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PLUTONIUM STORAGE
AT
MAJOR DEPARTMENT OF ENERGY FACILITIES

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PLUTONIUM STORAGE SAFETY AT MAJOR DEPARTMENT OF ENERGY FACILITIES

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I. OVERVIEW

This report reviews the safety of plutonium stored at the Rocky Flats Plant, the Hanford Site, the Los Alamos National Laboratory, and the Savannah River Site. It considers the inventory of bare (unencapsulated) plutonium metal, plutonium oxides, other plutonium compounds, solid plutonium scrap, and plutonium solutions. The report does not consider irradiated fuel, finished plutonium weapon components (pits), or plutonium-238.

The Department of Energy (DOE) shut down most plutonium facilities in the late 1980s. All plutonium operations at Rocky Flats, previously the largest plutonium processing site, were stopped in 1989. The main plutonium plants at Hanford (the Plutonium Finishing Plant) and the Savannah River Site (the F-Area facilities) suspended operations about the same time. None of those plants is expected to resume operations except as required for decommissioning or other short-term purposes. Only the Los Alamos National Laboratory (LANL) plutonium facilities have continued to operate.

The great majority of the plutonium in the shut-down plants--Rocky Flats, Hanford, and Savannah River--is stored in conditions that are not safe for the long term. Most liquids remain in the same tanks and bottles where they happened to be located when the shutdown orders came. Solid materials were usually removed from processing equipment, but were sometimes packaged haphazardly. Most of the plutonium in the shut-down plants has been declared by DOE to be surplus, but virtually none of it has been readied for permanent disposal or long-term storage. Some is in forms that are difficult to store safely even for short periods.

This report is based on Defense Nuclear Facilities Safety Board (DNFSB) staff visits to the four largest plutonium sites and conversations with local plutonium experts. The staff was assisted by three DNFSB outside experts who are prominent specialists in plutonium chemistry or processing. The general conclusions are:

A. With careful preparation and packaging, plutonium metal and stabilized plutonium oxide can be stored safely over periods as long as a few decades. The draft DOE standard on storage of plutonium metal and oxide is a good guide to established storage practice for metal and oxide.[1]

B. Most plutonium materials other than metal and oxide are not suitable for long-term storage, and there are significant quantities of such materials at all four sites.

C. The high-concentration plutonium solutions and reactive plutonium scrap stored at Rocky Flats pose the most severe and immediate safety risk of any stored plutonium in the DOE Weapons Complex (Complex). DOE is generally aware of the danger at Rocky Flats, but has done little so far to correct it.

D. Much of the plutonium at Hanford and Savannah River is reasonably safe for short-term storage, but DOE is rapidly foreclosing plutonium processing options at those sites. If that trend continues, Hanford and Savannah River may develop some of the same safety problems as Rocky Flats.

II. GENERAL STATUS OF PLUTONIUM STORAGE

This section reviews DOE's four largest inventories of separated, unencapsulated plutonium. "Separated" means plutonium removed from the reactor fuel or targets in which it was created. "Unencapsulated" means plutonium not contained in a finished weapon component. Irradiated fuel and targets have been excluded from this review because, due to their high fission product concentrations and low plutonium concentration, they have properties different from separated plutonium, and are stored and handled differently. Intact weapon components and sealed sources are not considered because their storage is based on an established encapsulation method that protects the plutonium from adverse chemical reactions. Plutonium contained in intact, undamaged components (an important qualification) should not be subject to the types of problems discussed below.

Five sites in the DOE Complex have inventories of separated plutonium in excess of 1 metric ton (1,000 kilograms), although there are smaller inventories at other sites. All of the plutonium at the Pantex site is encapsulated, and thus excluded from this report. The inventories of separated plutonium at the other four large sites have recently been de-classified by DOE:[2]

Rocky Flats Plant	12.9 metric tons
Hanford Site	3.8 metric tons
Los Alamos National Laboratory	2.6 metric tons
Savannah River Site	1.8 metric tons

The following summaries cover the highlights of each of the main sites. Appendix A provides more detail.

A. Rocky Flats Plant

Rocky Flats has more unencapsulated plutonium than all other sites combined. The DNFSB staff is convinced that Rocky Flats is also the one site with serious, immediate problems in dealing safely with its inventory.

Not counting encapsulated weapon components, Rocky Flats has several thousand containers of plutonium metal, compounds, and mixtures. They have a significant quantity of plutonium solution stored in tanks and bottles. They have thousands of containers of scrap of various kinds (called "residue" at Rocky Flats). The great majority of the inventory is either not in forms suitable for long-term storage or is not packaged correctly for long-term storage.

Plutonium operations were suspended at Rocky Flats in late 1989. No plutonium has been processed since then and very little of the inventory has been repackaged. There are three serious problem areas: metals, solid scrap, and solutions.

Plutonium metal corrodes fairly rapidly in the presence of air or water vapor. It corrodes very rapidly in the presence of hydrogen to form pyrophoric plutonium hydride.[3,4] Some metal items stored at Rocky Flats may be in direct contact with plastic bags, an unsafe situation for storage periods extending beyond a few weeks or months. A plastic bag in direct contact with plutonium is subject to rapid radiolysis, which generates hydrogen gas and weakens the bag at the same time. Hydrogen gas will react rapidly with the plutonium metal to generate plutonium hydride. As long as there is free oxygen in the container, hydride will react with it to form oxides. When the free oxygen is depleted, hydride will start to accumulate. The weakened bag is likely to eventually break open and admit air; it is particularly likely to do so when it is handled, but it may do so spontaneously. Either way, there will be an exothermic reaction as the hydride oxidizes, possibly sparking and flaming, and the risk of a larger fire if combustibles are nearby. Other possible adverse chemical evolutions involving plutonium metal and plastic are discussed in Appendix B.

A great deal of plutonium scrap, some containing high concentrations of both plutonium and americium, was also packaged without adequate consideration of long-term chemical and radiological effects. Some scrap was packaged directly in plastic bags or cartons, creating the possibility of high hydrogen generation rates, with the same adverse implications as apply to plutonium metal. (A considerable fraction of the plutonium in the scrap is in the form of metal, so some of the chemical issues are similar.) Many scrap packages are poorly vented. Hydrogen generation in poorly vented containers can create a fire hazard from the hydrogen itself, and can lead to the accumulation of pyrophoric forms of plutonium. Any mishap that results in the sudden introduction of air into such a container (a handling accident, for example, or corrosion of a container wall) could cause a fire or small explosion. The scrap situation is complicated by the fact that the exact chemical composition and packaging arrangements are not known in many cases. There are thousands of scrap containers, mostly 55-gallon and 10-gallon drums, stored in several different buildings. They are generally located in operating areas.

There are approximately 11,000 liters of plutonium solutions stored at Rocky Flats. There are twenty-four tanks of solution in Building 771 alone, with plutonium concentrations as high as 140 grams/liter. There are additional tanks in Building 371, plus several hundred plastic bottles of plutonium solution stored in six different buildings, plus an estimated 9,000 liters of plutonium-contaminated liquid in process piping and tank heels. There have been dozens of leaks from the tanks since the 1989 shutdown. One of the most serious was the July 1993 rupture of an overhead oxalic acid line, which sprayed plutonium-contaminated liquid over a radius of 6-7 meters. Most of the tanks and process piping are in routinely occupied areas, although no one was injured by the July pipe rupture. The solutions are not well characterized. No samples have been taken from any of the tanks since 1989. Some of the tanks are not geometrically safe, and their Raschig rings have not been inspected since 1989. The accumulation of plutonium polymer, the extent of plutonium precipitation, and the severity of tank wall corrosion are all essentially unknown. The DNFSB staff considers DOE's plans for dealing with these problems greatly inadequate.

B. Hanford

Aside from irradiated fuel, almost all of Hanford's plutonium inventory is in the Plutonium Finishing Plant (PFP). There are more than 3,000 containers of plutonium oxides, 370 metal items, somewhat more than one thousand containers of plutonium scrap, and about 250 bottles of plutonium solution. The oxides, metal items, and most of the scrap are packaged in double or triple food-pack cans. PFP has an active monitoring program, with much of the metal and oxide in well-instrumented storage racks that can detect increases in a can's temperature or bulging of a can's bottom. In a typical year, two or three cans are observed to be pressurized or otherwise defective, and their contents are removed and repackaged.

PFP's inventory seems to be fairly safe and stable, with some exceptions. The bottled solutions are old, over 20 years in some cases. There may not actually be any liquid left in the oldest bottles, so it may be difficult to retrieve the plutonium in a form that can be easily stabilized. PFP also has about one thousand polystyrene cubes impregnated with plutonium oxide. They generate gases rapidly and are difficult to store safely; there have already been two contamination incidents involving the cubes. There are also unstable precipitation and solvent extraction residues stored in vented plastic bottles inside glove boxes.

While the present plutonium storage situation at Hanford is not bad, there are some discouraging trends. The problems at Rocky Flats arose largely because plutonium operations were curtailed before accumulations of unstable plutonium materials could be processed into more stable forms. Rocky Flats has found it difficult to restart processes once they have been shut down. The potential exists for similar problems to arise at Hanford should upkeep of PFP be abandoned or processing systems dismantled before existing inventories of plutonium materials can be stabilized.

Until November 1993, DOE had planned to operate PFP's dissolution and solvent extraction facilities in order to eliminate the solutions and the most unstable residues. Now DOE has decided not to operate the PFP's main processes, but to study other ways of stabilizing the materials, such as leaching and thermal stabilization.[5]

This decision has some unfortunate implications. Dissolution and solvent extraction were routinely conducted at Hanford in the past, and are well-known technologies. Developing new processing methods will take time, and no one knows how well they will work. In the meantime, training and maintenance are being reduced to minimum levels, even though some of the plant's processing capabilities may be needed in the future. The option to restart the plant to stabilize materials may be hard to accomplish efficiently and economically.

C. Los Alamos National Laboratory (LANL)

LANL stores a wide variety of plutonium materials, mostly in Technical Area (TA)-55. LANL has around a thousand containers of plutonium oxides and other compounds.

They also have close to a thousand metal plutonium items, most of them high-purity ingots originally intended for shipment to Rocky Flats. Most solid materials are stored in double slip-lid cans. There are typically some plutonium solutions present at any given time as in-process inventory.

The scrap inventory at LANL is modest compared to Rocky Flats or Hanford. There are about 1200 containers of pyrochemical salt scrap, and a few hundred containers of miscellaneous scrap. LANL has active capabilities for processing nearly all forms of plutonium scrap and, with the exception of salts, has generally not allowed a large backlog to accumulate.

DOE has not revealed any plan to shut down the TA-55 facilities, so it should be possible for LANL to continue to process and eliminate unstable compounds and scrap materials. Few of the items in LANL's inventory are currently well packaged for long-term storage since few of them are intended for such. LANL's practices seem to be adequate for the short term, and they are embarking on a general repackaging campaign intended to comply with most provisions of the new DOE standard on long-term storage of plutonium oxides and metals.[6]

D. Savannah River Site (SRS)

SRS has a large quantity of plutonium solution in storage, far more than any other site. There are about 380,000 liters stored in eighteen tanks in F-Canyon and two tanks in H-Canyon (compared to around 20,000 liters at Rocky Flats and no more than 3,000 liters at Hanford). SRS has a significant amount of Pu-238, Pu-242, Am-243, and Cm-244 stored in solution form as well. Solution is not a suitable form for long-term plutonium storage because of the strong potential for leakage and corrosion, and because of the difficulty (due to radiolysis and evaporation) of controlling solution chemistry sufficiently to prevent precipitation or polymerization. Most of the SRS solutions have low plutonium concentrations, but they are still subject to most of those general problems.

