2.3 IMMOBILIZATION ALTERNATIVES

A total of six immobilization variants covering the three reasonable alternatives addressed in the PEIS are described in this section: vitrification, ceramic immobilization, and electrometallurgical treatment.

The following assumptions apply for the immobilization alternatives:

- The operational campaign of the immobilization facility will take no more that 10 years to complete.
- The nominal feed of plutonium to the facility is 50 metric tons. Nominal throughput is therefore 25 kg plutonium per day for 200 days of operation per year for ten years.
- Design for criticality safety will meet applicable DOE Orders and available NRC regulatory guides. Criticality is prevented by using batch mass control or equipment geometry as the preferred methods in the design. The use of appropriate neutron absorbers (e.g., gadolinium, samarium, or hafnium) has been assumed.
- The waste canister assumed for this study shall not exceed a 0.6 meter in diameter by 3.0 meter long cylindrical canister.
- The immobilized plutonium package will contain an added radiation field to increase proliferation resistance. The gamma radiation field will be greater than 100 R/hr at 1 meter from the package surface 30 years after initial fabrication.

The Immobilization Alternative team analyzed the variants described in Table 2-4 with a summary of the results shown in Table 2-5.

Table 2-4. *Immobilization Category Variants*

Variants	*Description*[1]
Vitrification Greenfield	• Combined plutonium processing and glass melter facility • A two step vitrification process • Plutonium immobilized in borosilicate glass with ^{137}Cs radiation barrier
Vitrification Can-in-Canister	• Existing facility on DOE site used for plutonium conversion and glass melter facility • Plutonium immobilized in glass in small cans; cans placed in DWPF canister with HLW as radiation barrier • Canister filling done at DWPF
Vitrification Adjunct Melter	• A two step vitrification process • Plutonium is first dissolved in glass frit in the plutonium processing plant in existing facility on a DOE site • New adjunct melter adjacent to DWPF as second stage melter • Final ^{137}Cs supernate from HLW at DWPF used as radiation source
Ceramic Greenfield	• Combined plutonium processing and ceramic immobilization facility • Plutonium immobilized in ceramic matrix with ^{137}Cs radiation barrier • Ceramic forms placed in canisters and backfilled with TiO_2
Ceramic Can-in-Canister	• Existing facility on DOE site used for plutonium conversion and ceramic immobilization facility • Plutonium immobilized in ceramic matrix in small cans; cans placed in DWPF canister with HLW as radiation barrier • Canister filling done at DWPF
Electrometallurgical Treatment	• Plutonium processing and immobilization performed at ANL-W using modified or new facilities • Immobilized form is in glass-bonded zeolite matrix with ^{137}Cs radiation barrier • Glass-bonded zeolite placed in canister and backfilled with appropriate filler

[1] Final formulation for plutonium-loaded immobilized forms is to be determined. All immobilized forms will be designed to be acceptable to the high-level waste repository.

Table 2-5. ***Summary of Characteristics of Immobilization Variants***

Alternative	Variant	Number of canisters to repository[1] Total/Incremental	Plutonium concentration in waste forms wt. %	Effective plutonium concentration in canister wt. %
Vitrification	New Greenfield Facility	600/600	5	5
Vitrification	Can-in-Canister	1000/200	10	3
Vitrification	Adjunct Melter	600/600	5	5
Ceramic	New Greenfield Facility	640/640	12	12
Ceramic	Can-in-Canister	1000/200	12	3
Electrometallurgical Treatment	None	960/960	5	5

[1] The total number of canisters is the total number of canisters containing surplus plutonium. The incremental number is the number of additional canisters required for plutonium disposition beyond requirements for ongoing DWPF operations. For can-in-canister variants, the incremental number of canisters results from small cans displacing approximately twenty percent of the volume inside the DWPF canisters already planned for the high-level waste program.

