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DISPOSAL OF DEFENSE SPENT FUEL AND HLW

FROM THE IDAHO CHEMICAL PROCESSING PLANT*

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ABSTRACT

Acidic high-level radioactive waste (HLW) resulting from fuel reprocessing at the Idaho Chemical Processing Plant (ICPP) for the U. S. Department of Energy (DOE) has been solidified to a calcine since 1963 and stored in stainless steel bins enclosed by concrete vaults. Several different types of unprocessed irradiated DOE-owned fuels are also in storage at the ICPP. In April, 1992, DOE announced that spent fuel would no longer be reprocessed to recover enriched uranium and called for a shutdown of the reprocessing facilities at the ICPP. A new Spent Fuel and HLW Technology Development program was subsequently initiated to develop technologies for immobilizing ICPP spent fuels and HLW for disposal, in accordance with the Nuclear Waste Policy Act. The Program elements include Systems Analysis, Graphite Fuel Disposal, Other Spent Fuel Disposal, Sodium-Bearing Liquid Waste Processing, Calcine Immobilization, and Metal Recycle/Waste Minimization. This paper presents an overview of the ICPP radioactive wastes and current spent fuels, with an emphasis on the description of HLW and spent fuels requiring repository disposal.

INTRODUCTION

Irradiated nuclear fuel has been reprocessed at the Idaho Chemical Processing Plant (ICPP) since 1953 to recover uranium-235 and krypton-85 for the U. S. Department of Energy (DOE). The resulting acidic high-level liquid radioactive waste (HLLW) has been solidified to a high-level waste (HLW) calcine since 1963 and stored in stainless steel bins enclosed by concrete vaults. Residual HLLW and radioactive sodium-bearing liquid waste are stored in stainless-steel tanks contained in concrete vaults. Several different types of unprocessed irradiated DOE-owned fuels are also in storage at the ICPP.

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In April, 1992, DOE announced that spent fuel would no longer be reprocessed to recover enriched uranium and called for a shutdown of the reprocessing facilities at the ICPP. A new Spent Fuel and HLW Technology Development program was subsequently initiated to develop technologies for immobilizing ICPP spent fuels and HLW for disposal, in accordance with the Nuclear Waste Policy Act. The ICPP Spent Fuel and Waste Management Technology Development Program consists of the following basic elements:

- Systems Analysis
- Sodium-Bearing Liquid Waste Processing
- Calcine Immobilization
- Spent Graphite Fuel Conditioning
- Other Spent Special Fuel Conditioning
- Metal Recycle/Waste Minimization

Systems Analysis will include the identification and evaluation of all elements related to waste disposal to provide the basis for integrated, proactive, strategic decision making to accomplish the technology development mission. Radioactive sodium-bearing liquid waste processing will involve the evaluation and development of treatment technologies which will minimize the quantities of waste to be generated in the future and to be disposed in the repository. Calcine immobilization will investigate and develop methods to minimize resulting high-level waste volumes, considering all feasible options, with glass-ceramic as the baseline waste form. Graphite fuels disposal will examine, evaluate and develop one of three potential disposal paths and the technologies associated with one of those options. Other spent fuel disposal will be concerned with the evaluation and development of technologies for characterizing and processing or conditioning, for geologic disposal, metal alloy or metal-clad fuels presently stored or slated for future storage at the ICPP. Metal Recycle/Waste Minimization is concerned with the development of technology to reuse the contaminated and activated metals from decommissioned structural material and vessels in the nuclear and DOE defense programs. Conditioned spent fuel, immobilized calcine, and possibly a small quantity of material resulted from processing the sodium-bearing liquid waste will be considered for disposal in a geologic repository. A schedule of major milestones of the proposed program is shown in Figure 1, assuming requested funding levels.

This paper will present an overview of the ICPP calcined HLW, sodium-bearing radioactive liquid waste, and graphite and special fuels, with emphasis on the description of ICPP HLW and spent fuel requiring repository disposal.