Savannah River has a modest amount of plutonium metal in storage, mostly as "buttons" that would have been sent to Rocky Flats. There is a fairly large inventory of plutonium oxide and other compounds, about 1200 containers, plus a few hundred containers of miscellaneous process residues. The residues, metal items, and most of the oxides are packaged in double food-pack cans. There have been some instances of rapid oxidation of metal items, but in general the SRS inventory of solid plutonium materials seems to be well monitored and acceptably stored. SRS is the only site that regularly weighs randomly selected metal items to monitor oxidation, a practice DOE may encourage at other sites.[7]

Until recently, DOE had planned to operate the F-Canyon to remove the plutonium from the stored solutions and process it into a stable form. They decided in January 1994, that the planned clean-out and stabilization run will be delayed until an Environmental Impact

Statement can be prepared.[8] The situation is now similar to the PFP at Hanford. The plutonium in storage may be relatively safe for now, but if F-Canyon's processing capabilities are allowed to atrophy, it may become difficult to eliminate the solutions and other unstable plutonium materials before they develop into serious safety risks.

III. STANDARDS FOR PLUTONIUM STORAGE

Complex-wide safety standards that apply directly to plutonium storage, such as the DOE Orders on safety analysis, unresolved safety questions, and criticality safety, are general in nature. That is, those standards do not provide much detail (criticality is a partial exception) on how plutonium should be prepared for storage, in what sort of containers it should be placed, and how the containers should be monitored. Detailed requirements, where they exist, are generally to be found in site-specific documents.[9, 10, 11] The lack of Complex-wide guidance has had the consequence that plutonium storage practices vary among sites, as discussed in Section II and Appendix A.

A. Long-term Storage Standards

DOE has recently developed a standard for long-term storage of plutonium metal and oxide.[1] It is a DOE consensus standard, based on consultation with experts from all major DOE facilities.[12] The standard is still in draft form as of April 1994; it was issued for comment in mid-April.

The draft standard applies to pure plutonium metal, selected plutonium alloys (such as gallium and aluminum alloys), and oxides that contain at least 50 percent plutonium by weight. It does not apply to sealed weapon components, liquids, scrap, waste, or material containing more than 3 weight percent Pu-238. The goal of the standard is to define handling and packaging practices that would keep the plutonium safe with minimum monitoring for at least 50 years.

It is not clear yet how DOE intends to implement the standard. The normal procedure is for each affected field office to submit an implementation plan to headquarters describing how the field office plans to deal with the standard. The basic uncertainty is what philosophy DOE will adopt about the urgency and timing of bringing the sites into conformance. DOE may decide to systematically process and repackage all unencapsulated plutonium to the terms of the standard on an aggressive schedule. LANL seems to be adopting this approach unilaterally.[6] DOE may take a more gradual approach, imposing the standard only on material that is being processed or repackaged for other reasons, or they may just invoke the standard for material that they specifically want to place in special long-term storage facilities, which would mean that most of the plutonium inventory would not have to be brought into conformance for many years.

The main features of the standard are summarized below:

1. Material form and preparation

Metal must be more than 1 mm thick in all dimensions, or have a specific surface area less than 1 cm^2/gram. Metal must be free of loose oxide. Oxide must be thermally stabilized in an oxidizing atmosphere at 1000°C for at least 1 hour, and must not lose more than 0.5 percent of its weight in a standardized test (called "loss-on ignition"). There is no restriction on the specific surface area of the oxide, nor any stipulation as to the process by which the oxide should be formed.

2. Atmospheres

Following stabilization, oxide must be cooled, handled, and packaged for storage in an atmosphere having a water vapor concentration of 100 ppm or less. Metal must be packaged in a very dry (≤100 ppm water), mildly oxidizing atmosphere.

3. Container design

All material must be packaged in a DOE-approved container made of stainless steel or an equivalent, free of all organics, hermetically sealed, helium leak tested, and free of transferrable contamination on its exterior. Material may be packaged in a container meeting somewhat less strict standards on sealing and leak testing if it is overpacked with a container meeting all the standards. In either case, the inner container will eventually be packaged in another, heavier-duty container, intended to serve as a shipping container, the design of which will be supplied by DOE.

4. Inspection

Containers must be leak tested and weighed after closure. Surveillance of containers in storage must be performed using statistical sampling methods and must include weighing, measuring the outer container, and analyzing the gas inside the outer container.

B. Interim Storage Standards

The terms of the long-term storage standard are sufficiently demanding that all of the sites will need new equipment and procedures to comply. DOE has developed interim storage guidance as a near-term safety measure.[7] The interim guidance is fairly basic. The sites are instructed to come as close as they can to complying with the long-term standard, and there are some common-sense suggestions for handling packages in the meantime. The plan is that the interim guidance will be issued to all DOE sites in April or May 1994. The principal terms of the interim guidance are summarized below:

1. Material form and preparation

Plutonium is not supposed to be stored in the form of solutions, metal turnings, or particles with specific surface area greater than 1 cm^2/gram. Hazardous or pyrophoric corrosion products materials are supposed to be removed from metal items. Oxide from sources other than metal should be stabilized at 1000°C for one hour, if possible. Otherwise a combination of lower temperature and longer time should be used to produce material with the lowest possible loss-on-ignition characteristics (a standard test of the purity of plutonium oxide).

2. Packaging

No plastic is supposed to be in contact with metal or oxide, and use of plastic in outer layers of packaging should be minimized. Metal should be packaged in an atmosphere as dry and inert as possible. Oxide should receive priority over metal for storage in robust vaults due to its higher potential for dispersal in an accident.

3. Inspection

All packages containing more than 0.5 kg of plutonium metal should be weighed annually. Beginning as soon as possible, existing metal packages should be inspected on a random-sampling basis to determine whether significant corrosion has occurred.

C. Conclusions

The long-term standard is a well-documented and technically well-supported consensus standard. The DNFSB staff and outside experts have discussed the standard with many plutonium specialists around the Complex, and have found them to generally support the standard's terms, although there are strong minority opinions on several points, particularly on the issue of sealed containers versus vented containers. Section V of this report will review the basic technical issues in plutonium metal and oxide storage and make a few more observations about sealing versus venting.

There is a problem with the interim guidance. It does not acknowledge the reality that much of the current plutonium inventory is in forms that are unsuitable for even interim storage. The interim guidance simply states that solutions should not be stored, even though three of the four main sites have solutions in storage. It does not mention reactive scrap at all. DOE could provide guidelines on how best to monitor reactive scrap materials until they can be processed and eliminated. DOE could also provide guidelines on how to decide whether to repackage reactive scrap. It is conceivable that developing such short-term storage guidelines might distract effort from the more essential goal of eliminating the solutions and reactive scrap, but it appears at this point that DOE plans to store some of those materials for years. It is necessary to determine how to keep them as safe as possible over that period.

IV. PROCESSING AND HANDLING UNSTABLE PLUTONIUM

The large majority of DOE's unencapsulated plutonium inventory is not configured for safe long-term storage or ready for permanent disposal. Much of the inventory is in the form of scrap or process intermediates, forms that are usually inherently unsuitable for long-term storage or disposal. Most of the rest is in the form of metals and oxides, which can be suitable for long-term storage if they are properly purified and packaged, but most of the metals and oxides in the inventory are not properly stabilized or packaged.[12]

Unstable forms of plutonium are being stored because plutonium operations, including scrap treatment, have been effectively suspended at three of the four sites-- Rocky Flats, Hanford, and Savannah River. At Rocky Flats all plutonium processing was shut down suddenly in 1989, without giving the operators a chance to finish processing solutions and other intermediates. It also happened that the site was holding a particularly large scrap inventory at the time. In the general turmoil that has since prevailed at Rocky Flats, it has proven difficult to make progress in stabilizing the plutonium inventory. It has even been difficult to convince all parties that the plutonium inventory is hazardous in its present storage configuration.

A. Necessary Processing and Handling Capabilities

Almost all unencapsulated plutonium in the Complex's inventory will eventually require processing or repackaging for disposal or long-term storage. For most metal, cleaning and repackaging may suffice. All other forms of plutonium will require stabilization processing to control the risk of container failures and exothermic plutonium reactions if they are stored, or to produce an acceptable waste form if they are to be disposed. Most oxides can be stabilized by heating to a high temperature in air. For other plutonium materials, including solutions and most scrap, it will probably be necessary to extract most of the plutonium in order to make storable or disposable end products, and wastes containing disposable amounts of plutonium.[3,4]

It may on first consideration seem unnecessary to process plutonium scrap when DOE has no need for the plutonium. There are reasons, though, why additional processing will be necessary. Many forms of plutonium scrap are chemically unstable and become more so with time. In most cases there is no accepted way to store or directly dispose of the scrap safely. There are no established standards on the subject because no one has ever viewed the scrap as even potentially suitable for long-term storage. The intention was always that most of the plutonium would be extracted, and the remainder of the material disposed as low-level or transuranic waste. The safety of some scrap materials (particularly at Rocky Flats) can be temporarily improved by repackaging, but there is currently no prospect that repackaging, overpacking, or improved monitoring methods will make the scrap safe for long-term storage. It must eventually be processed for long-term storage or disposal. As a secondary point, it is probably desirable for economical waste disposal to process the scrap into a relatively small volume of plutonium-rich material and a larger volume of low-plutonium waste. There may be additional value in removing or destroying chemically hazardous constituents so that the scrap need not be treated as mixed waste.

1. Characterization

Before any processing is done, feed materials must be adequately characterized. Particularly for older scrap, the composition and condition of the material is often not well enough known to develop a safe, effective stabilization process. It is not clear that all parties at DOE appreciate this. It took great effort by the DNFSB to convince DOE that even minimal feed characterization was warranted for the planned Building 707 thermal stabilization campaign at Rocky Flats.

2. Processing Methods

The principal processing capabilities needed to stabilize the existing inventories are dissolution or leaching, plutonium extraction, and calcination, although treatment of salt scrap will require some other capabilities. Most processes would recover plutonium as an oxide. Most sites (Savannah River may be an exception) will probably not propose to convert the plutonium to metal since producing metal would normally require an additional step after oxide has been produced. Most of the necessary processing equipment is already at hand for use with familiar, long-established technology.

a. Dissolution and leaching

Dissolution or leaching will generally be necessary before plutonium can be extracted from solid materials such as ash, SSC (sand, slag, and crucible), sludges, metal filters, or polystyrene slabs. This has normally been done by grinding (if necessary), then leaching with nitric acid, although other acids are sometimes used. The result is a plutonium solution and a relatively plutonium-free solid residue.