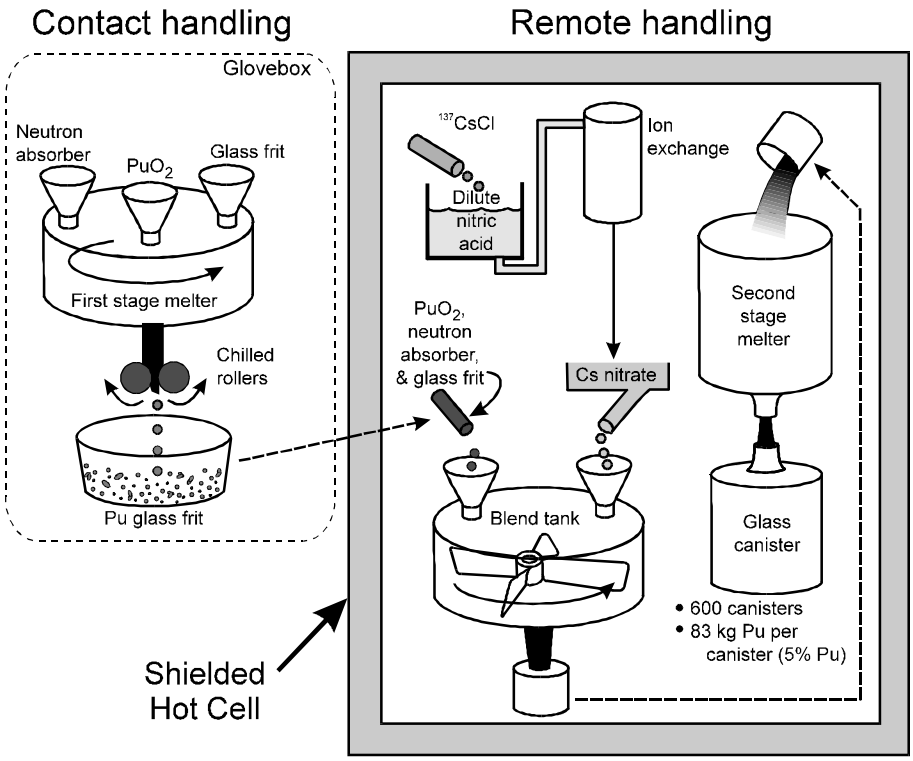
2.3.1 Vitrification Alternative

In several countries including the United States, radioactive high-level waste is being incorporated into molten glass in a process known as vitrification, producing highly radioactive glass "logs" to be stored for an interim period and then disposed. Vitrification has also been identified as a promising technology for the disposition of surplus weapons-usable plutonium, although such a process has not been demonstrated on an industrial scale. The immobilization technology considered here is to vitrify plutonium in borosilicate glass to produce a final waste form suitable for disposal in a high-level waste repository. Borosilicate glass has been chosen as the waste form for high-level waste in the western world because it combines high radioactive impurity solubility and high tolerance to high-level waste variability with excellent leach resistance and high thermal and radiation stability. Borosilicate glass can also be produced at temperatures 200^{o} C below more conventional glasses, thus minimizing volatility of fission products (e.g., ^{137}Cs) and dose to workers.

2.3.1.1 Vitrification Greenfield Variant

The Vitrification greenfield variant immobilizes plutonium through a two-step vitrification process in a borosilicate glass with ^{137}Cs uniformly distributed in the glass matrix to produce a radiation field in the final product ("internal radiation barrier"). The vitrification greenfield variant is shown conceptually in Figure 2-4.

Figure 2-4. ***Vitrification Greenfield Variant***



The plutonium feed materials to the vitrification facility will be plutonium oxide. This oxide is fed to a first stage melter which incorporates the plutonium in a borosilicate glass frit as shown on the left side of Figure 2-4. The first stage melter is located in the contact-handled portion of the facility. The frit will subsequently be blended with ^{137}Cs, with a neutron absorber, and with additional glass frit and fed to a second stage melter as shown on the right side of Figure 2-4. The ^{137}Cs is from 54 million curies of CsCl capsules at Hanford. The molten glass from the second stage melter containing the plutonium and the ^{137}Cs will be poured into a canister that will subsequently be welded closed, decontaminated, and stored onsite pending permanent disposal at a high-level waste repository.

