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Plutonium Storage Phenomenology

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Westinghouse
Hanford Company Richland, Washington

Management and Operations Contractor for the
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ABSTRACT

Plutonium has been produced, handled, and stored at Department of Energy (DOE) facilities since the 1940s. Many changes have occurred during the last 40 years in the sources, production demands, and end uses of plutonium. These have resulted in corresponding changes in the isotopic composition as well as the chemical and physical forms of the processed and stored plutonium. Thousands of ordinary food pack tin cans have been used successfully for many years to handle and store plutonium. Other containers have been used with equal success. This paper addresses the chemical and physical forms of plutonium in storage and presents examples of the norm and exceptions to this satisfactory experience. To aid in understanding the challenges of handling plutonium for storage or immobilization the lessons learned from past storage experience and the necessary countermeasures to improve storage performance are discussed.

HISTORY OF PLUTONIUM PROCESSING AND STORAGE AT THE DOE SITES

The original mission of the DOE plutonium sites was to supply plutonium metal for national defense. Plutonium was produced by uranium irradiation in nuclear reactors and irradiated fuel elements were processed in chemical separations facilities at Hanford and Savannah River to separate the plutonium from fission products and remaining uranium. The plutonium portion or product of the separation facility was a nitric acid solution of plutonium. Direct pipe transfer or heavy-wall stainless steel containers were used to handle and transport this solution to other facilities where the plutonium solution was converted to metallic plutonium. Urgent demands for large quantities of plutonium metal during the 1940s and 1950s limited amount and time plutonium was held in storage. Food pack type tin cans and other containers saw relatively short-term (i.e., weeks or months) use as a package for plutonium transport and short term storage of metal product. Production residues stored in such containers, however, were typically stored for longer periods.

In the 1960s, plutonium processing started to change in several ways. Demands for plutonium metal declined while research in power reactor technology indicated that plutonium in the form of plutonium dioxide could be used in place of fissionable ^{235}U in power reactor fuel. Later, breeder reactor research showed how to utilize plutonium oxide as a fuel for breeder reactors. A new product form, a fine powdery oxide, joined the traditional product form of a single metallic piece weighing about 2 kilograms. Demands for the oxide powder form increased from grams to kilograms to thousands of kilograms. In the late 1960s and 1970s the Hanford plutonium facilities provided almost all of the plutonium oxide used in this country. This included thousands of kilograms for several Fast Flux Test Facility (FFTF) cores.

The declining demand for metallic plutonium initiated interim and finally long-term storage of the metal. Some metal has been in storage over 25 years. Production strategies for the oxide dictated

scheduling production between metal production campaigns and a degree of overproduction to allow for process losses, out of specification product, and ability to fulfill unanticipated requests for kilogram quantities of plutonium oxide for research efforts. Storage of oxide product prior to shipment as well as production overrun material became common.

In the 1960s the Hanford plutonium facility was provided with plutonium scrap recovery capability. Plutonium scrap from military application research and virtually all from commercial and breeder reactor research was returned to Hanford for recovery of the plutonium. This scrap, which had a wide variety of chemical and physical forms, was added to the growing inventory of plutonium metal and oxide being stored at Hanford and other sites. Cans stored soon numbered in the thousands. Radiation levels and ambient temperatures in storage vaults increased.

Vaults such as the typical pedestal storage vault shown in Figure 1 were used to store these cans. Fundamentally, it is an open room with floor-to-ceiling vertical pedestals to which steel storage cups have been welded. Cans of plutonium are placed in the cups for storage. A large storage vault contains over a thousand storage positions. A later version of pedestal storage incorporated concrete shielding around small groups of pedestals for radiation shielding of personnel is shown in Figure 2.