In the past, plutonium-bearing combustibles such as paper and cloth were incinerated at most sites, and the resulting ash was leached to dissolve its plutonium content. Most incinerators have shut down in the face of intense public opposition, and it seems likely that few will resume operations in the foreseeable future. As an alternative to incineration, leaching processes can probably be developed for most types of combustible scrap. Hanford engineers hope to leach their acid-soaked rags (a particularly unstable form of scrap) in a solution of divalent silver ions and precipitate the plutonium as a sulfate salt, which could be safely stored for an interim period.

b. Plutonium extraction

Plutonium has usually been extracted from solutions by precipitation, ion-exchange, solvent extraction, or direct de-nitration.[3,4,13] For materials being converted to waste, a non-selective precipitation that also extracts other heavy elements such as uranium and americium is the most likely candidate, since this will produce a relatively clean, easily disposed supernate. The most promising precipitating agent is probably oxalate, since it was used extensively in past operations in a well-established technology.

Ion exchange was used extensively at Rocky Flats and Savannah River's FB-Line for plutonium extraction. Since high-purity plutonium is no longer a goal, highly selective ion-exchange processes will probably not be used to recover plutonium from solutions or dissolved scrap, although they may still be attractive for polishing supernates or other waste water.

Solvent extraction was used to separate plutonium from dissolved irradiated fuel at the Hanford PUREX plant and the Savannah River canyons, and to recover plutonium from dissolved scrap at PFP. Until recently, PFP and F-Canyon were planning to restart their solvent extraction processes as part of a program to stabilize reactive plutonium scrap and solutions, but current DOE planning is now focused on other alternatives. The prospect of resuming solvent extraction at PFP has become particularly remote because its equipment is designed to work with a carbon tetrachloride diluent, use of which may be prohibited under chloro-fluorocarbon restrictions. (Since CCl_4 is heavier than water and other common diluents are lighter than water, changing diluents would be a major modification to PFP's extraction process.)

Plutonium precipitated from nitrate solution can be directly converted to oxide by heating in air. This process, called direct de-nitration, avoids a separate processing step to extract plutonium from solution before converting it to oxide.

c. Calcination

Plutonium oxides can be stabilized by calcining (heating in air). Other plutonium compounds and plutonium metal can usually be converted to plutonium oxide by this method as well. The draft standard on long-term storage of plutonium would require calcination at 1000°C to drive off water and other volatile constituents sufficiently to allow long-term storage of the oxide in a sealed container.[1] Calcination at a significantly lower temperature (400-500°C is common) can produce a fairly stable oxide, depending on the feed material. The "low-fired" oxide may generate gases by radiolysis of residual impurities or water.

d. Salt treatment

Rocky Flats has a large inventory of residues from molten salt processing of plutonium, and LANL has a smaller inventory. The traditional method used at Rocky Flats for removing most of the plutonium and americium from salt scrap was to re-melt the salt and add aluminum (this was called the "scrub alloy" method). The aluminum product was sent to Savannah River for dissolution and plutonium recovery in the F-Canyon. LANL is developing a new method of plutonium and americium removal that involves melting the salts, sparging with oxygen to convert the plutonium to oxide, and vacuum distilling the salt.

3. Packaging

Packaging for short-term storage can be done with existing equipment and established procedures. Savannah River and Hanford have traditionally used hermetically sealed "food-pack" cans for storing both oxides and metals. Rocky Flats and LANL have more often used non-airtight slip-lid cans. All sites have used plastic bags to remove the inner container from the glove box through a bag-out port. The inner container and plastic bag have usually been placed in a second container. Again, Savannah River and Hanford have most commonly used food-pack cans for the outer container, while Rocky Flats and LANL have most commonly used slip-lid cans. These practices, possibly with small variations, will apparently remain in effect as plutonium metal and oxide are packaged or repackaged for interim storage (LANL is the exception, as discussed below).

Traditional packaging methods will not satisfy the requirements of the draft DOE standard on long-term storage of plutonium metal and oxide, as discussed in detail in Section III. The standard defines containers and atmospheres necessary to keep plutonium metal and oxide in safe storage for up to 50 years. All of the sites will need some new equipment and procedures. In particular, the sites will have to have "bag-less" transfer systems for removing items from glove boxes because the standard does not allow any organic material, such as bag-out plastic, to be included in the packaging configuration. The containers will have to be hermetically sealed and cannot contain elastomer seals, which eliminates conventional food-pack cans. The atmospheres in the containers must be very dry, which will require more stringent moisture controls than have existed in the past at most sites. Containers of metal could be backfilled with a dry atmosphere, but oxide has to be handled in a dry atmosphere from stabilization to final packaging in order to prevent absorption of water. Except for the moisture controls, LANL plans to implement the draft standard's packaging practices and repackage all metals and oxides before the end of 1995.[6]

B. Existing Processing and Handling Capabilities

The four major plutonium sites are physically capable of stabilizing most of their unstable plutonium materials. They have the necessary equipment, infrastructure, and trained personnel. Except for LANL, none of them is actually doing much stabilization work, but none is stopped by basic lack of capability. There are other reasons why they are not stabilizing the plutonium inventory: lack of funds for stabilization, unsatisfactory environmental or safety documentation, waste disposal problems, uncertainty about DOE's long-term plans for the surplus plutonium, or a general belief that stabilization is not urgent, compounded in some cases by local public opposition to processing operations.

The situation at the four main sites is summarized in Table 1. The rows list the basic plutonium processing, handling, and packaging capabilities required to put the plutonium into a stable form. The columns indicate the site's capabilities in each area. An "X" indicates that the site has the basic equipment, infrastructure, and personnel necessary for the task. It does not mean that anything is actually being done; in most cases nothing is. It does not mean that all desirable safety and environmental precautions are in place, or that all equipment is in optimum condition, or that all personnel are optimally trained. It only means that the capability is physically there.

Most of Rocky Flats' capabilities are still potentially available if modest repairs are made. As of April 1994, no processing systems, and only a few handling and repackaging capabilities, are officially active. Plutonium operations at the site were suspended in 1989. The only plutonium facility that has been officially restarted is Building 559, the analytical laboratory. A small-scale, low-temperature calcining capability has been prepared for restart in Building 707, although DOE has not yet authorized its restart. No other processing capabilities are scheduled to become available in 1994.

The only readily usable plutonium processing capabilities at Hanford are in the Plutonium Finishing Plant (PFP). The PUREX Plant had additional capabilities, but has been shut down longer than PFP and is already partly decommissioned. PFP can do most types of plutonium processing on a large scale. It is a canyon-type facility that can process reactor-grade plutonium and other fairly high-activity materials. Unfortunately, PFP is essentially shut down awaiting improved environmental documentation. It looks as though it may be several years before it operates. The main solvent extraction process used carbon tetrachloride as a diluent for the tributyl phosphate, and may never be able to operate again because of prohibitions against chloro-fluorocarbons.

LANL has the only general purpose plutonium processing capability in the Complex that is fully operational at this time. The TA-55 facility can process any locally generated scrap materials, convert plutonium from any form into any other, within reason, and comply with current packaging and storage standards. Their facilities are generally of smaller capacity than the other three sites.

Table 1 - Plutonium Processing Capabilities

OPERATION	ROCKY FLATS	HANFORD	LANL	SRS
Package metal for short-term storage	X	X	X	X
Package metal for long-term storage			X	
Package oxide for short-term storage	X	X	X	X
Package oxide for long-term storage				
Eliminate solutions	X	X	X	X
Inspect and repackage scrap	X	X	X	X
Incinerate combustible scrap				
Treat salt scrap			X	
Dissolve/leach and treat solutions		X	X	X

SRS has significant plutonium processing and handling capabilities in F-Canyon, H-Canyon, and associated support facilities. The canyons are reprocessing plants, so they are heavily shielded and capable of handling the highest activity materials. The past mission of F-Canyon was to extract plutonium from irradiated fuel and targets, and convert the extracted plutonium to metal. Its plutonium processing capabilities are extensive and of large capacity. Unfortunately, improved environmental documentation will be required before F-Canyon can resume operations.

V. FORMS OF PLUTONIUM FOR LONG-TERM STORAGE

Most forms of plutonium are chemically reactive, and plutonium is sufficiently radioactive that it subjects its surroundings to significant radiolytic attack. Most potential storage safety problems arise from the interaction of the plutonium itself with air, moisture, or radiolytic breakdown products. The breakdown products can come from the container, especially if it contains plastic, or from impurities in the stored material.

DOE long ago developed methods of keeping plutonium metal stable in weapon components, and until recently there was little need to store separated plutonium in other forms for long periods. The reactivity of plutonium was well known from many incidents of ruptured containers, fires, and small explosions. The underlying physical and chemical reasons for some of the incidents are still not well understood, although the plutonium plants learned how to take precautions. There has not been a large fire at a U.S. plutonium plant since the 1960s.

This section summarizes the storage properties of the two best-known forms of plutonium - metal and purified oxide - and compares their advantages and disadvantages. Metal and oxide are the forms identified in the draft storage standard as suitable for long-term storage. This section also discusses the prospects that other forms of plutonium can be developed for long-term storage. Most of the discussion here concerns the inherent properties of plutonium metal and oxide. Packaging issues are only discussed in a general context. The reactions that can occur when plutonium is incorrectly packaged or left too long in a package intended for short-term storage are cataloged more completely in Appendix B.

A. Storage Properties of Plutonium Metal

Plutonium metal reacts fairly rapidly with both free oxygen and water vapor. The rate of oxidation is controlled by diffusion of the oxidizing gas through a protective layer of oxide on the surface. As it forms, the oxide expands relative to the metal, cracks, and flakes off, exposing fresh surface for oxidation. The exact mechanism depends on the temperature and concentration of oxidizing gas, and on the exact composition of the metal. In fairly dry room air at room temperature, bulk alpha-phase plutonium metal oxidizes at a rate of about 1 micrometer per day. The bulk metal will not spontaneously ignite in air at room temperature, although the finely divided metal is quite reactive.[14,15]

Oxidation of the metal is catalyzed by the presence of water vapor. The mechanism of the reaction depends on the concentrations of water vapor and oxygen. Water vapor in the absence of air is the most corrosive environment for the metal. Recent investigation of the plutonium-water reaction has revealed evidence for the formation of a binary oxide containing Pu(VI), which is formed by the reaction of PuO_2 with water vapor. This material, written as $PuO_{2.2}$, has implications for the storage of PuO_2 and will be further discussed below.[16]

If the bulk metal is sealed in an inert atmosphere it can be preserved for a long time. Even a small leak in the container will eventually cause complete oxidation of the metal. Since the oxide is much less dense, extensive oxidation can rupture a container. There have been several such incidents, including some recently (see Appendix C).

Direct contact between the metal and any sort of plastic, such as bag-out plastic, is highly undesirable. The radiolysis of most plastics forms hydrogen, which quickly reacts with the metal to form pyrophoric hydrides. There are several adverse reactions and sequences of events involving plutonium metal and plastic (see Appendix B).

The form and composition of the metal influence the rate of the oxidation and hydriding reactions described above. The delta phase metal is more resistant to corrosion than the others, with an oxidation rate in air ten to one hundred times slower than alpha metal.[17] It is interesting that the draft standard for long-term storage does not rely on rate differences as a safety element in storage; the alloyed and unalloyed metal will be packaged the same. It is known that certain oxidizing atmospheres can actually stabilize the material by a passivation process, a fact that can be used to advantage in long-term storage.

B. Storage Properties of Plutonium Oxide

Plutonium dioxide, PuO_2, is a thermodynamically stable form resulting from the oxidation of the metal in an excess of oxygen or from the heating of plutonium compounds to high temperatures in air. The sesquioxide, Pu_2O_3, and a higher oxide, $PuO_{2.2}$, are also postulated to be present under certain circumstances.[15] PuO_2 itself is well characterized. Depending on how it is processed, it may have a high specific surface area, and may strongly adsorb many types of molecules. The effects of radiation on the adsorbates, and the consequences of radiolytic decomposition of the adsorbates, are not always understood.