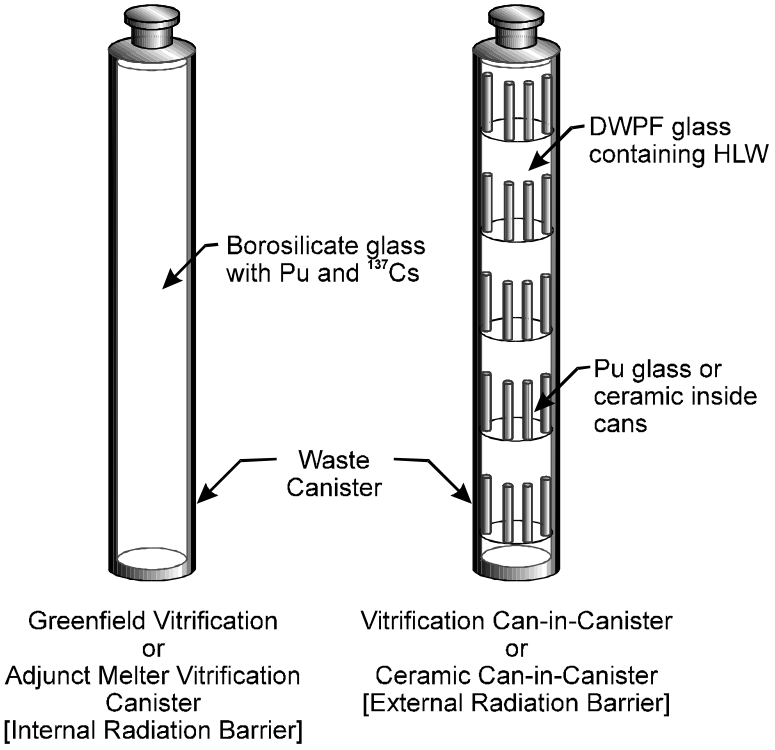
The plutonium loading in the borosilicate glass is a design parameter involving multiple tradeoffs that will be optimized based upon research, testing, and repository criticality analysis during later phases of the design. The final design loading selected will consider fission product availability as well as form quality, facility size, safety factors, and high-level waste acceptance criteria. For this early design phase, 5% (by weight) plutonium loading has been assumed. Lower plutonium loadings would increase the number of canisters going to a repository while higher loadings would reduce conservatism in safety assessments.

The facility is assumed to be constructed and operated on a generic site. After actual site selection, more specific site-related information will be evaluated.

2.3.1.2 Vitrification Can-in-Canister Variant

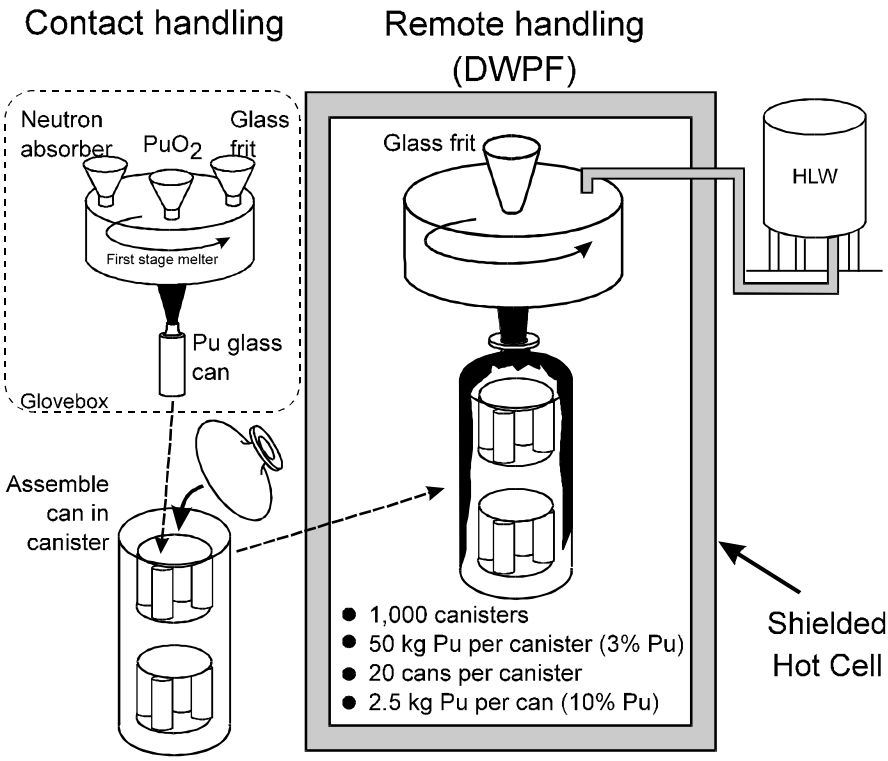
The Vitrification Can-in-Canister variant immobilizes plutonium in borosilicate glass in individual cans and utilizes high-level waste (HLW) glass produced at DWPF to provide an external radiation barrier for proliferation resistance. Molten plutonium glass is poured into small stainless-steel cans. The cans are then loaded onto a frame and placed inside an empty stainless-steel DWPF canister which had the top/neck removed. The canister is then assembled and transferred to the DWPF facility where molten HLW glass is poured around the small cans. After the filled canisters are decontaminated and welded closed, they will be stored onsite until they are sent for final disposal at a high-level waste repository. Figure 2-5 shows a comparison of the canisters. Figure 2-6 shows a schematic representation of the vitrification can-in-canister variant.