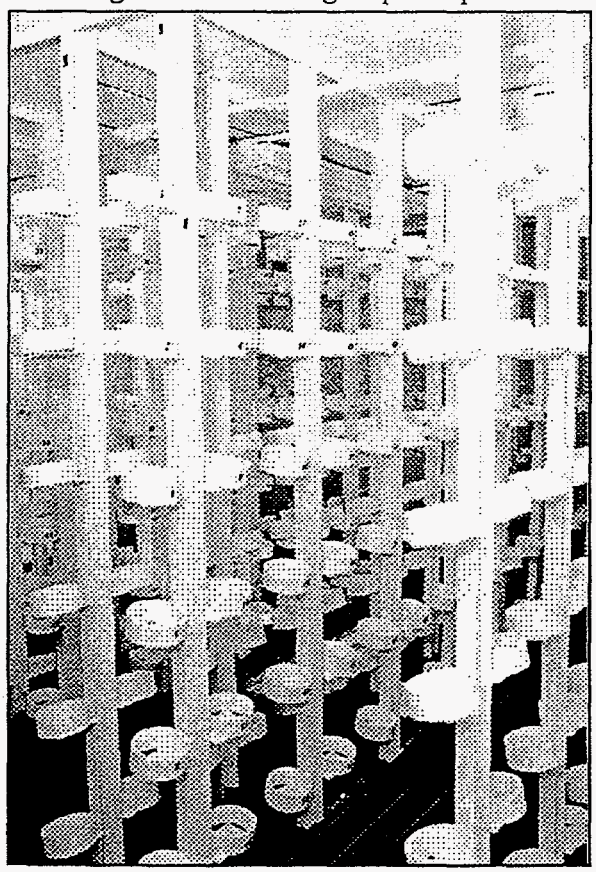

Figure 1 Open room storage vault contains hundreds of storage positions for food pack type cans. Personnel entering this room are directly exposed to radiation from every can.

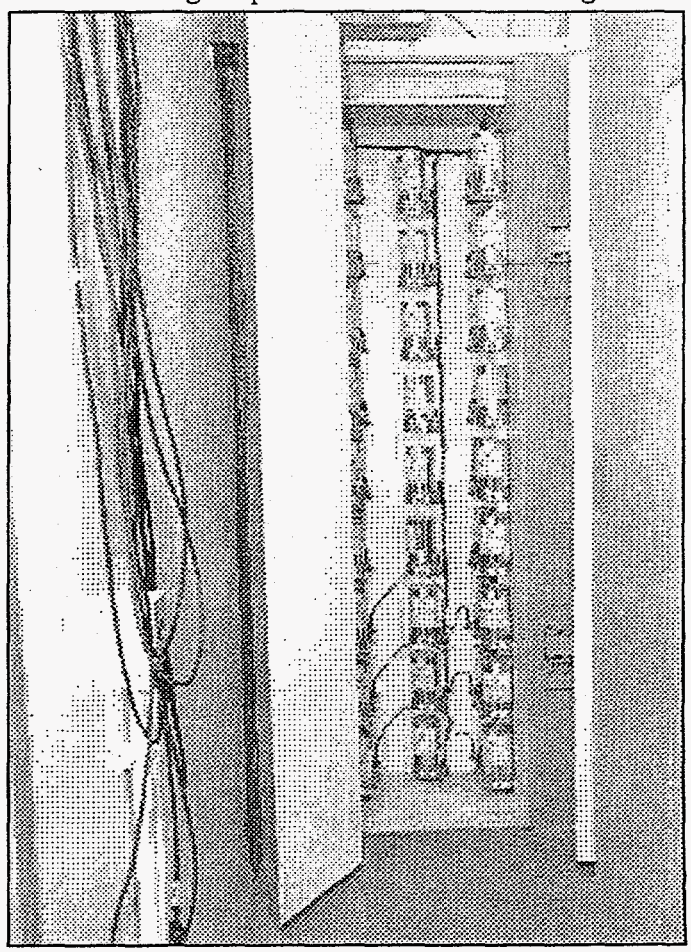

Figure 2 Newer storage vault design utilizes concrete cubicles with concrete shielding doors to reduce personnel radiation exposures.

