in the form of item description codes of the origin of the material, MBAs of origin of the material, etc.

Exceptions to the Specifications

These specifications are written primarily for large lots of material -- primarily over 100 kg of plutonium involved. There are many small batches of materials, such as the holdings at Oak Ridge, Argonne, and some at Rocky Flats, and with the Central Scrap Management System at Hanford, Los Alamos, and at Savannah River. With the scrap Management materials, frequently, the holding knows very little about the material and the personnel at the shipping site who shipped the materials no longer works there. There will also be cans of material opened during the stabilization process that just are not what the label says that it is. These materials should be labeled as suspect and the known information transferred to the Immobilization Program. A joint decision must then be reached between the holder of the material and the Immobilization Program as to whether or not the Immobilization Program will accept the material or the holder will process it in some fashion, or the material will be prepared for disposal at WIPP.

If it is accepted into the Immobilization Program the material being labeled as suspect will flag the operators to send the material first to the characterization staging area instead of the blending staging area.

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TABLE I. Preliminary specification limits on blended feed Impurities

	Category	Moles per mole PuO ₂	Impurities
1	volatiles	0.60	NaCl, KCl, CaCl ₂ , CaF ₂ , MgF ₂ , Carbon etc. Plus CuO _{0.5} , KO _{0.5} , NaO _{0.5} , HgO _{0.5} , ZnO, etc.
2	Zirconolite Stabilizers	0.75	AlO _{1.5} , FeO _{1.5} , GaO _{1.5} , CrO _{1.5} , MgO, ZrO ₂ , HfO ₂ , VO _x , etc.
3	Pyrochlore Stabilizers	0.40	WO ₂ , MoO ₂ , TaO _{2.5} , NbO _{2.5}
4	Glass Formers (Si > B)	0.30	SiO ₂ , BO _{1.5}
5	PO _{2.5}	0.10	PO _{2.5}
6	BaO	0.45	BaO
7	NiO	0.1	NiO
8	Total of all impurities included in 1 to 7	1.75	All in 1 to 7
9	"Rare earth" oxides	X + 1.40	LaO _{1.5} , GdO _{1.5} , AmO _{1.5} , etc.
10	CaO	X + 0.25	CaO
11.	"Actinide oxides" excluding UO ₂	1.00	ThO ₂ , NpO ₂ , CeO ₂ , etc. (Not UO ₂)
12	Total of all impurities included in 1 to 11	3.00	All in 1 to 11
13	"Actinide oxides" including UO ₂	2.00	All in 11 plus UO ₂

*CaF₂, MgF₂, etc. will not vaporize at 1350°C, but the fluorine will come off as other fluoride compounds such as SiF₄ and BF₃. Using CaF₂, MgF₂, etc. to represent the compounds actually distilling off is conservative.