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**Office of  
Fissile Materials  
Disposition**

United States Department of Energy

**Technical Summary  
Report  
For  
Surplus Weapons-Usable  
Plutonium Disposition**

October 31, 1996

Rev. 1

## EXECUTIVE SUMMARY

### ES.1 INTRODUCTION

The United States has declared 38.2 metric tons of weapons-grade plutonium surplus to national security needs.<sup>1</sup> Additional inventories of plutonium are expected to bring the total amount of plutonium that is surplus to approximately 50 metric tons.

To establish a framework for selecting plutonium disposition options which would achieve a high degree of proliferation resistance, the National Academy of Sciences (NAS) reviewed a number of options and concluded that the national objective should be to make the surplus "plutonium roughly as inaccessible for weapons use as the much larger and growing quantity of plutonium that exists in spent fuel from commercial reactors,"<sup>2</sup> a state the NAS defined as the *spent fuel standard*. The Department of Energy (DOE) has enhanced this statement to read:

*DOE Spent Fuel Standard*

A concept to make the plutonium as unattractive and inaccessible for retrieval and weapons use as the residual plutonium in the spent fuel from commercial reactors.

The DOE enhancement makes explicit the concept of material attractiveness which was implicit in the NAS usage of the term. The spent fuel standard is not a specification-type standard. It encompasses a range of barriers which deter accessibility to and use of plutonium, including such barriers as a radiation field, dilution, inaccessible location, and size and weight. In the aggregate, these barriers achieve a degree of inaccessibility and a difficulty of extraction of the plutonium comparable to that of plutonium in "typical" commercial spent fuel. Once having achieved the spent fuel standard, the formerly weapons-usable plutonium is rendered no more attractive for use in nuclear weapons than the much larger and growing inventory of plutonium in commercial spent fuel.

Building on the NAS work, the DOE completed a *screening process* in March 1995<sup>3</sup> in which a large set of proposed, conceptual options for the disposition of plutonium were evaluated. The options that remained after the screening process were identified as

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<sup>1</sup> President Clinton's March 1, 1995, Address to the Nixon for Peace and Freedom Policy Conference and the Department of Energy Openness Initiative, February 6, 1996.

<sup>2</sup> National Academy of Sciences, Committee on International Security and Arms Control, Management and Disposition of Excess Weapons Plutonium, National Academy Press, Washington, DC, 1994.

<sup>3</sup> U.S. Department of Energy, DOE/MD-0002, "Summary Report of The Screening Process, March 29, 1995. Referred to as "The Screening Report" in this document.

### ES.2.3 Reactor Alternatives

*Existing light water reactors* can be readily converted to enable the use of MOX fuels. Many European light water reactors operate on MOX fuel cycles and at least three companies are actively involved in MOX fuel fabrication. Although some technical risks exist for the alternative, they are all amenable to engineering resolution.

The MOX fuel cores which are currently operating in Europe are partial cores. The cores analyzed in this report are full core MOX fuel cycles. Full core MOX fuel designs were selected to complete the disposition mission faster with fewer reactors. The full core MOX fuel designs can be implemented with or without integral depletable neutron absorbers, where the absorbers provide enhanced plutonium throughput capability but require an extensive fuel qualification demonstration program. For cores not using integral neutron absorbers, there is no substantial difference between partial versus full core MOX fuel cores for fabrication; the differences will reside in reactor performance since additional analyses will be required to confirm the adequacy of the new full core MOX fuel designs.

*CANDU* reactors appear to be capable of operating on MOX fuel cycles, but this has never been demonstrated on any industrial scale. Therefore, additional development is required to achieve the level of maturity for the *CANDU* reactors as exists for light water reactors.