During the 1960s and 1970s other changes were taking place which would ultimately impact the plutonium storage. The isotopic composition of much of the plutonium changed. The declining demand for plutonium metal caused N Reactor production to be changed from nominal 6% ^{240}Pu weapons-grade plutonium to nominal 12% ^{240}Pu. The 12% ^{240}Pu material as used for breeder reactor research and in the FFTF cores. Commercial nuclear reactors and some government experimental reactors produced plutonium with higher ^{240}Pu content which was commonly referred to as fuels-grade plutonium. Much of this material has been stored in Hanford plutonium storage vaults as metal, oxide, or scrap. Since the late 1980s many of the defense related plutonium activities in the DOE have been shut down and the number of nuclear warheads reduced. This has resulted in a buildup of large inventories of excess weapons grade plutonium metal and other forms at Rocky Flats, Pantex and to lesser degrees at other sites.

PLUTONIUM CHARACTERISTICS

Plutonium is a mixture of isotopes with varying important physical properties (i.e,, those relating to storage). The exact mixture of isotopes produced is the result of many complex physical characteristics of nuclear reactor hardware and operation as well as fuel cycle operations. Reactor power level, length of fuel exposure, neutron flux energy spectrum and other factors influence the production rates of the individual plutonium isotopes. Table 1 shows some plutonium isotopic distributions which may be considered typical and illustrates the wide distribution of plutonium isotopes.

TABLE 1. Plutonium Isotopic Mixtures.

Plutonium Type	Weight Percent				
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Weapons Grade	0.004	92.84	6.48	0.61	0.23
12 %	0.04	86.20	11.80	1.70	0.15
Fuels Grade	1.40	70.12	15.67	10.70	2.10
High Exposure or Reactor Grade	0.57	65.08	23.36	7.66	3.34

The mixtures shown in Table 1 are listed in order of increasing ^{240}Pu content. The ^{240}Pu content is the usual way that the general pedigree of the plutonium is characterized. Technically, no precise line separates the types although certain ^{240}Pu levels have been administratively designated as cut-off points.

Table 2 presents properties of the individual isotopes that are of interest in plutonium handling and storage.

TABLE 2. Properties of Plutonium Isotopes.

Property	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Half-life (years)	89.6	24,000	6,600	13.2	380,000
Principal means of decay	Alpha	Alpha	Spontaneous Fission	Beta	Spontaneous Fission
Significant daughters of decay	None	None	None	^{241}Am	None
Curies per gram	17.0	0.061	0.22	112	0.004
Decay heat (watts/gram)	0.56	0.002	0.007	0.003	0.001

Except for ^{242}Pu, each isotope has some attribute that is of concern in plutonium handling and storage:

- ^{238}Pu--high decay heat (0.56 watts/gram) must be dissipated
- ^{239}Pu--fissionable material requires nuclear criticality prevention considerations
- ^{240}Pu--relatively high spontaneous fission rate results in neutron exposures
- ^{241}Pu--short half-life decay to ^{241}Am which has high decay heat (0.12 watts/gram) and high gamma radiation.

In addition to the nuclear properties outlined, the physical and chemical forms of plutonium have characteristics that also affect plutonium handling and storage. Based on experience gained in successfully storing ton quantities of various forms of plutonium for decades and the lessons learned from the relatively few incidents of less than adequate storage conditions, the behavior of plutonium in storage is predictable. The following discussion and illustrations are intended to display the breadth of plutonium characteristics that must be dealt with whether the plutonium will be stored, stabilized or immobilized.

PLUTONIUM METAL

- Very high density (about 19.4 g/cc) material.
- Chemically stable in most finished product forms.
- Somewhat pyrophoric if impurity content or decay heat is high. Can combine with oxygen at ambient temperature to form plutonium oxide.
- When reacted with hydrogen or carbon, unstable compounds can result.

Figure 3 Two kilogram plutonium metal button shown shortly after being produced in 1988.