Figure 2-5. ***Canister Comparisons***



The plutonium loading in the borosilicate glass is a design parameter involving multiple tradeoffs that will be optimized based upon research, testing, and repository criticality analysis during later phases of the design. The selection of the final design loading will consider radionuclide availability as well as form quality, facility size, safety factors, and repository waste acceptance criteria. For this early design, a plutonium loading of 10% (by weight) within the small can has been assumed. There will be about 20 such cans per canister. This results in an average plutonium concentration of about 3% of the weight of the glass in the larger canister.

Figure 2-6. ***Vitrification Can-in-Canister Variant***

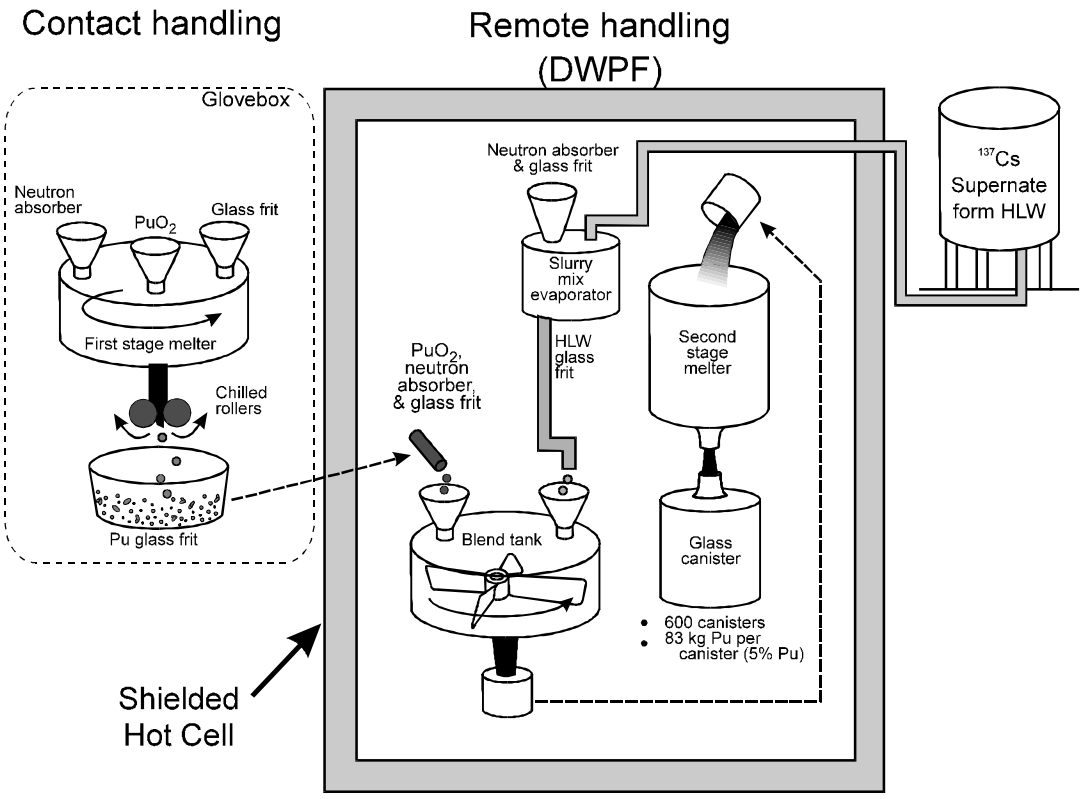


2.3.1.3 Vitrification Adjunct Melter Variant

The Vitrification-Adjunct Melter to DWPF variant is similar to the Vitrification greenfield variant, except this immobilization variant uses the existing facility at the SRS in conjunction with a new adjunct melter built next to DWPF. Figure 2-7 shows a schematic of the vitrification adjunct melter variant.