The *partially complete* and *evolutionary light water reactor* alternatives are similar to the existing light water reactor alternative, except that the reactors need to be completed or built, respectively, and the core designs would differ somewhat. There is more technical risk for these alternatives, relative to the existing light water reactor alternative. The increased technical risks are due to two factors, namely: (1) the partially complete and evolutionary reactor alternatives core designs both require integral neutron absorbers—a novel MOX fuel technology not currently in use—to perform the mission with only two reactors; and (2) there are inherent uncertainties associated with completing or building reactor facilities. These reactors would generate additional spent fuel above that for existing light water reactors.

### ES.2.4 Immobilization Alternatives

All of the immobilization alternatives will require qualification of the waste form for the high-level waste repository.

All *vitrification* alternatives require additional research and development prior to implementation of immobilization of weapons-usable plutonium. However, a growing experience base exists relating to the vitrification of high level waste. These existing technologies can be adapted to the plutonium disposition mission, though different equipment designs and glass formulations will generally be necessary.

The facility requirements for *ceramic* immobilization are generally similar to those for vitrification. Vitrification and ceramic immobilization alternatives are similar with regard to the technical maturity of incorporating plutonium in their respective matrices. Ceramic

immobilization offers the potential for superior plutonium confinement over geologic time frames.

The technical viability of the *electrometallurgical treatment* has been demonstrated for treatment of spent nuclear fuels, but has not yet been fully established for the plutonium disposition mission. The experimental data base for the alternative is limited, and critical questions on waste form performance are not yet resolved. This alternative is considered practical only if the underlying technology is developed.

#### **ES.2.5 Deep Borehole Alternatives**

The most significant uncertainties for the deep borehole alternatives relate to selecting and qualifying a site and to obtaining the requisite licensing approvals. These uncertainties can be resolved but will first require a mandate. The front-end feed processing operations for the deep borehole alternatives are much simpler than for other alternatives because no highly radioactive materials are processed, thus avoiding the need for remote handling operations. Emplacement technologies are comprised of largely low-technology operations which would be adaptations from existing hardware and processes used in industry, requiring only a system integration of the various components for this application. One of the chief safety advantages of the deep borehole alternatives is their ability to isolate plutonium from the biosphere on geologic time scales.