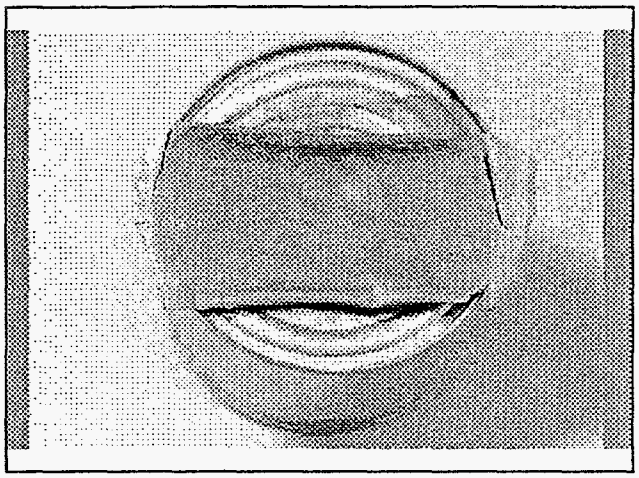

Figure 4 High decay heat of fuels grade plutonium can result in high temperatures as evidenced by this charred label.

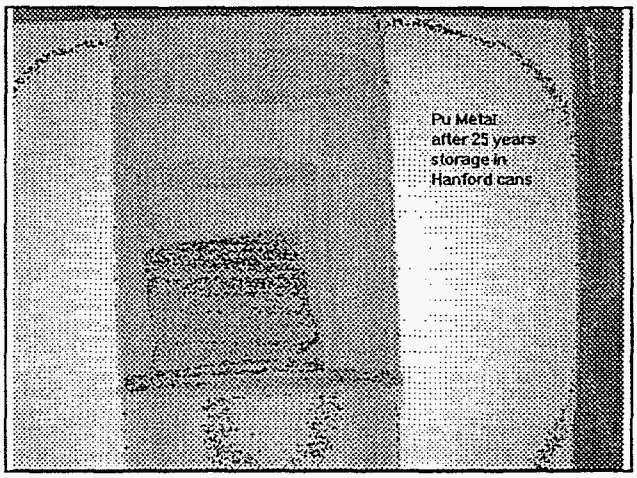

Figure 5 Radiograph of 2 kilogram metal ingot stored for 25 years in sealed food pack cans shows little indication of corrosion.

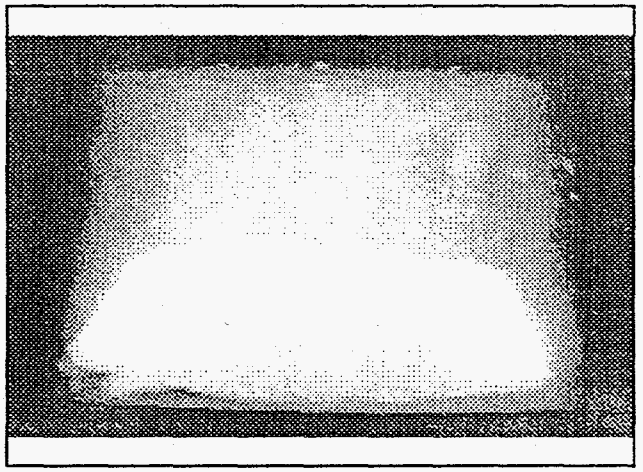

Figure 6 Radiograph shows inner can filled with oxide from corrosion of this metal piece. Note irregularity of top surface of metal.

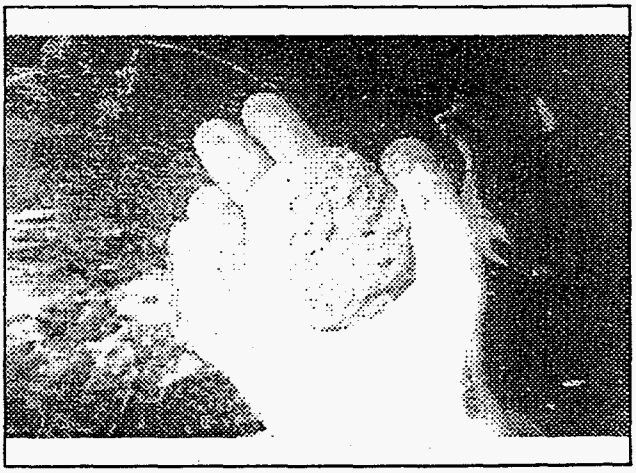

Figure 7 Metal piece removed from can shown in Figure 6. The cans were not properly sealed allowing air to enter and oxidize nearly 25% of the metal in 4 years.