Plutonium oxide will be fed to vitrification equipment also located in existing facility on DOE site to produce glass frit containing plutonium. This glass frit will then be sent to the new Adjunct Melter facility adjacent to DWPF where it will be mixed with ^{137}Cs from the SRS tank farms, then melted in a second stage melter. The molten glass containing the plutonium and ^{137}Cs will be poured into 0.6 meter diameter x 3.0 meter high stainless steel canisters, which will subsequently be welded closed, decontaminated, and stored onsite until sent to final disposal at a high-level waste repository.

Figure 2-7. *Vitrification Adjunct Melter Variant*



2.3.2 Ceramic Alternative

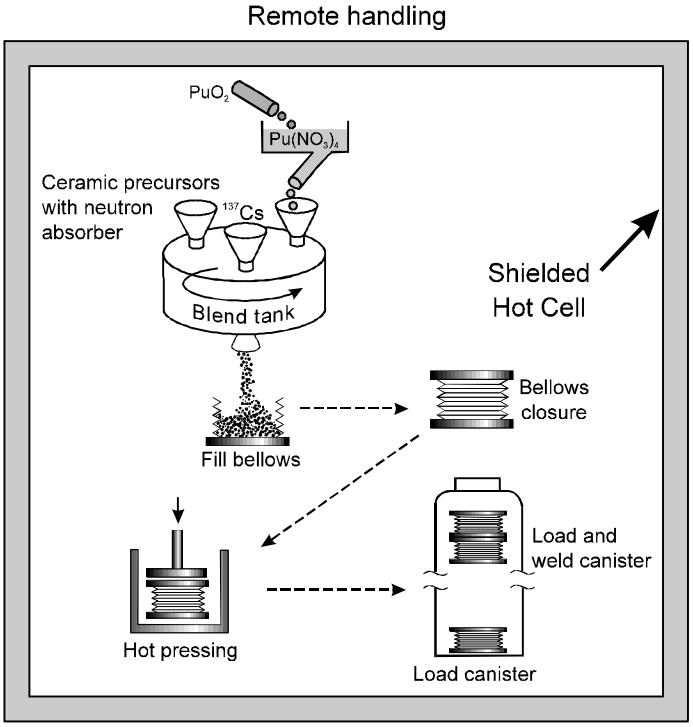
Since the late 1970s, various ceramic waste forms have been considered for immobilization of high-level waste; however, no industrial experience exists for high-level ceramic waste forms unlike borosilicate glass forms. The ceramic waste form is attractive for immobilization purposes because of its extremely low leachability, existence of natural mineral analogues that have demonstrated actinide immobilization over geologic time scales, and the high solid solubility of actinides in the ceramic resulting in a reasonable overall waste volume. Ceramic immobilization of simulated high-level waste in a Synthetic Rock (SYNROC) material has been demonstrated at full scale at the Australian Nuclear Science and Technology Organisation (ANSTO). Small scale samples have been made with greater than 10% plutonium. Although immobilization in ceramic has not replaced various existing and planned vitrification facilities for high-level waste, a considerable amount of research and development has been performed, particularly with higher mass plutonium isotopes and higher actinides procured from reactor recycled plutonium.

2.3.2.1 Ceramic Greenfield Variant

The Ceramic greenfield variant accepts plutonium oxide and, through a ceramic immobilization process, converts the plutonium into a form that can be disposed of in a high-level waste repository. Plutonium is immobilized in a titanate-based ceramic with ^{137}Cs

spiking to produce a radiation field that is uniformly distributed in the waste form. Figure 2-8 shows the greenfield ceramic variant.

Figure 2-8. ***Ceramic Greenfield Variant (dry feed)***



The plutonium feed materials to the ceramic fabrication facility will be plutonium oxide. The plutonium oxide is converted to plutonium nitrate and blended with ceramic precursors, neutron absorbers, and a cesium loaded titanate. The mixture is then calcined (heated), loaded into bellows, and hot pressed to produce a densified product. Twenty hot pressed bellows, 30 cm diameter, will be loaded into a canister, 36 cm diameter by 2.4 m long, with TiO_2 granules that are used as a packing material. The canisters are then stored onsite until they can be transferred to the high-level waste repository for disposal.