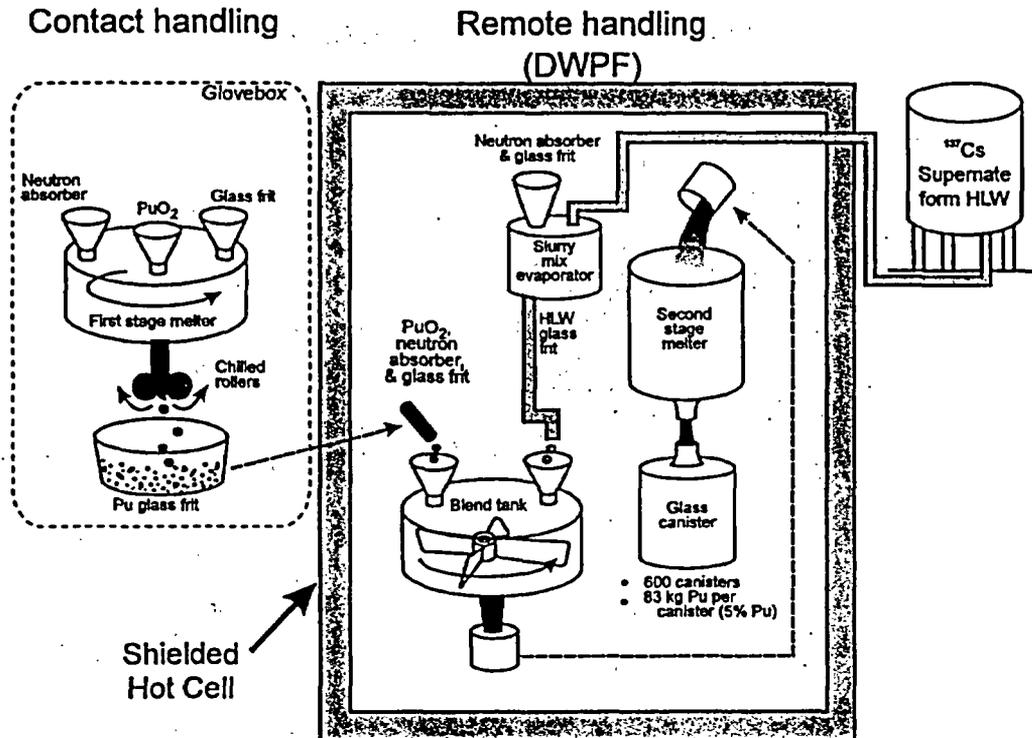
#### **ES.2.6 Hybrid Alternatives**

Two hybrid alternatives were considered as examples of how different technologies might be combined to effect disposition of all the nation's surplus plutonium. Since hybrids combine technology from different categories that were deemed technically viable, both hybrid alternatives are technically viable. The hybrid alternatives benefit by combining the strengths of two different technology approaches and thus provide robustness since they provide a dual path for implementing plutonium disposition.

### **ES.3 COST SUMMARY**

The variants discussed in this report are based on pre-conceptual design information in most cases. As such, large uncertainties in the point estimates for cost and schedule estimates provided in this report apply. The key parameters that drive the uncertainties are identified explicitly in Chapters 4 and 5 for the cost and schedule estimates, respectively. These parameters include: *for all alternatives*: how will the alternatives develop and comply with regulatory and oversight requirements and how will front-end plutonium processing be configured (existing facility, co-located, or new facility); *for reactor alternatives*: how many and what kind of reactors will be used, what core management strategies are adopted, and what are the business arrangements for implementation; *for immobilization alternatives*: what are the material throughputs and facility schedules and how will waste form processing and qualification proceed; *for deep borehole alternatives*: how will site selection

Figure 2-7. Vitrification Adjunct Melter Variant



### 2.3.2 Ceramic Alternative

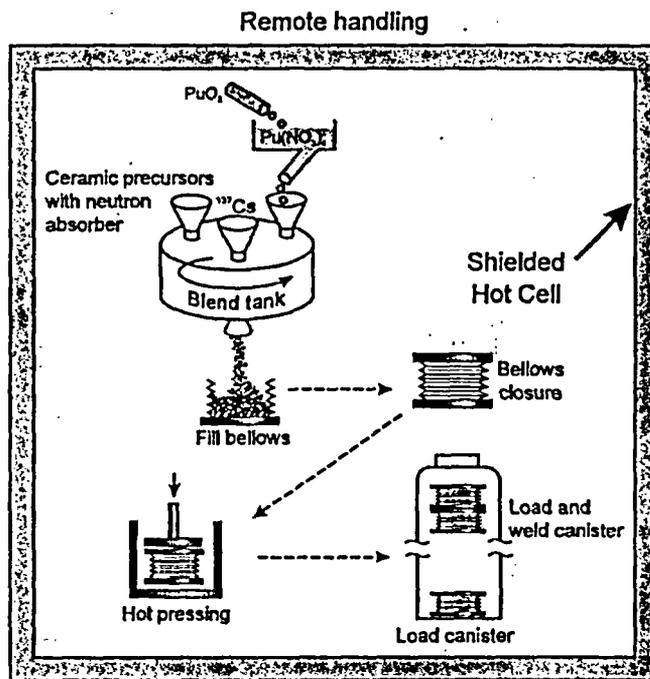
Since the late 1970s, various ceramic waste forms have been considered for immobilization of high-level waste; however, no industrial experience exists for high-level ceramic waste forms unlike borosilicate glass forms. The ceramic waste form is attractive for immobilization purposes because of its extremely 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 <sup>137</sup>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  $\text{TiO}_2$  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 ( $\text{CaZrTi}_2\text{O}_7$ ), hollandite ( $\text{BaAl}_2\text{Ti}_6\text{O}_{16}$ ), perovskite ( $\text{CaTiO}_3$ ), and rutile ( $\text{TiO}_2$ ). 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.