PLUTONIUM OXIDE

- Density typically about 2 gm/cc (theoretical 10.3 g/cc).
- Dispersibility spans a wide range from fine powders to pressed pellets
- Hygroscopic.
- Impurities may result in generation of gases due to radiolysis.

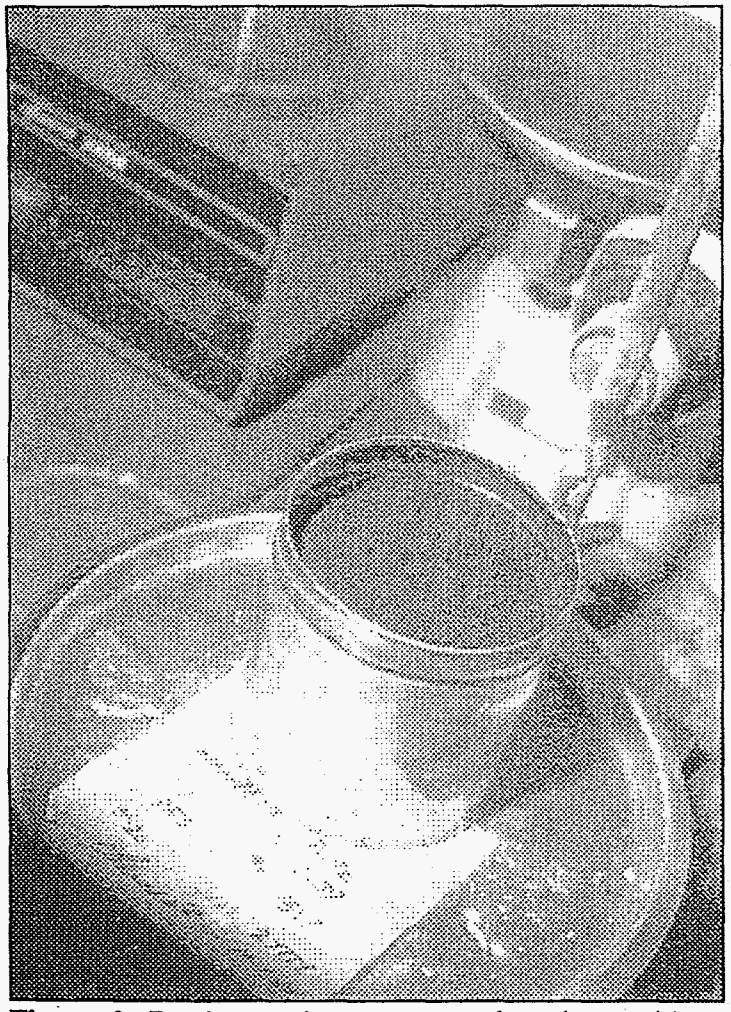

Figure 8 Product and some scrap plutonium oxide is typically a fine dry appearing powder. Hundreds of cans of product oxide produced at Hanford and other sites have been stored for over a decade without incident.

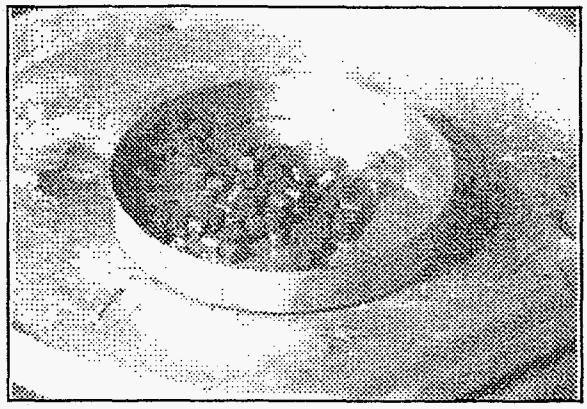

Figure 9 Some oxide is in the form of pellets of varying quality depending on previous fabrication or testing steps.