If the oxide has a high specific surface area, there are several possible problems.[12] First, water and other molecules are tenaciously held on the oxide. Heating to 1000°C removes most of the water, but depending on the details of treatment, a monolayer of water molecules may persist. The water can oxidize the dioxide to produce the higher oxide and generate large quantities of hydrogen gas if stored for an extended period. The second problem is that radiolysis of adsorbed water and atmospheric gases could produce gaseous products such as NO_x and H_2, which would pressurize an unvented container and create an atmosphere that may be corrosive to the container. Radiolytic gas generation rates can be high because the adsorbates are normally distributed throughout the oxide and in intimate contact with the plutonium.

Calculations done at LANL suggest the magnitude of the pressurization problem for long-term storage.[12] Although such calculations involve many assumptions, a worst case can be evaluated in which thermal desorption, radiolytic decomposition, chemical reactions, and helium generation from alpha decay produce gas at maximum credible rates over a period of 50 years. A container is assumed to contain 4.5 kg of PuO_2 in 1850 cm^3 of free volume. A poorly processed oxide, containing 20 mg of H_2O per gram of PuO_2, would generate an internal container pressure as high as 2550 psia. For a well-processed oxide containing only 1 mg of H_2O per gram of PuO_2, the pressure could be as high as 85 psia. Due to the pressurization issue, there is a school of thought opposed to using sealed containers. DOE is taking the position that correctly processed and tested oxide would be acceptably safe for long-term storage in sealed containers. The majority of plutonium experts with whom the DNFSB staff and outside experts have spoken seem to accept this position, although there is a significant minority of experts who differ.

There are differences between metal and oxide in their potential dispersability in the event of a severe accident. Fundamentally, metal is not dispersable and oxide is. The oxide source term would depend on the temperature at which oxidation takes place. Room temperature oxidation of the metal or low-temperature firing (less than 500°C) of most oxide precursors leads to very small particles, nearly 100 percent dispersable with a large fraction (40 weight percent) in the respirable range.[19] Treatment of the oxide at 1000°C causes some sintering of the particles, and reduces the dispersable and respirable fraction (the latter may be 10 percent).[19] The oxidation of metal at high temperatures, as in a building fire, results in an oxide with far fewer than 1 percent of its particles in the respirable range.[20] The experiments in which these results were obtained are old, but the results are consistent with conceptual models of the physical processes involved in the surface oxidation. Low temperature oxidation leads to a brittle cracking of the oxide as it is formed due to the brittle underlying metal. At high temperatures, the metal becomes much more malleable and allows the oxide layer to grow thicker before it cracks, resulting in the spallation of larger particles.

C. Alternative Forms for Long-Term Storage

There does not appear to be much interest around the Complex in further research into the safety implications of long-term plutonium behavior. There does not appear to be any sponsorship of such research by DOE or other parties. DOE has sponsored a series of meetings to collect opinions and formulate criteria for storage of oxide and metal, but the report issued after the meeting did not identify a specific need for further research.[12]

The DNFSB staff and outside experts are of the opinion that DOE could sponsor some further thinking, if not necessarily actual research, into some alternative plutonium storage forms and containers. There are three areas that call for more attention:

1. storing plutonium oxide as sintered pellets;
2. storing plutonium metal as a corrosion-resistant alloy; and
3. using vented containers for some materials.

It would not be difficult to form plutonium oxide into hard, dense pellets. Pellets of PuO_2 have been fabricated in the past in DOE facilities and mixed oxide pellets have been fabricated in commercial facilities, although none of those commercial facilities are operating now. The technology is still active at DOE facilities for making heat sources from Pu-238 oxide. Oxide pellets for storage would not have to meet tight dimensional specifications or be of perfectly uniform density like reactor fuel pellets. The idea would be to put the oxide into a less dispersable form and a form with a lower specific surface area. That would reduce the potential health consequences of future accidents and would reduce the problem of gas generation by radiolysis of adsorbates.

Corrosion of stored metal is undesirable for several safety-related reasons: corrosion converts the plutonium to a more dispersable form, expanding oxide may rupture the container, and, under some circumstances, pyrophoric hydride may be formed. DOE has a great deal of experience with delta-phase plutonium alloys, which are known to be much more resistant to corrosion in most storage environments than alpha-phase plutonium. Many other plutonium alloys have been investigated on a laboratory scale.[4] It is possible that some of those alloys may have properties that would make them especially corrosion resistant in long-term storage. As far as the DNFSB staff has been able to ascertain, DOE is not sponsoring any investigation of this subject.

Vented containers would offer several benefits. As long as the vents do not get plugged, the containers could not pressurize when gases are generated. Also, if air could freely flow into the container, pyrophoric reaction products could not accumulate. Pressurization and accumulation of pyrophorics are the two main long-term safety problems with sealed containers. A vented container would also probably be easier to design and fabricate than the elastomer-free sealed container currently envisioned by DOE. Vented containers would not require an inert atmosphere or leak testing, so the packaging operation itself would be simpler.

Vented containers would be most attractive if the plutonium were in the form of pressed pellets or corrosion-resistant metal. Loose oxide, not pressed into pellets, may be prone to obstructing vent holes. An obstructed vent hole would be highly undesirable since it would cause the container to pressurize to the unpredictable point where the obstacle is blown out. Unalloyed plutonium metal could only be stored in vented containers if the atmosphere in the storage vault was dry and inert, or if it was considered acceptable for the metal to gradually corrode to oxide.

As a longer-term solution, the National Academy of Sciences has recommended that surplus plutonium be disposed by incorporation into glass for deep geologic disposal or burned as fuel in nuclear reactors.[21] It is beyond the scope of this report to evaluate those propositions. It is clear that the National Academy is motivated primarily by proliferation concerns rather than safety concerns. If DOE adopts one of the disposition options preferred by the National Academy, it would become academic to debate the long-term safety nuances of plutonium metal and oxide. The Secretary of Energy has recently constituted a task force to consider all points of view on the subject of long-term plutonium disposition, but it is too early to predict what its recommendations will be.[22] The White House Office of Science and Technology Policy has recently organized an interagency group, including representatives from DOE, to consider the same general issues.[23]

VI. ORGANIZATIONAL RESPONSIBILITIES

Figures 1 through 4 show the DOE line management chain for plutonium processing and storage at the Hanford, LANL, Rocky Flats, and Savannah River sites. The figures show the positions at DOE involved in direct line management of plutonium. There are other DOE positions involved in related areas such as safeguards, stockpile management, and the development of new standards. In addition, the newly formed Plutonium Disposition Project, which is a matrix organization with resources temporarily drawn from line organizations, will apparently be responsible for making decisions on many plutonium issues.

Figure 1 shows the DOE organization for management of plutonium at the Rocky Flats Plant. Of the four sites discussed in this paper, the DOE organization for management of plutonium at Rocky Flats is the most complex. Plutonium responsibilities are divided among three divisions: Transuranic (TRU) Waste Management, Material Management, and Operations. The TRU Waste Management Division includes teams to follow planning and actions taken by the contractor for disposition of liquids and solid residues. The Material Management Division is responsible for material control and accounting, plus other material management activities. The Operations Division has direct oversight of day-to-day operations, including safe storage of plutonium.

Figure 2 shows the DOE organization for management of plutonium at the Hanford site. The Facilities Transition Branch of the Operations and Transition Division has the responsibility for Richland Operations Office oversight of all Plutonium Finishing Plant operations, which would include the safe storage of Hanford's inventory of separated plutonium.

Figure 3 shows the DOE organization for management of plutonium at LANL. Direct facility operations oversight of the TA-55 Plutonium Facility is provided by the Facility Operations Branch of the Los Alamos National Laboratory Area Office. The Environment, Safety, and Health Branch is responsible for overseeing safety issues associated with plutonium processing and storage. The Operations Management Division and the Safety Programs Division at the Albuquerque Operations Office have a higher level responsibility for providing oversight of safety issues associated with plutonium processing and storage at LANL.

Figure 4 shows that at the Savannah River Site, direct DOE oversight of operations in the F-and H-Areas is provided by the Separations Division. The responsibilities of the Separations Division include direct oversight of safety-related plutonium storage issues in H-Canyon, F-Canyon, FB-Line, and the 235-F Vaults. The Technical Division provides technical matrix support for the Separations Division.

For each site's organization chart there is a dotted line connecting the Office for Field Management (FM) at DOE HQ to each of the operations offices. This is meant to indicate that it is still unclear what role FM, a relatively new organization, will eventually play in plutonium storage and processing at the sites. A dashed line is used to connect the Office of Defense Programs (DP) and the Office of Environmental Restoration and Waste Management (EM) of DOE HQ to the operations offices to indicate that DP and EM generally interact with many levels of the operations offices, not just with the operations office manager.