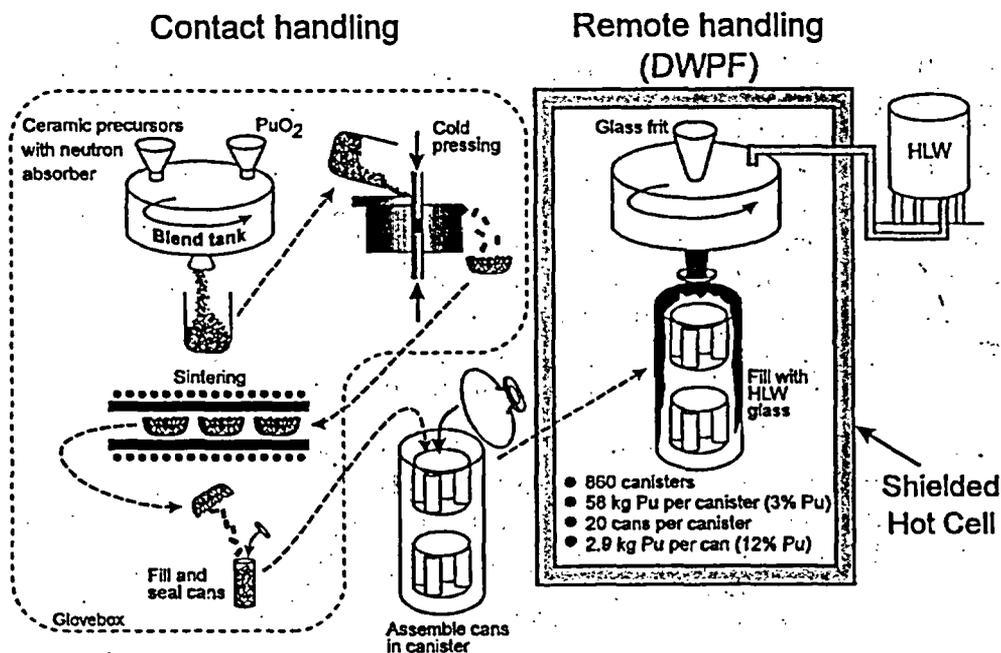
The facility is assumed to be constructed and operated on a generic site. After actual site selection, more specific site-related information will be required.

### 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  $^{137}\text{Cs}$ ) in the sintering process is not an issue.

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



of that experience is limited to applications where actinide concentrations were very low (generally less than 0.1% by weight). However, an experience base for vitrification with higher concentrations of plutonium is beginning to emerge.

Conceptual designs of systems and components have been identified for the vitrification variants, and technologies have been demonstrated at laboratory scale. Crucible melts with plutonium nitrate feeds have successfully been dissolved in glass. For example, plutonium loading has been demonstrated at the laboratory scale at 11 wt % for Löffler glass (the proposed high-temperature glass form for the can-in-canister glass variant) and at 5% for the lower temperature alkali-tin-silicate (ATS) glass (the proposed glass for incorporating the cesium radiation barrier in the greenfield and adjunct melter variants). Key processing parameters requiring further development and demonstration are plutonium oxide (high and low fired) solubility in glass, uniform mixing in the melter, and processing time and temperatures for production-reliable operation of the melter at the required glass physical properties.

The can-in-canister variant appears the more viable since the glass containing the plutonium does not have to simultaneously incorporate the  $^{137}\text{Cs}$  because the radiation source is the vitrified high-level waste outside the can. In addition, this approach allows use of the Defense Waste Processing Facility (DWPF) at Savannah River, eliminating the need for a new hot cell. The can-in-canister variant has been successfully demonstrated cold (i.e., without radionuclides) at the DWPF.