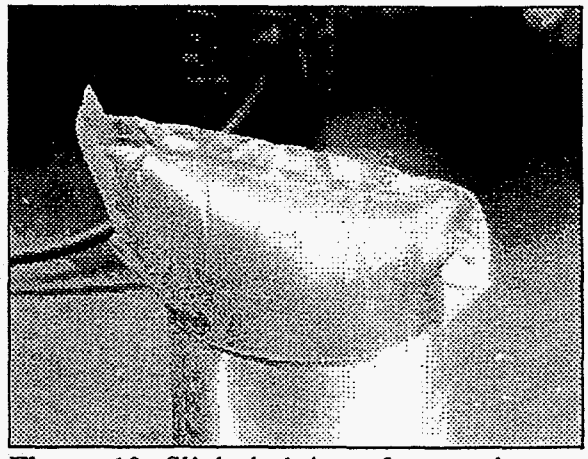

Figure 10 Slight bulging of can end plates indicated internal pressure of about 3 psig prompting this can to be opened. After several years in storage the plastic bag still holds pressure.

PLUTONIUM SCRAP AND RESIDUES

- Properties span the range of plutonium metal and oxide identified above.
- May include foreign matter which may be chemically inert or chemically reactive. This can lead to container corrosion or pressurization.
- Exact chemical composition or impurities sometimes not known.
- Chemical reactivity of scrap can cause the material to be pyrophoric or release flammable or explosive gases, such as hydrogen and oxygen.

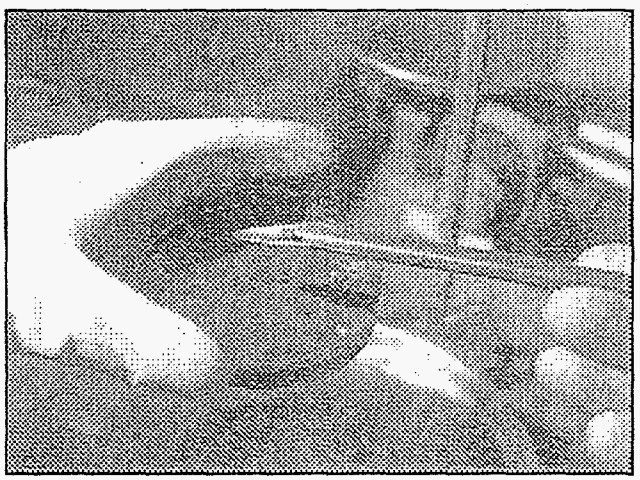

Figure 11 Some oxide scrap may contain powders, pellets or other physical forms. In this container pellets were found in the powder oxide.