Figure 1 - DOE Rocky Flats Organization

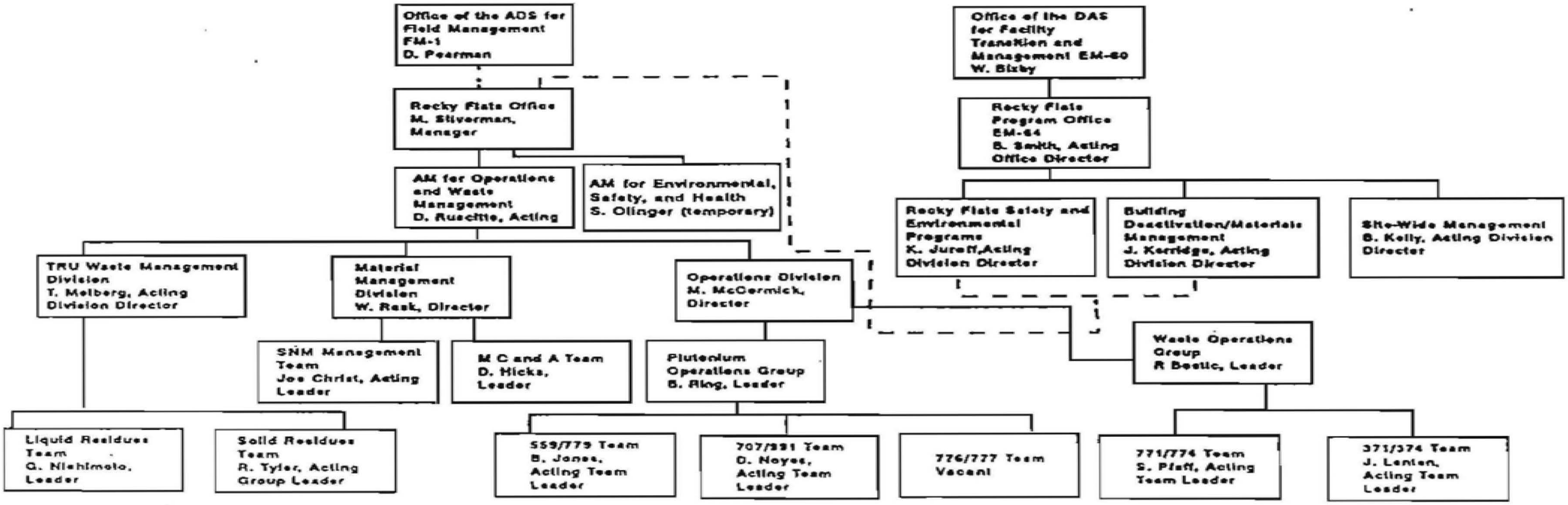


Figure 2 - DOE Hanford Organization

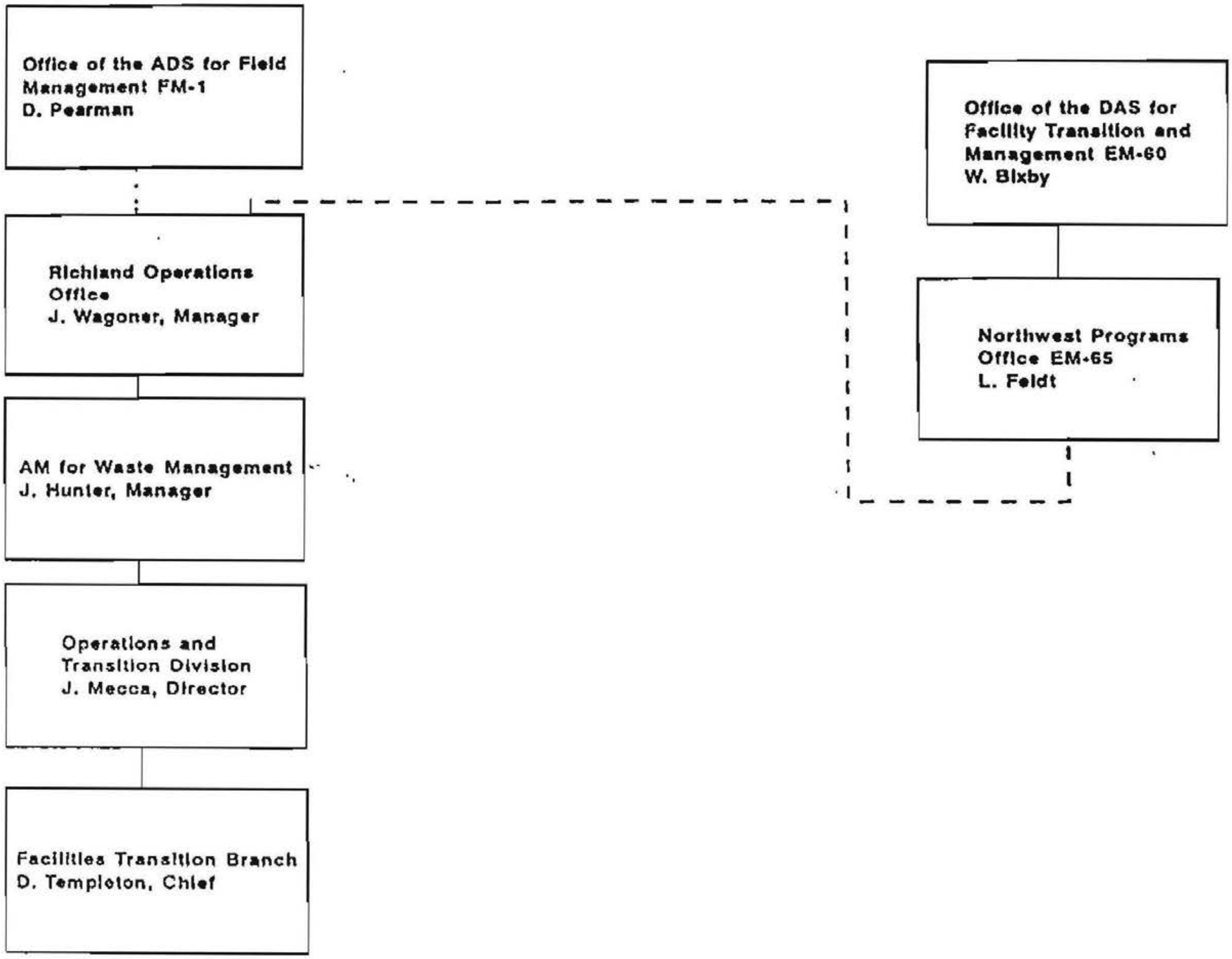


Figure 3 - DOE Los Alamos National Laboratory Organization

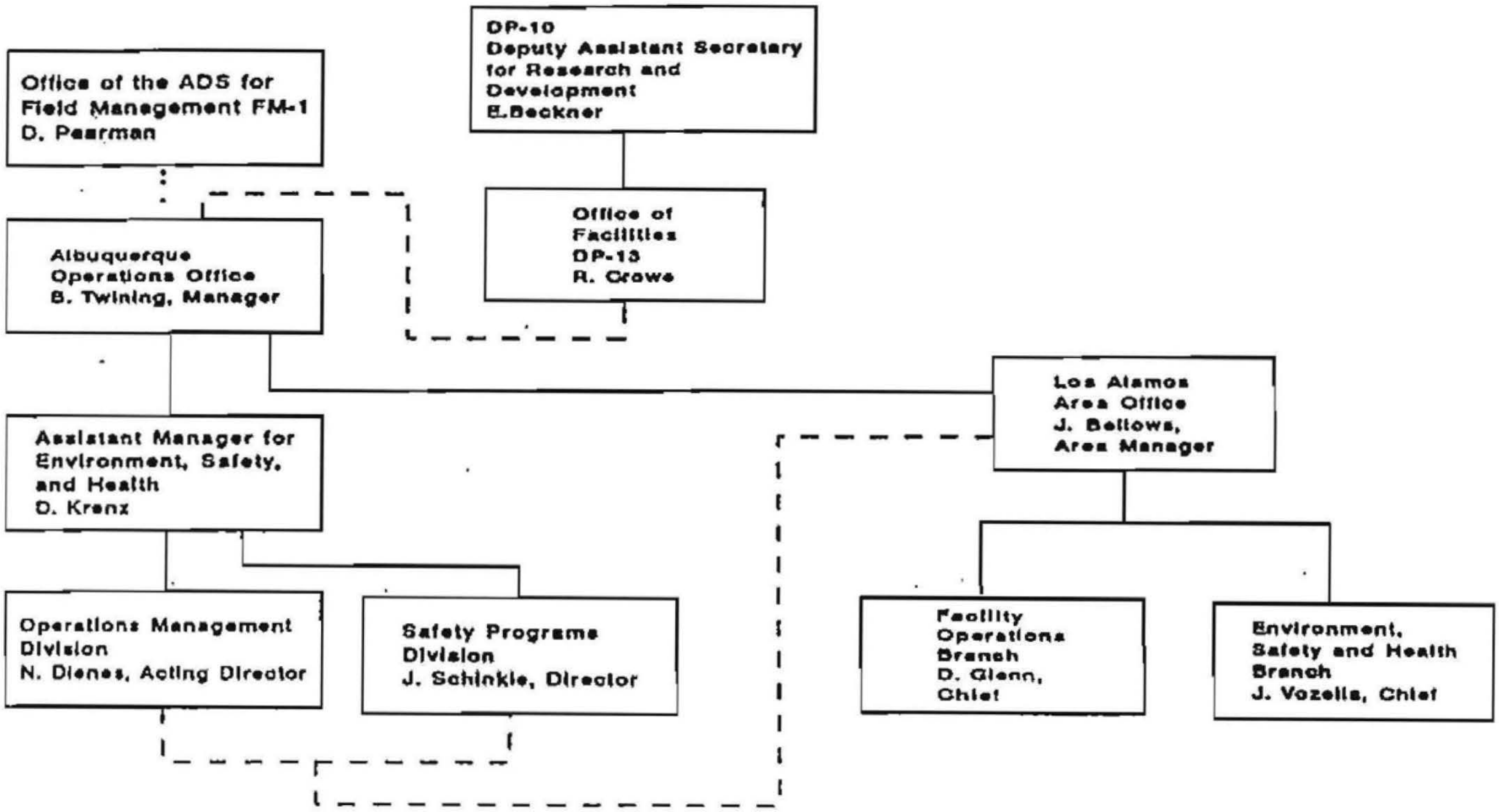
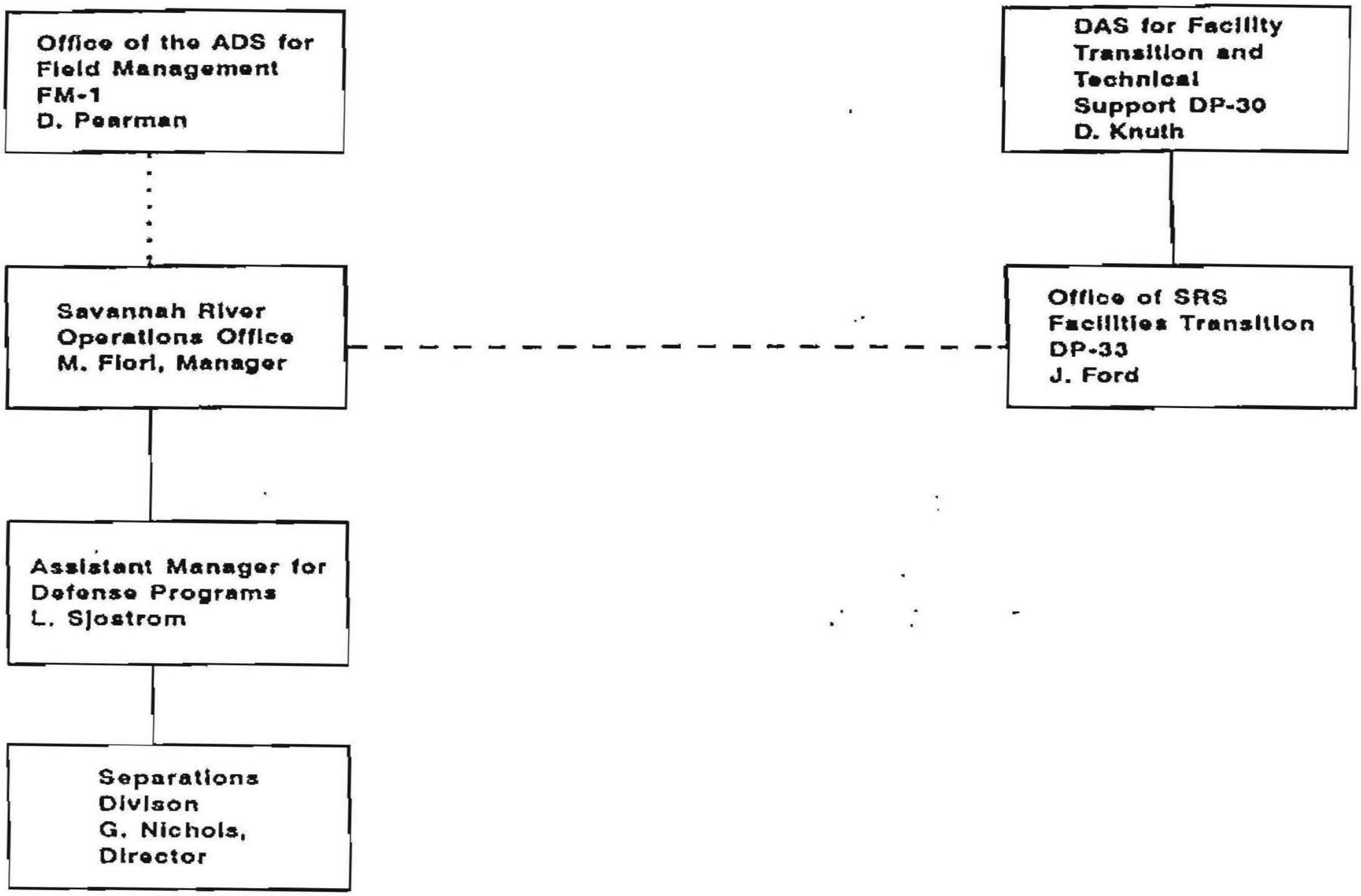


Figure 4 - DOE Savannah River Organization



Appendix A - Plutonium Storage Conditions at Four Major DOE Facilities

I. Introduction

Many former DOE facilities that produced weapons materials and components are now undergoing a transition from their original purpose to a different mode of operations, generally involving shutdown and cleanup. During the production years, most of these facilities processed plutonium materials at a rapid pace to meet production goals and schedules. Any materials that contained easily recoverable quantities of weapons-quality plutonium were usually processed as rapidly as practicable. As a result, most of the sites kept only relatively small amounts of plutonium in storage. In most cases stored plutonium materials were considered to be in-process stocks and were stored for only relatively short periods of time. They were converted to weapons materials as rapidly as facilities and operations would permit. Scrap materials that were difficult or time consuming to convert to weapons-usable material were in some cases held in storage for extended periods. Some such scrap materials have been in storage for a decade or more by now.