CALCINED HLW

The calcining process operates by feeding an acidic HLW to a fluidized-bed calciner operating at 500° C which forms a mixture of particles (0.2 - 0.5 mm) and fines (10 - 200 μm). Alumina and zirconia calcines were generated from wastes resulting from reprocessing aluminum and zirconium-based fuels, respectively. Fluorinel-Na and zirconia-Na calcine were produced from a blend of sodium-bearing waste and HLW resulting from reprocessing a more recent fluorinel fuel and zirconia-based fuel, respectively. Radionuclide content in all of the calcine types is less than about 1 wt%, and the Curie content and heat generation is approximately 24 kCi/m^3 and 70 W/m^3, respectively.

Calcine is also a mixed hazardous waste, and the treatment process for calcine immobilization must meet current Resource Conservation and Recovery Act (RCRA) Land Disposal Restrictions (LDR) regulations. The EPA Third Thirds Rulemaking specifies vitrification as the best demonstrated available technology (BDAT) for mixed HLW, and has proposed in another rulemaking that a glass-ceramic process is also a BDAT for calcine.

The calcined waste is stored near-surface in stainless steel bins within concrete vaults. The bin sizes are approximately 4-m diameter by 12.5 to 18.5-m high. Some of the bins are cylindrical and others are of an annular configuration. Currently there is an inventory of 3,600 m^3 HLW calcine at ICPP with compositions shown in Table I. Not shown in Table I is zirconia-Na calcine, which has a similar composition to fluorinel-Na calcine. The amount of alumina, zirconia, zirconia-Na, and fluorinel-Na calcines is approximately 560, 1250, 950, and 600 m^3, respectively. The remaining 240 m^3 calcine inventory consists of calcines from processing other minor fuels and start-up bed material.

CALCINE IMMOBILIZATION TECHNOLOGIES

The objective of the Calcine Immobilization Program is to develop and demonstrate a process to immobilize ICPP HLW calcine using BDAT in an acceptable form at minimum volume for final disposal. Areas of effort included in this task are 1) defining disposal criteria based on applicable regulations, 2) evaluating alternative technologies for feasibility and overall volumes, 3) developing waste form formulations for the feasible alternatives, 4) conducting nonradioactive and radioactive verification studies of various technologies, including grinding, degassing, densification, robotic areas, and waste form formulations, and 5) testing of subsystem components to provide operation parameters needed for full-scale design.

Several technologies have been identified to date that could immobilize calcine; these include vitrification and glass-ceramic processing. Nonradioactive and some radioactive laboratory tests have been carried out to

Table I. Composition of ICPP Calcine

Type of Calcine and Composition, wt%			
Component	Alumina	Zirconia	Fluorinel -Na Blend*
Al_2O_3	82-95	13-17	9
Na_2O	1-3	---	4.8
K_2O	---	---	1.2
ZrO_2	---	21-27	17-18
CaF_2	---	50-56	41-42
CaO	---	2-4	12
So_4	---	---	3
B_2O_3	0.5-2	3-4	3.0-3.4
CdO	---	---	6.7-7.0
Misc.	0.5-1.5	0.5-1.5	0.5-1.5
Fission Products and Actinides	<1	<1	<1