Additional assumptions for the variant are:

- The plutonium loading in the ceramic form is assumed to be 12% (by weight). This parameter is taken from demonstrated fabrication sizes (~33 kg using actinides), typical plutonium limits in glove box processing (~4 kg plutonium), and known plutonium loading data in ceramics (>12%). The final plutonium loading selected will consider form quality, facility size, safety factors, high-level waste repository acceptance criteria, and other considerations.

- The final ceramic product is contained in canisters and is stored onsite until it is transported to a high-level waste repository. Each product canister contains 20 compressed bellows with about 660 kg of ceramic, which includes approximately 80 kg of plutonium.

The ceramic product is assumed to be similar to Synroc-C which contains the mineral phases zirconolite ($CaZrTi_2O_7$), hollandite ($BaAl_2Ti_6O_{16}$), perovskite ($CaTiO_3$), and rutile (TiO_2). The actual phases selected will be the result of a research program, and it is assumed that the composition of the ceramic-forming chemicals (precursors) will not affect the processing equipment or sequence.

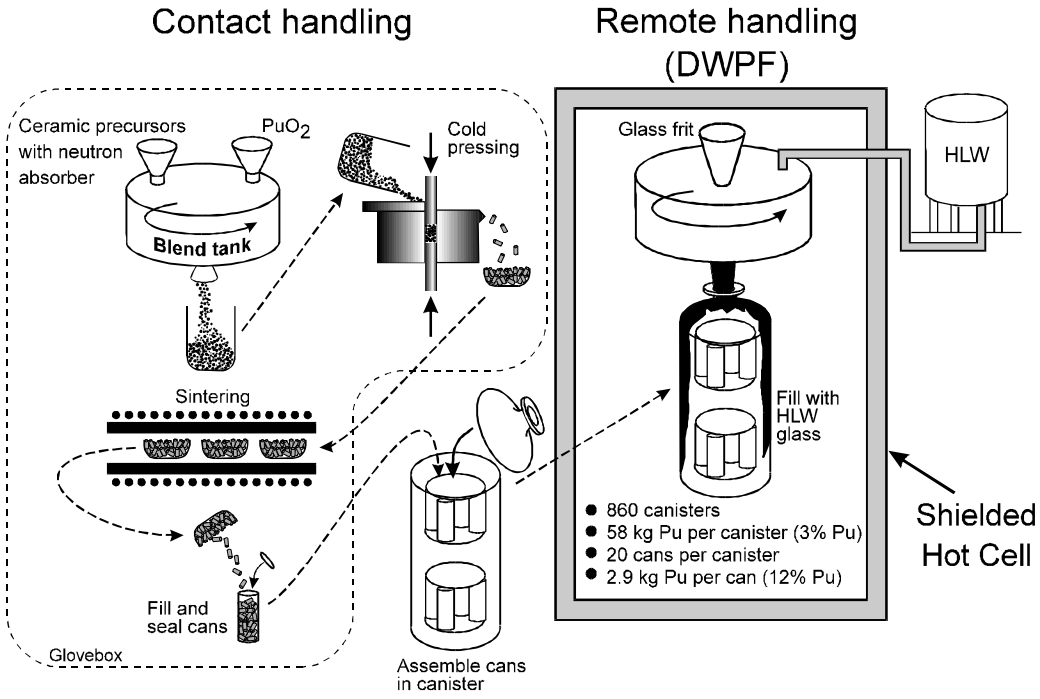
The facility is assumed to be constructed and operated on a generic site. After actual site selection, more specific site-related information will be required.

2.3.2.2 Ceramic Can-in-Canister Variant

This variant is analogous to the vitrification can-in-canister variant. The difference is that the plutonium that is inside the can is immobilized in a ceramic form, rather than a glass form. The ceramic product for this variant is formed using a cold press and sintering process, rather than the hot press process in the greenfield ceramic variant. Plutonium oxide is blended with ceramic precursors and neutron absorbers. This mixture is calcined, cold pressed and sintered to produce the densified product to be loaded into small cans. Figure 2-9 shows the ceramic can-in-canister variant.