Several accidents have occurred, both in the US and abroad, involving pressurization of containers, fires, explosions, and spread of contamination as the result of improper storage of plutonium. It is not clear that present plutonium storage practices at some DOE facilities reflect a recognition of such incidents. There have been several plutonium storage incidents within the past year at DOE facilities (see Appendix C for a summary). In fact, several incidents have occurred at DOE facilities subsequent to the visits by the DNFSB staff and outside experts.

The DNFSB staff and outside experts have visited the four main DOE plutonium facilities to review the safety of plutonium in storage. The method of gathering information was to conduct interviews with experienced DOE or contractor personnel who had first-hand experience in the handling, processing, packaging, or storage of plutonium. The scope of this review was plutonium in liquid solutions, and in such solid forms as metals, oxides, other compounds, mixtures, and scrap. This review did not consider plutonium in encapsulated forms such as weapons components or sources since the technology for safe long-term storage of such items has been well established for many years. The review did not consider irradiated spent fuel or target materials containing plutonium. It did not consider materials considered to be wastes which may contain plutonium in low concentrations.

II. Summary of Information from Individual Sites

A. Visit to the Rocky Flats Plant on September 22-23, 1993

From the 1950s to 1980s, Rocky Flats was the major plutonium materials processing and fabrication operation for the DOE Complex. It handled the largest inventory of plutonium materials and had a large variety of plutonium processing operations. It retains in storage the largest quantity of unencapsulated plutonium in the Complex. The inventory of plutonium at Rocky Flats is 12.9 metric tons. This consists of 6.6 metric tons of metal and 6.3 metric tons of compounds and mixtures.

1. Liquid Materials

At the time of the shutdown of operations in 1989, 11,000 liters of plutonium solutions were in tanks and bottles in Buildings 771 and 371. As much as 9,000 additional liters of plutonium solutions are contained in tank heels and process lines, some of them overhead in occupied areas.

Some of the tanks are of geometrically safe configuration, while others contain solid poison materials to assure the criticality safety of the contents. Normal practice would be to examine poisoned tanks annually to assure the integrity of the poison material and solution composition. However, such examinations have not been carried out for a number of years and thus the exact status of the solutions and contents are not clearly understood. Many of the tanks have bottom connections with flanges and valves. As time passes, there are ever more leaks from the flanges and valves, causing contamination of occupied operating areas.

DOE understands these hazards and corrective actions are being planned. However, the time required to get such action completed appears to be far too long in light of the existing hazards to operating personnel, and the fact that handling such liquids was a routine matter in the past.

2. Solid Materials

During the clean out of glove box and processing systems (circa 1989-1990) many forms of material containing plutonium were rapidly packaged for what was intended to be a short time. Due to changing circumstances, the storage period has extended and many items remain in storage to this date (four to five years later). Many different material forms were placed in storage in a variety of packages.

Most weapon components are well packaged, in accordance with established practice for such items. Some metal items were bagged out of glove boxes with the plastic in direct contact with the plutonium metal. These items are subject to rapid oxidation and possible generation of pyrophoric plutonium hydride. They should be repackaged as soon as possible.

For most scrap materials, no packaging standards exist. Some scrap was in storage before the 1989 shutdown; others were packaged in the rapid clean out that was undertaken in 1989. There are over 4,000, 55-gallon drums of scrap in storage, containing a wide variety of materials in a wide variety of internal packages, many of which contain plastic or other organic materials. There are thousands more 10-gallon drums containing similar scrap materials. The contents of many of these are not well characterized.

A recent report on scrap stored in 55-gallon drums was prepared by a senior Rocky Flats engineer.[24] This report evaluated the safety risks of the residue drum storage packages. It assigned risk factors to the various types of scrap. The conclusion of this evaluation was that about a thousand drums contain materials or combinations of materials that are in the most dangerous category and may be susceptible to reactions that could generate pressure or flammable products. An additional thousand drums are in the second highest category of safety risk. Thus about one half of the residue drum packages in storage are considered to be relatively high safety risks. It should be noted that this assessment was based on assumptions as to the most likely material contents and packaging materials, not positive knowledge of scrap composition and packaging.

According to the report, the following generic categories of materials are the most suspect from a stability standpoint:

- salt residues, including electrorefining, molten salt extraction, and direct oxide reduction salts;
- combustibles such as filters, ion-exchange resins, and potentially nitrated cellulose materials;
- skulls, which are metallic residues from casting molds;
- sand, slag, and crucible (SSC), which may contain unreacted metal; and
- peroxide precipitation residues.

The exact composition of the materials in many packages is not certain, and this causes additional concern. The salt residues may contain unreacted calcium or magnesium, and the combustibles may contain organic materials that may have nitrated to a reactive form. Such possibilities are of concern since they can act as ignition sources.

There is evidence that these packages contain organic materials in their inner packaging, or in the form of plastic containment bags or tape. There is ample evidence from past occurrences that organic materials in contact with plutonium are subject to radiolysis accompanied by generation of flammable or corrosive gases, and heat. Degradation of organics and radiolytic generation of hydrogen will continue as long as organic materials are in contact with or in the same package as plutonium.

3. Storage Locations and Conditions

There is little standardization in the design and operation of the many storage rooms and vaults at Rocky Flats. Apparently, as the number of residue packages grew with increasing production pressure, and later when process lines and equipment were emptied in the early 1990's, almost any available usable area was pressed into service for storage with little concern for the services provided in the area.

Now certain areas in Building 371 are being planned for use as fairly long-term storage areas. From a standpoint of resistance to the effects of natural phenomena, this building is the most suitable structure on the site and thus should be an improvement over the other buildings in that regard.

B. Visit to the Los Alamos National Laboratory (LANL) on October 27-28, 1993

(Outside expert, Dr. Joseph Leary did not participate in this visit.)

The LANL plutonium processing facilities have operated in a significantly different manner from Rocky Flats. LANL operated mainly as a developmental laboratory for special materials and processes, although for a period in the 1980's LANL did produce a large quantity of high-purity plutonium metal for use at Rocky Flats. The plutonium inventory at LANL is 2.6 metric tons.

In general, the quantities of plutonium handled and now stored at LANL are much smaller than at Rocky Flats. In addition, the LANL staff has always been technically oriented while the Rocky Flats staff was production oriented. Partly because of the safety issues, and partly because of a shortage of storage space, LANL did not accumulate large quantities of plutonium scrap. As such materials were generated, they were usually processed into more stable forms fairly quickly.

Since the plutonium processing facilities at LANL are currently operational, they are able to continue reducing scrap inventories and to improve the condition of stored materials as necessary.

1. Liquid Materials

The only liquids in storage at LANL are solutions associated with on-going processing operations. The practice has been to only generate solutions as they are needed for processing campaigns and then to process them to solid forms. There appears to be no current issue at LANL concerning liquids in storage.

2. Solid Materials

LANL personnel appear to be aware of the hazards of improper storage of solid plutonium materials. They have avoided the use of organic packaging material in direct contact with plutonium. They have used plastic bags only outside of an inner package.

As at Rocky Flats, LANL has generated significant amounts of cellulose waste material that has been in contact with nitric acid. However, the LANL approach to storing such materials is much different from that at Rocky Flats. At LANL such material is stored under water in metal cans in glove boxes where it can be easily monitored. LANL personnel are sufficiently concerned about the safety of potentially nitrated cellulose that they monitor such items on a daily basis. At Rocky Flats such materials are stored in packages in storage rooms or vaults without any specific monitoring or surveillance.

LANL has accumulated a significant quantity of pyrochemical salt scrap, as well as some sand, slag, and crucibles from metal production. However, the quantities at LANL are far less than at Rocky Flats. LANL's salt recovery process is operational and the inventory of salt scrap is being reduced.

3. Storage Locations and Conditions

Most of the plutonium material at LANL (more than 90 percent) is stored in the vaults in the TA-55 area. These vaults are small and LANL has decided to prepare an additional vault. LANL has developed criteria for plutonium storage vaults and has served as the lead laboratory for criteria development for the Complex 21 plutonium storage effort.

C. Visit to the Hanford Site on December 7-8, 1993

Hanford was the first site to make production quantities of plutonium and to process large amounts of plutonium metal in the early part of the DOE weapons program. This work was carried out at the Plutonium Finishing Plant (PFP) in the 200 West Area. Later in the program, as Rocky Flats and Savannah River came into operation, the mission at Hanford was reduced, and in recent years the quantities of plutonium handled and processed have been relatively small. The inventory of separated plutonium at Hanford is 3.8 metric tons. There are almost 8000 containers of plutonium material, with about 7350 items in vault storage, 350 items in glove box storage, and 200 items retained in the PFP laboratory. A majority of the plutonium at Hanford is not weapons grade. For several years Hanford was the Central Scrap Management Office (CSMO) for the DOE Complex and received many unusual plutonium materials from other sites. Many of these items are still stored in the original shipping packages at Hanford.

1. Liquid Materials

Plutonium solutions have been in storage at PFP for decades. Most of these are retained in the original shipping containers (several hundred) that consist of an inner plastic bottle in a stainless steel flask within a conventional steel drum. While most of these solutions are nitrate, there are also some (25-30) that contain chlorides and fluorides.

The inner containers are supposed to be vented so that gases generated from radiolytic decomposition of water or degradation of the plastic bottle are released. However, neither the condition of the vent openings nor the plastic bottles is known. There had been some concern that the plastic bottles may have degraded to the extent that they no longer contain the liquids, and that the solutions may have been significantly concentrated by evaporation, to the point of being in the form of sludges or other solids. Since the group's visit to Hanford, PFP operators used X-rays to evaluate the packages containing chloride or fluoride, and concluded that the plastic bottles are still intact in those cases.

2. Solid Materials

There are sludges from oxalate precipitation and solvent extraction operations stored in plastic jars in glove boxes. These materials are probably the most reactive in storage at Hanford, which is why they are kept in glove boxes and not consigned to vault storage. In addition there are polystyrene cubes used for criticality experiments that contain plutonium. These are being stored in plastic bags in vented metal cans. In the past, some of these cubes have been processed by destructive distillation to remove the styrene. However this technique deposited some styrene in glove boxes and will not be used again.