### **3.3.2 Ceramic Alternative**

The ceramic variants are expected to provide superior confinement of plutonium over geologic time scales. This argument is supported by the existence of mineral forms found in nature ("natural analogs") that have demonstrated the immobilization of actinides for periods exceeding 100 million years. Ceramic waste forms have been under development for high-level waste for many years; however, the application of ceramic technology for the immobilization of plutonium is currently developmental. Key technical issues for plutonium immobilization include achieving simultaneous high densities, reacting plutonium from oxides to an incorporated phase, and attaining compatibility with expected impurities. Success in each of these areas depends on the ceramic mineral formulation, as well as the methodology selected for fabrication (including the technology for densifying the ceramic and whether the plutonium feed is dry oxide or a nitrate solution).

The two fabrication methods for ceramic immobilization being considered for this mission are generally well known: hot pressing in bellows and cold pressing and sintering. Hot pressing generally achieves higher densities and can retain  $^{137}\text{Cs}$  added as a radiation barrier but accommodates a relatively lower throughput. Cold pressing and sintering is generally more cost effective due to a higher throughput and is suitable only for the can-in-canister approach because it will likely not retain  $^{137}\text{Cs}$  at the high temperatures in the sintering furnace.

The can-in-canister variant appears the more viable since the ceramic containing the plutonium does not have to simultaneously incorporate the  $^{137}\text{Cs}$ . In addition, this approach allows use of the Defense Waste Processing Facility (DWPF) at Savannah River with minimal interference on the ongoing high-level waste operation and eliminates the need for a new hot cell.

Hot pressed ceramic samples containing 10 to 100 grams of plutonium at a loading of 12% have been prepared which indicate that full-scale production is viable. Cold pressing and sintering has produced ceramic pellets with oxide powder loading of 12%. Full characterization of these samples have not yet been completed. The technology for cold-press and sinter is similar to that used for production of MOX fuel and is a mature production advantage for this waste form.

The baseline feed approach for producing hot press ceramics is the use of plutonium nitrate solution. This "wet" feed approach generally results in a more fully reacted plutonium ceramic product; however, it requires an off-gas system (thus larger capital equipment) and could result in greater volumes of secondary waste. A more desirable approach would be to use a "dry" plutonium oxide feed, which results in significantly reduced secondary waste but is more difficult to obtain completely reacted plutonium in the ceramic matrix and is less well demonstrated at the present. Additional developmental work to reduce technical uncertainties would be required to select the dry feed approach.

### **3.3.3 Electrometallurgical Treatment Alternative**

The electrometallurgical treatment alternative requires further development to confirm its applicability as an immobilization option for plutonium disposition. Although the technical viability of several components of this alternative is well established for spent nuclear fuels, questions regarding the technical viability of this alternative for the plutonium disposition mission remain. Most of the technical risk associated with this alternative is due to a small experience base of several unit processes with pure plutonium. The lithium reduction step of the process has been demonstrated with uranium oxide and with mixed uranium and plutonium oxides but not with pure plutonium oxide or plutonium containing large quantities of inert material. The zeolite waste form has been demonstrated at a few gram scale (total mass) using plutonium-loaded chloride salt. The electrorefining process is currently being operated with irradiated Experimental Breeder Reactor-II fuel and blanket assemblies on a limited demonstration basis at ANL-W using some of the same facilities, equipment and processes that would apply to fissile materials disposition.

Regarding the qualification of the zeolite waste form for the high-level waste repository, a NAS National Research Council Report noted several concerns with the long-term performance of this waste form, including radioactive decay effects and chemical and thermal

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Table 4-3. Immobilization Alternatives Costs