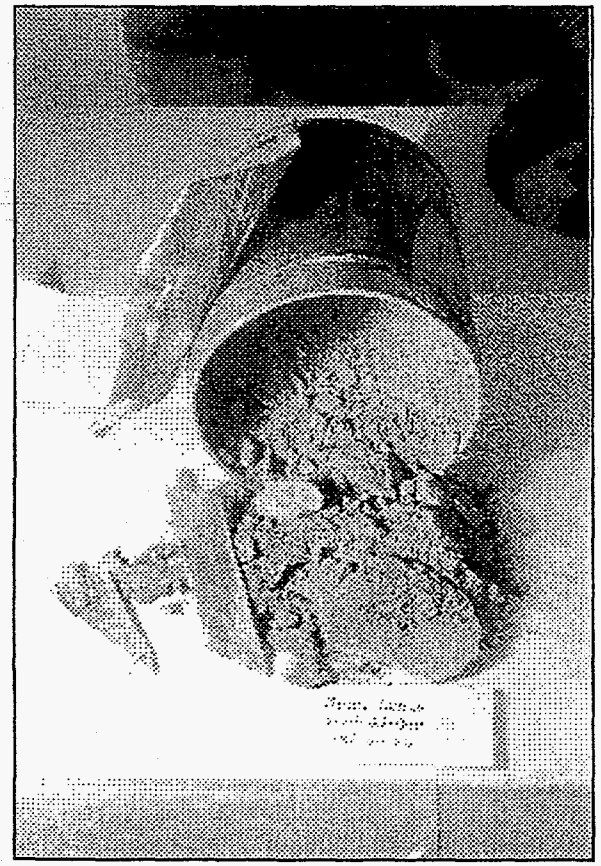

Figure 13 Tools, in this case a broken hacksaw blade, used in gloveboxes sometimes are found in residue containers.

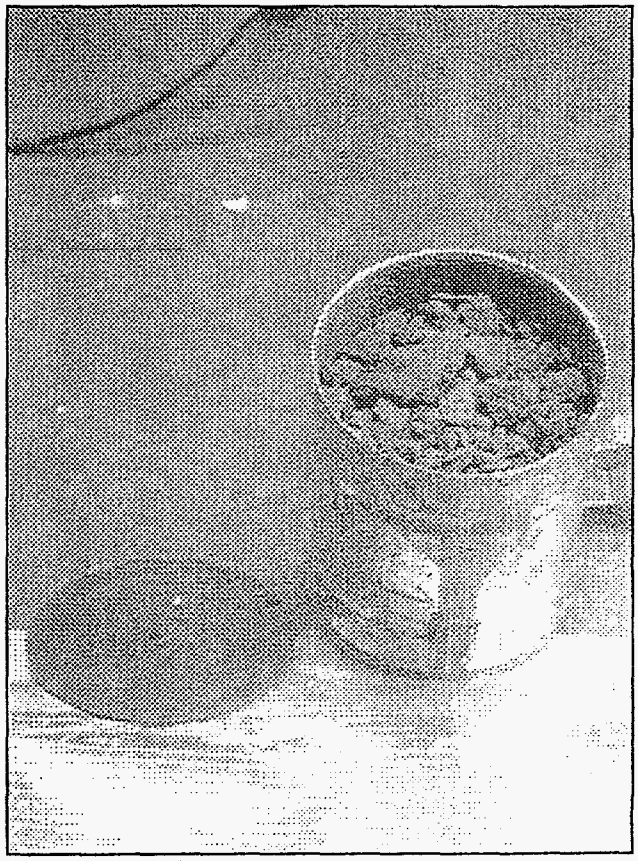

Figure 12 The phyical form of residues may be clumps of a wide range of sizes and brittleness.

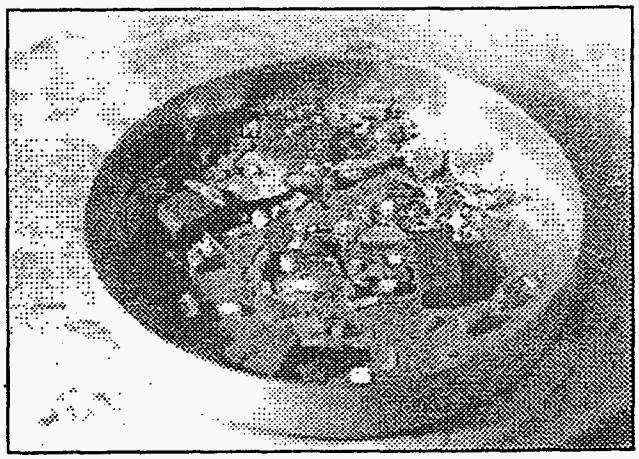

Figure 14 Non plutonium materials are sometimes found in glovebox residues. This material was screened from about a kilogram of oxide powder.