Plutonium metals and oxides at Hanford are generally packaged in small steel food-pack cans with a crimped lid containing an elastomer seal in the crimp. This can is contained in a plastic bag within at least one larger steel food-pack can of the same design. Both the elastomeric gaskets in the crimped seals and the plastic bags are subject to radiolytic degradation over the long run. This type of packaging is fairly standard at Hanford and Savannah River, and has been satisfactory for interim storage of clean metal and pure oxide.

Sand, slag, and crucibles from metal production operations have been packaged in a crimped sealed inner steel food-pack can within a plastic bag, then placed in a lard can with a taped lid. There are several hundred of these packages, and they are not considered as safe as the double-canned materials. There have been some incidents of these cans bulging and requiring repackaging. In addition, a considerable amount of incinerator ash from Rocky Flats is now at Hanford and will require some action to improve its long-term stability.

3. Storage Locations and Conditions

The vaults at Hanford seem to be well designed. In some vaults, packages are stored on pedestals that contain temperature alarms and an apparatus to detect bulges on the bottoms of the cans. The vaults also contain continuous air monitors and other surveillance systems, and undergo inspections on regular intervals. Many of the items in the vaults were repackaged around 1980. Typically, two or three cans are repackaged annually when visible signs indicate a build up or decrease of pressure.

D. Visit to the Savannah River Site (SRS) on January 5-6, 1994

SRS was the major site in recent years for producing and processing plutonium for the weapons program. Plutonium processing activities were primarily carried out in the F-Canyon and FB-Line. In general, the F-Canyon was used to separate plutonium from irradiated targets. FB-Line was used to produce plutonium metal from F-canyon product solutions.

The unencapsulated plutonium inventory at SRS is 1.8 metric tons, of which 0.5 metric tons is metal and 1.3 metric tons is oxide and other compounds. There are over 2200 solid items in storage, of which slightly over 300 contain Pu-238.

1. Liquid Materials

The F-Canyon was shut down in 1989 without working off in-process solutions. Despite several attempts to restart the plant to process these liquids, nothing has been accomplished in this regard in the past five years. About 380,000 liters of solution containing plutonium is contained in 18 tanks in the F-canyon. The FB-Line was cleaned out much more thoroughly.

The shutdown of F-Canyon was so abrupt that some undissolved target elements were left in the dissolver. (They are dissolved by now.) It even seems that not all of the solvent extraction contactors were flushed out, so that highly radioactive solutions still remain in intimate contact with some organic liquids. A similar situation, with liquids remaining in tankage for several years, exists in H-Canyon on a smaller scale.

In early 1993, operations personnel detected that the liquid level in one F-Canyon tank had decreased dramatically from the original level at shutdown. Upon sampling, it was found that the solution had concentrated and some plutonium precipitate had been formed. The situation posed some risk of accidental criticality. SRS eventually added water, dilute acid, and neutron poison material to the tank.

Fortunately the process equipment in both canyons is designed without connections or gaskets below the tank liquid level. Thus corrosion or deterioration of bottom connections, gaskets, or valves is not possible as at Rocky Flats. Also, since the tanks are located in remotely operated canyons, behind heavy shield walls, the direct risk to plant personnel from accidental criticality or leaks is much lower than at Rocky Flats.

2. Solid Materials

There are plutonium metal buttons in storage, half weapons grade and half reactor grade. The buttons are packaged in double food-pack cans, similar to Hanford.

Plutonium oxides, some relatively pure and some mixed with other oxides, are stored in several manners. The most prevalent form is a crimped food-pack can similar to the inner Hanford storage can. The cans are contained in a plastic bag inside a steel five-gallon pail. A small amount of oxide from Lawrence Livermore National Laboratory is stored in plastic screw-top jars. A considerable amount of weapons-grade oxide from Hanford is stored in double food-pack cans inside shipping containers.

A wide variety of plutonium alloys, compounds, and residues is stored at SRS from the days when it served as the Central Scrap Management Office. The packaging of these materials is diverse, and in many cases the condition is not well known. One residue material containing plutonium oxide and organic die lubricant was packaged in crimped food-pack cans. One of the cans pressurized in the 1980's and caused contamination that required six months to clean up. Following this incident, all remaining packages of this material were repackaged in cans with small HEPA filters to permit venting. The filters are now checked for plugging quarterly.

There are a few drums of Rocky Flats incinerator ash in the original shipping containers. Most sand, slag, and crucible (SSC) generated at SRS has been processed. However, about 100 packages of SSC from other sites are stored in slip-lid cans inside five-gallon pails. This material can contain unreacted calcium that can generate hydrogen gas on exposure to moisture.

3. Storage Locations and Conditions

SRS has several vaults in the FB- and HB-Lines for the storage of packaged solid materials. The vaults are not instrumented as they are at Hanford, although randomly selected metal items are weighed at regular intervals to check for excessive oxidation.

Late in 1993, a plutonium metal button packaged in a double food-pack can was noted to have gained about 70 grams over its nominal button weight after about four years of storage. It was examined by X-ray, and the inner can was observed to be almost completely full of oxide. Apparently the crimp seals on both cans were defective. This illustrates the value of careful monitoring of stored packages.

III. Conclusions

A. Rocky Flats Plant

Rocky Flats has the most hazardous plutonium storage conditions in the Complex.

The continuing storage of plutonium solutions in tanks and piping is one of the most severe hazards at Rocky Flats or anywhere in the Complex. The connections in the tanks and piping are already leaking. Criticality safety is increasingly difficult to guarantee. The Raschig rings in the non-geometrically favorable tanks, which are supposed to be inspected every year, have not been inspected for several years. The systems required for moving liquids have not been operational for several years and thus mixing of the liquids for proper sampling or concentration control can not be carried out. The tanks are in occupied areas; a criticality incident could cause fatalities. These liquids should be processed to a stable solid form as a very high priority item.

There are so many types of solid materials that might not be stable in their present storage form or packaging that it is difficult to choose the single most hazardous category. Salt residues are a major concern and should be repackaged to eliminate as much plastic material as possible. A second category of materials that should receive attention are the combustibles that may be nitrated.

Many conditions at Rocky Flats are in conflict with good packaging practices for plutonium and storage conditions leading to a potential incident will continue to worsen with time. The fact that this site has not had many recent incidents is no reason to be complacent. Some poor practices used at this site give cause to considerable concern that incidents may be expected.

B. Los Alamos National Laboratory

It appears that LANL is proceeding with the processing of its stored materials in an appropriate manner and that the present course is appropriate for the residues that are on hand at that site. There have been some recent incidents with stored plutonium at LANL, showing that continual vigilance is required.

C. Hanford Site

The leading concern at Hanford is probably plutonium solutions in bottles. These liquids should be processed to a stable solid form as soon as possible. The solutions of most concern are those containing chlorides or fluorides due to their potential to corrode the secondary stainless steel containers. Other reactive materials retained in glove boxes should be stabilized as rapidly as practicable.

D. Savannah River Site

The liquid storage conditions at the F-Canyon appear to be the most pressing issue. The solutions in the F-Canyon should be processed to a solid form as a high priority item. While there may be other possible alternatives for placing the liquids in a more stable form, one logical solution to this problem, as suggested by site personnel, would be to restart the F-Canyon to work off the present inventory of solutions and to convert the plutonium to a stable solid form. In any event, some early action should be taken to process these liquids.

Appendix B - Adverse Interactions of Stored Plutonium with its Surroundings

This appendix presents a brief analysis of physical and chemical reactions that are known to affect plutonium metal and plutonium oxide in storage. The phenomena are not mutually exclusive; two or more of them may operate at the same time on a particular container of stored plutonium. For example, a piece of plutonium metal may be stored in a moist, inert atmosphere (case III) inside a plastic bag (case I), and as oxide forms on the metal, the oxide would itself be in contact with the plastic (case VIII).

I. Metal in direct contact with a plastic bag, isolated from ambient air.

All experts agree that plutonium metal should not be stored for any length of time in direct contact with plastic, and no facility has made a general practice of storing plutonium that way. The concern is that some metal items at Rocky Flats may have been stored directly in bag-out plastic as a temporary measure, and that there has not been opportunity yet to repackage them.

Almost all plastic is susceptible to radiolytic decomposition, particularly under the influence of alpha radiation. Radiation causes the plastic to become brittle and weak, and causes the release of hydrogen gas and sometimes hydrochloric acid vapor, depending on the type of plastic. The gases can be expected to be released mostly on the inside of the plastic bag, although a certain percentage may be released on the outside. The hydrochloric acid may corrode both the plutonium metal and, if the bag leaks, the container. The hydrogen gas will pressurize the bag and eventually the container as well, plus the hydrogen is a fire hazard should an ignition source be introduced.

Any hydrogen released inside the plastic will react rapidly with the plutonium metal, producing plutonium hydride.

$$\mathrm{Pu} + \mathrm{H_2} = \mathrm{PuH_2} \qquad (1)$$

Hydride is pyrophoric, meaning it will spark and flame upon exposure to ambient air.

$$\mathrm{PuH_2} + \mathrm{O_2} = \mathrm{PuO_2} + \mathrm{H_2} + \text{Heat} \qquad (2)$$

As long as free oxygen is present inside the plastic, the hydride will react quickly with the oxygen. Experience indicates that when plutonium is in direct contact with plastic, it does not take long before sufficient hydrogen is generated to form enough hydride to deplete whatever oxygen was initially inside. Plutonium metal is often handled and packaged in a low-oxygen atmosphere, so the initial oxygen concentration may be minimal. In any case, once the oxygen is depleted, hydride can accumulate.

The plastic will embrittle under the influence of alpha radiation and will eventually break open. If the plastic breaks before the container is opened, hydrogen that has accumulated outside the plastic but inside the container will rapidly react with the metal to generate more hydride. At the same time, any free oxygen within the container (which would have been isolated from the hydride up to now) would quickly oxidize some of the hydride, releasing more hydrogen and generating heat. It may be theoretically possible that a combustible mixture of hydrogen and oxygen could be ignited inside the container by the heat of the hydride oxidation reaction, although there are no documented cases.

If the plastic is intact when the container is opened (which is impossible to know in advance) the container itself can be safely opened as long as any trapped hydrogen can be dispersed without igniting. The main danger point is when the worker tries to pick up the plastic bag, which will probably break and allow air to come into contact with the hydride. In that case, there will be sparking and flaming, with the spread of contamination and risk of a larger fire if other combustibles are in the vicinity. This phenomenon has occurred many times, some recently (see Appendix C).

II. Metal in direct contact with a plastic bag, not isolated from ambient air.

In this case, the plutonium metal will undergo reactions (1) and (2) as before. If the bag is still intact when the container is opened, this case is identical to the previous one. If the bag breaks before the container is opened, the situation is somewhat different.

Free oxygen inside the container will quickly oxidize any accumulated hydride, generating a burst of hydrogen and heat. Again, ignition of hydrogen inside the container is a theoretical possibility, but probably unlikely. It is more likely that the hydrogen will react with the metal to produce more hydride. Since air can flow into the container from the surrounding atmosphere, this reaction will continue until all the metal is oxidized. The hydride formation process will accelerate the oxidation reaction far beyond the rate characteristic of dry, relatively hydrogen-free air.