Major advantages of the cold press and sinter option are increased throughput, simplicity, and proven production experience in the MOX fuel industry. An automated cold press can process 12.2 kg of plutonium an hour. Cold "pressing" is an option for the ceramic can-in-canister variant because the volatility of fission products (specifically ^{137}Cs) in the sintering process is not an issue.

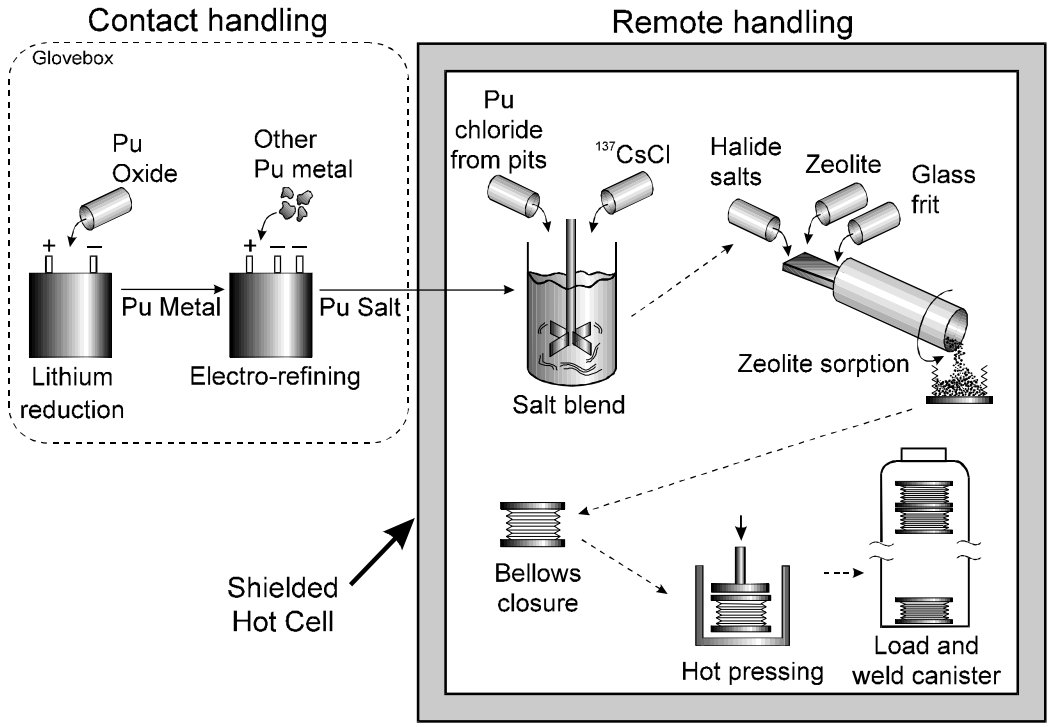
Figure 2-9. ***Ceramic Can-in-Canister Variant***



2.3.3 Electrometallurgical Treatment Alternative

In the Electrometallurgical Treatment Alternative, plutonium metal and oxide are converted to chlorides, dissolved in a molten salt solution, sorbed on zeolites, and then immobilized in a glass-bonded zeolite (GBZ) waste form. The immobilization operations will be integrated with operations in the ANL-West hot cells to treat DOE-owned spent fuels. The fission products from these fuels will contribute some radiation to the immobilization forms, but ^{137}Cs from the Hanford capsules will provide most of the radiation field to create a radiation barrier. Figure 2-10 shows the Electrometallurgical Treatment variant.

Figure 2-10. ***Electrometallurgical Treatment Alternative***



Feed to the immobilization operations will be plutonium metal, oxides, and chlorides. Oxide feeds will be converted to metals in a lithium reduction step and then sent, along with metal feeds, to an electrorefining cell. The electrorefiner converts plutonium metal to chloride using an anodic dissolution process. Metal from pits would be converted to chlorides directly in front-end processing using a hydride/chloride process; the ARIES process would have to be modified to accommodate the conversion to chloride. Plutonium chlorides from the electrorefiner and front-end processing are blended with salt to which CsCl is added to provide the radiation barrier. The blended salt is sorbed onto zeolite, and the zeolite is mixed with a suitable glass frit and hot pressed to make the monolithic mineral form (GBZ). The GBZ forms are loaded into canisters and stored onsite until they can be transferred to a high-level waste repository for disposal.