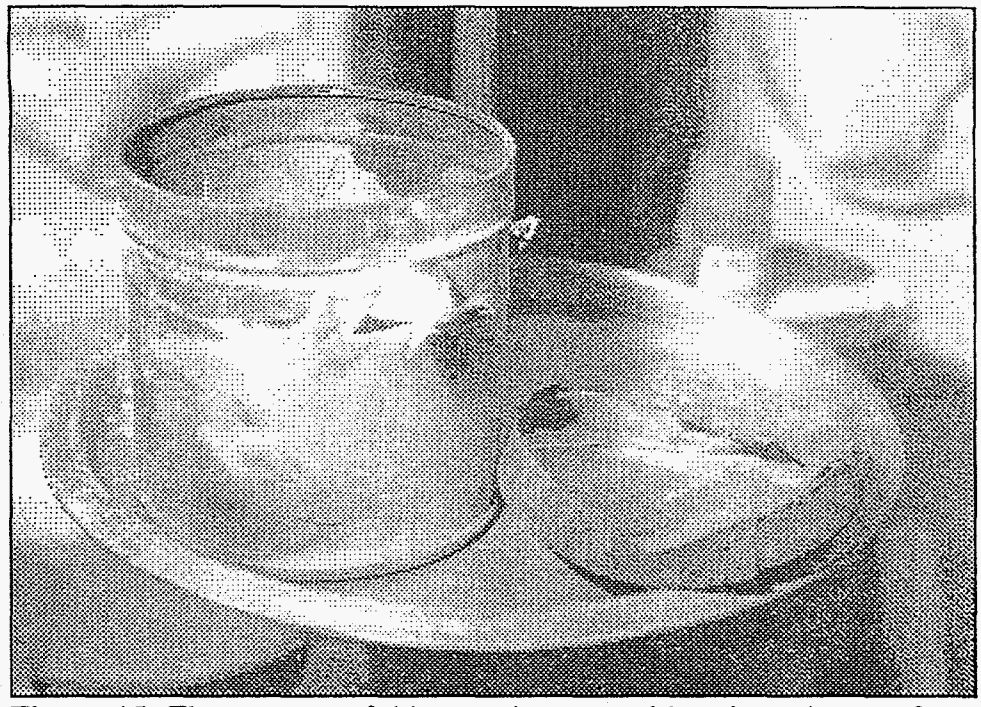

Figure 15 The contents of this container started burning minutes after it was repackaged and removed from a glovebox in 1980.

Figure 16 This burned bag was the result of spontaneous ignition of residue materials that occurred minutes after the can containing the plutonium residues was bagged out of a glovebox in 1980.

LESSONS LEARNED AND CORRECTIVE ACTIONS

Table 3 is a summary of the lessons learned from unsatisfactory storage or packaging conditions. Implementation of the corrective actions have been successful in precluding similar occurrences.

TABLE 3. Summary of Lessons Learned and Corrective Actions

Lessons Learned	Corrective Action
Size increase of oxidizing plutonium metal can be sufficient to rupture cans.	Plutonium metal overpacked in can large enough to accommodate complete oxidation of largest metal piece handled.
Personnel can become contaminated by walking into a vault where a can has ruptured.	Storage vaults equipped with continuous air monitors.
Poor can seals will allow oxidation of plutonium metal to continue to completion.	Can seal inspection procedure and canning machine preventative maintenance implemented.
When plutonium metal oxidizes, inner containers can rupture without an outer container appearance change.	Periodic weighing instituted to detect oxidation.
If properly sealed, tin cans will bulge when pressure builds up.	Straight-edge bulge test instituted to detect pressurizing containers well in advance of dangerous can deformation.
Plutonium oxide containing moisture or impurities can pressurize cans to the point of rupture.	Glovebox storage mandated for all materials suspected of being capable of causing pressurization. Thermal stabilization and weight loss on ignition testing criteria established.
High decay heat can degrade plastic bags and can seals allowing oxygen to enter to sustain oxidation of plutonium metal.	Limits established on decay heat of plutonium stored in tin cans. Plastic bags are not relied on to exclude oxygen.
Misidentification of material can allow storage in cans incompatible with the material.	Rigid material identification requirements have been imposed. Monthly visual container inspection program instituted.

DISCLAIMER

The views expressed in this paper are those of the author and do not necessarily reflect any biases, proposed action, or decisions of the United States Government or any agency thereof.

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