Immobilization Alternative	Facility	Constant \$ (millions)			Discounted \$ (millions)		
		Investment	Operating	Net Life Cycle Cost	Investment	Operating	Net Life Cycle Cost
Vitrification Greenfield	Front-end	1000	980	1980			
	Immobilization	1030	1800	2830			
	Repository	0	300	300			
	Total	2030	3080	5110	1250	1300	2550
Vitrification Can-in-Canister	Front-end	360	980	1340			
	Immobilization	220	170	390			
	Repository	0	100	100			
	Total	580	1250	1830	410	640	1050
Vitrification Adjunct Melter	Front-end	340	980	1320			
	Immobilization	680	1330	2010			
	Repository	0	300	300			
	Total	1020	2610	3630	680	1150	1830
Ceramic Greenfield	Front-end	860	820	1680			
	Immobilization	950	1720	2670			
	Repository	0	320	320			
	Total	1810	2860	4670	1120	1200	2330
Ceramic Can-in-Canister	Front-end	360	980	1340			
	Immobilization	220	170	390			
	Repository	0	100	100			
	Total	580	1250	1830	410	640	1050
Electrometallurgical Treatment <sup>1</sup>	Front-end	730	890	1620			
	Immobilization	460	870	1330			
	Repository	0	480	480			
	Total	1190	2240	3430	770	940	1710

<sup>1</sup> Costs are based upon a stand-alone plutonium disposition mission. Cost sharing with DOE programs for the treatment of spent fuel has the potential to reduced costs by approximately \$600 million.

Existing facilities and waste disposal operations provide the opportunity for significant cost savings for the plutonium disposition mission. As indicated by the data, the investment cost of the vitrification can-in-canister variant is approximately one fourth the greenfield vitrification variant investment cost. The cost ratio is about a factor of three for the ceramic greenfield versus the ceramic can-in-canister variant. Less dramatic investment savings can be realized using an adjunct melter strategy for vitrification, where costs are one half of the greenfield vitrification investment costs. Note that the front-end costs account for half of the investment costs for the two greenfield variants and well over half of the can-in-canister variants. The costs for the can-in-canister variants appear identical in the table; however, the variants were costed separately on their own bases.

The investment costs for the vitrification greenfield front-end facilities are approximately \$150 million more than the ceramic greenfield front-end due to the inclusion of a first stage melter in the vitrification front-end facility. The investment cost of the electrometallurgical treatment variant is less than the cost of greenfield variants, but more than the cost of can-in-canister variants. The front-end facility for electrometallurgical treatment accounts for approximately two thirds of the investment costs. However, those costs could be reduced by performing some of the front-end process steps at other locations, thereby avoiding the need to add additional facility space necessary to co-locate all operations at ANL-W.

Operating costs range from \$1.2 billion for the can-in-canister variants to over \$3 billion for the vitrification greenfield variant. Use of DWPF reduces immobilization facility operating costs by a factor of ten relative to greenfield immobilization facilities for the vitrification and ceramic immobilization approaches. Use of DWPF facilities for the can-in-canister variants relative to the greenfield variants reduces overall operating costs by a factor of two. Repository costs refer to the canisters resulting from disposition operations. The electrometallurgical treatment alternative is assumed to process plutonium independent of a mission to treat spent nuclear fuel. If the plutonium disposition mission is conducted simultaneously with the operations to treat spent nuclear fuel, then approximately \$600 million could be saved through the sharing of concurrent operating, storage, and waste disposal costs.

Life cycle costs of can-in-canister concepts are also significantly lower than for other immobilization variants. Discounted life cycle costs range from \$1.0 billion for the can-in-canister variants to \$2.6 billion for the vitrification greenfield variant.

*(Information previously here was moved to Chapter 6.)*

#### **4.4 DEEP BOREHOLE ALTERNATIVES COSTS**

##### **4.4.1 Assumptions**

Deep borehole alternatives, described in detail in Chapter 2, incorporate the following economic assumptions:

- 1) Government ownership of plutonium processing and borehole facilities is assumed.
- 2) Front-end and immobilization facilities are collocated at a government-owned site with plutonium processing infrastructure. Front-end processes are located in existing buildings where possible.
- 3) Borehole facilities are sited at a generic, non-DOE site.

##### **4.4.2 Cost Analysis**

Investment costs, operating costs, undiscounted life cycle costs, and discounted life cycle costs of borehole alternatives are summarized in Figure 4-4, with supporting detail of costs by facility shown in Table 4-4.