The main risk is that oxidation could rupture the container since the oxide has a much lower density than the metal. In general, the sites try to allow enough free space in the container to hold the oxide. In practice, there have been examples of containers rupturing even when there was still free space remaining, apparently because of a "wedging" phenomenon, where the metal is pushed against one corner of the container by rapid oxidation on one side of the metal. There was one example of this quite recently (see Appendix C).

If oxide formation causes a container to rupture, there may be severe spread of contamination in areas where such is not expected. Also, if oxidation is not complete when the container ruptures, any remaining hydrides may react violently with air, dispersing contamination. Some sites weigh containers of metal to monitor oxide formation, which is useful, but the oxidation can proceed so rapidly in some cases that intermittent weighing is not certain to detect the problem in advance.

III. Metal in moist atmosphere, isolated from ambient air.

Plutonium metal will react vigorously with the water vapor. The basic reaction forms plutonium oxide and hydrogen.

$$Pu + 2H_2O = PuO_2 + 2H_2 \qquad (3)$$

The hydrogen will react with the metal to form hydride as in equation (1). The hydride can react with free oxygen if there is any, or with water as in equation (4). Oxygen seems to inhibit the reaction of water with plutonium metal, so the hydrolysis may proceed faster in an oxygen-free atmosphere than in air.[24]

$$PuH_2 + 2H_2O = PuO_2 + 3H_2 \qquad (4)$$

The exact mix of products from this sequence of reactions can be quite variable depending on the conditions, and is the object of continuing investigations. Plutonium hydroxide, $Pu(OH)_3$, may be a product of the reaction of the hydride with water. This would react with available oxygen to form plutonium dioxide.[17] Pyrophoric hydrides and even metal fines have been found mixed in with the oxides.[17] More recently, studies of the high-temperature reaction of water with plutonium suggest that the hydrolysis reaction forms a multilayered oxide containing hydrogen.[16]

These reactions will continue until all the water and oxygen in the container are used up, resulting in a potentially pyrophoric mixture in a hydrogen atmosphere. The amount of hydrogen generated is probably not enough to seriously pressurize a container, but if the container is opened in air, the pyrophorics can be expected to cause an energetic reaction, spreading contamination and creating a fire risk.

IV. Metal in moist atmosphere, not isolated from ambient air.

This case is similar to the one above except that the supply of air is unlimited. If the air is moist, the metal will completely oxidize in a fairly short time. Drier air will result in a slower oxidation rate, but the end result will eventually be the same. The container could rupture if there is not enough room for the oxide to expand.

V. Metal in nested non-airtight containers with airtight plastic bag between them.

Much of the unencapsulated metal in the DOE inventory is stored in this arrangement, particularly at Rocky Flats and LANL. It is probably satisfactory for periods of 1-2 years, but not for much longer.

Nominally, the inner container will protect the plastic bag from the plutonium's alpha radiation. (The plastic will still be subject to gamma radiation, but that tends to act on the plastic more slowly.) The protection is only effective, though, if the outside of the inner container is almost totally free of plutonium contamination, a condition not often met. Even the thinnest layer of plutonium contamination will subject the plastic to approximately the same alpha flux as if plutonium metal were directly in contact with the plastic. The plastic bag in this situation will still weaken and give off hydrogen gas as discussed above.

Since the outer container is not airtight, the plastic bag will tend to expand and contract with changing atmospheric pressure. This may pump hydrogen from inside the bag into the inner container and may draw powdery plutonium corrosion products out of the inner container. As more of the metal oxidizes, more PuO_2 will escape the inner container, accelerating radiolytic destruction of the plastic bag. The free oxygen inside the bag will be depleted and hydride will start to accumulate. As long as the bag is still intact, it would be safe to open the outer container. The danger would be in opening the plastic bag or inner container, exposing the latter's potentially pyrophoric contents to air. The plastic bag would presumably be weak from radiolytic damage, so it may break when handled, creating the same danger.

If the plastic breaks while the package is still in storage, air will enter the inner can and rapidly oxidize any accumulated hydride. This may produce more hydrogen, which would react with any remaining plutonium metal. This phenomenon is characterized by a small initial weight gain, followed by a sudden, larger weight gain after the plastic bag breaks. Rupture of the inner container is possible as oxidation occurs. Rupture of the outer container is much less likely, but still possible. There have been fairly recent examples of both (see Appendix C).

VI. Metal in nested airtight containers with airtight plastic bag between them.

This is also a common storage configuration for metal, particularly at Savannah River and Hanford. As long as neither of the airtight containers leak, and as long as the atmosphere in the inner container is suitable, this method works well for periods of several years. The long-term problem is that the elastomer material used in the airtight seal will gradually be damaged by the radiation. Sooner or later one or both of the containers will start to leak, creating a situation similar to case V above.

It is worst if the inner container leaks first. The metal in the inner container will be exposed to hydrogen and other radiolytic breakdown products of the plastic, while the outer container remains airtight. Hydride will be formed as in equation (1), and free oxygen in the package will be depleted as in equation (2), leading to a situation similar to case I.

VII. Oxide in an unvented container.

PuO_2 is stable chemically but it usually has a high specific surface area (5-50 m^2/gram), and is hygroscopic. Even well-characterized material calcined at 1000°C (as recommended by the draft DOE standard) can generate a significant amount of off-gas. There are four mechanisms:

A. Adsorbed water can react slowly with the oxide to form hydrogen, according to equation (5), where $PuO_{2.2}$ is a binary oxide, $Pu(IV)_{3-x}Pu(VI)_xO_{6-x}$, with x approximately equal to 0.5.[16]

$$PuO_2 + 1/5H_2O = PuO_{2.2} + 1/5H_2 \qquad (5)$$

B. Radiolysis of water or other adsorbed species can liberate gases and pressurize the container.[24]

C. Adsorbed gases can desorb.

D. Helium is created from alpha decay of the plutonium.

There have been many examples of pressurization of unvented oxide containers, including some recent ones (see Appendix C). If an unvented container ruptures, it would spread highly dispersable and respirable plutonium contamination.

VIII. Oxide in contact with plastic.

Oxide will cause radiolytic degradation of any plastic with which in comes into contact. It will probably degrade plastic faster than bulk metal because of the more intimate contact possible with oxide powder. The hydrogen from this source would be added to the gases enumerated above, and would accelerate the pressurization of the container.

IX. Oxide in nested non-airtight cans with airtight plastic bag between them.

As in case V, fluctuations in external atmospheric pressure will cause the atmosphere inside the bag to exchange with the atmosphere inside the inner container, possibly depositing more water on the oxide and carrying more oxide into contact with the plastic bag. Radiolysis of the plastic will eventually break the bag. Plutonium oxide powder would then be relatively free to seep out of the outer container. If gas generation causes a significant pressure increase inside the bag before it breaks, oxide may be even more likely to escape the outer container. Another issue is that atmospheric nitrogen will be adsorbed on the oxide powder, producing NO and NO_2, which could combine with any water present to form corrosive acids. The inner and outer containers may be breached by the corrosion, spreading contamination.

APPENDIX C - Recent Plutonium Storage Incidents

Plutonium storage incidents are not purely hypothetical or purely events of the past. The following is a listing of recent incidents of the general sort relevant to this paper, of which the DNFSB Staff is aware.

March 1994 - plutonium precipitation at SRS

Flaky, plutonium-bearing solids were found in accountability samples taken from two tanks in H-Canyon. Based on preliminary analysis, the solids are believed to be corrosion products from the stainless steel tank walls, to which plutonium ions have adhered. This illustrates the difficulty of controlling solution chemistry over long periods of time well enough to keep plutonium completely in solution.

February 1994 - pressurized cans at SRS

Food-pack cans of plutonium oxide, mixed in some cases with other actinide oxides, were shipped to SRS from Hanford in the mid-1980's. The oxides originated at two commercial plutonium processing facilities that have since shut down. Seven of the cans have pressurized in storage to the point where the cans are visibly swollen. Not all of the cans in this general category have been checked yet (as of March 15, 1994), so more than seven may ultimately be involved. None of the cans have been opened yet and the exact cause of the pressurization is not known.

December 1993 - rapid button oxidation at SRS

Plutonium metal buttons packaged in two nested food-pack cans are regularly weighed to monitor for oxidation. One button, packaged approximately 4 years earlier, had gained enough weight by December 1993 to indicate that 25% of the metal had oxidized, even though there was no visible flaw in the outer can. The can was X-rayed at that point, and it was found that oxide had completely filled the inner can. Without regular weighing and an X-ray capability, there would probably have been a serious contamination incident.

November 1993 - welded container rupture at LANL

Oxidation of plutonium metal ingots ruptured a welded container in TA-55. The ingots had been packaged between five and ten years earlier. The welded container had been wrapped in double plastic bags and placed in an outer slip-lid can. In November 1993, an operator handled the outer can and subsequently discovered plutonium oxide contamination on her clothing. When the outer can was opened, the plastic bags inside were found to be badly deteriorated and the welded inner container was ripped open on one end. The ruptured container was placed in a low-oxygen glove box. After two hours, it started to swell and became warm to the touch. The container was moved to an argon glove box, and the reaction stopped.

July 1993 - line rupture at Rocky Flats

An overhead pipe containing plutonium-contaminated oxalic acid ruptured in Building 771 and sprayed liquid over a radius of 20 feet. Buildup of hydrogen gas pressure from radiolysis of water is believed to have been a factor in the rupture, as was corrosion of the pipe. The spraying occurred in a potentially occupied area, but it happened that no one was present at the time.

March 1993 - plutonium precipitation incident at SRS

Unmonitored evaporation from a process vessel in F-Canyon resulted in precipitation of a plutonium-phosphorus compound. The vessel was not intended for plutonium solution storage, but was left full of solution when F-Canyon ceased operations. Had the vessel agitators been turned on, dispersing the precipitate into a larger slab of water, there could have been a criticality excursion.

March 1993 - drum explosion (uranium) at Livermore

A drum containing metal uranium fuel rods accumulated hydrogen gas, apparently generated by the reaction of uranium with water. When a worker started to remove the lid, there was a low-grade hydrogen explosion, blowing the lid into the air and setting the packing material on fire. The ignition source is unknown.

January 1993 - pyrophoric incident at LANL

A container of actinide metal was opened in a fume hood in TA-55. The metal was directly wrapped in bag-out plastic. When the operator picked up the plastic, it tore apart and there was a flash. Pyrophoric actinide compounds apparently ignited as air entered the bag. No one was injured, but the room was seriously contaminated.

December 1992 - British can rupture incident

A plutonium metal ingot packaged in 1985 ruptured both its inner and outer containers by expansion during oxidation. The inner container was a slip-lid can, the outer container was a food-pack can, and there was a plastic bag between them. The operators were weighing the container regularly and noticed a weight increase in 1990. They did not take action, apparently because they thought the outer container was large enough to accommodate the oxide. The oxidation continued until the outer container ruptured and plutonium oxide was released into the vault atmosphere.

1990 - drum explosions at Fernald (uranium)

There were two separate incidents of 55-gallon drums exploding when hydrogen inside the drums ignited. The hydrogen was probably generated by the reaction of uranium metal with water. In both cases the drums ignited when they were being moved. There was no obvious ignition source; static electricity is the suspected ignition source.

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