* Contains additional nitrate at 10-15 wt%

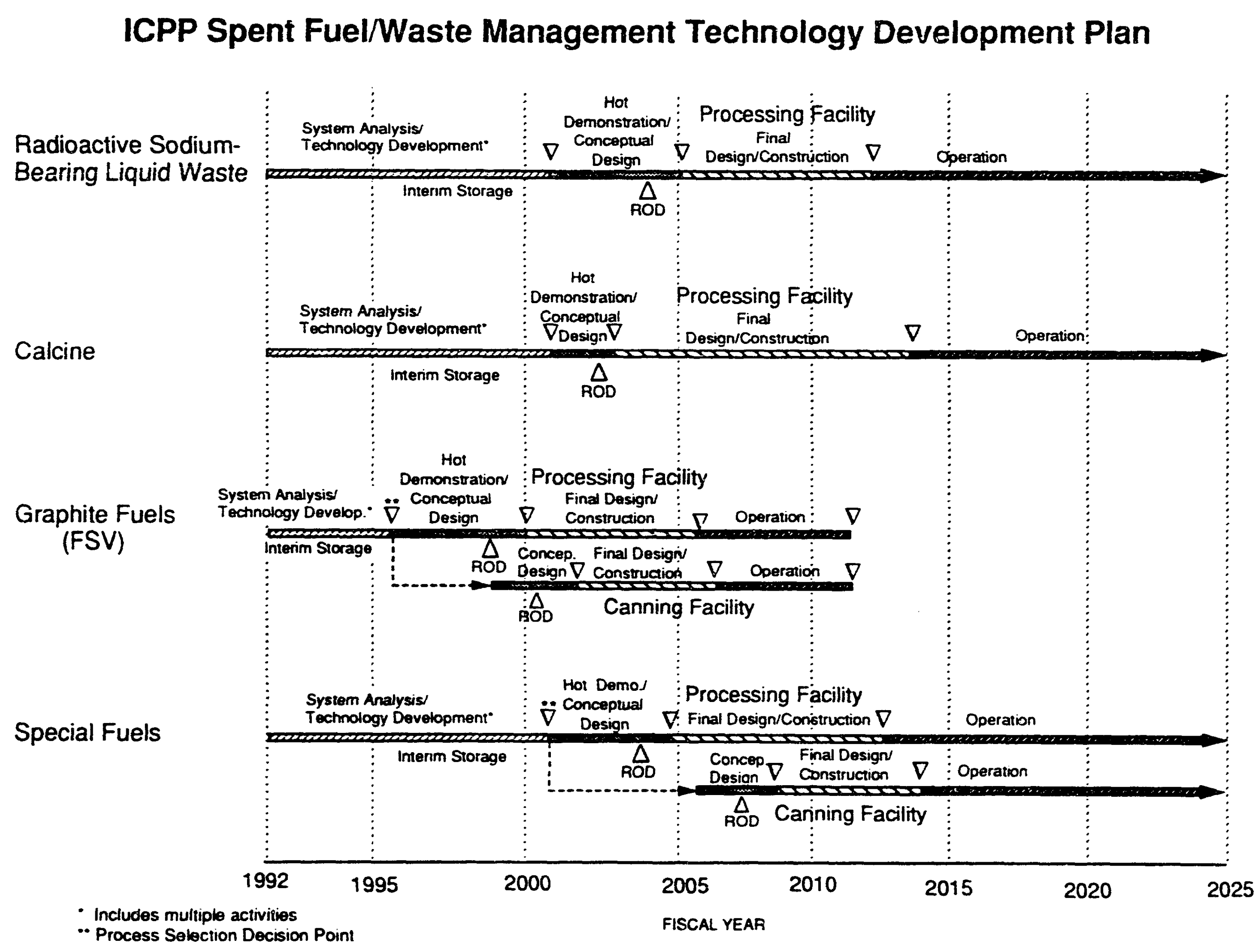

FIGURE 1. Schedule of Major Milestones of the Proposed ICPP Spent Fuel/Waste Management Technology Development Plan, Assuming Requested Funding Level

develop glass waste forms for existing calcines. Some nonradioactive glass-ceramic forms with high waste loadings of 50 to 70 wt percent have been prepared using simulated calcine and have shown leach rates similar to the glass products. Limited small-scale component and mock-up tests have been performed for selected unit operations of the glass-ceramic process, including calcine grinding, calcine transport, and vessel filling. Simplified, small-scale calcine retrieval mock-up tests have been run using calcium carbonate as a nonhazardous stimulant. This work, while not complete, provides confidence that acceptable processes can be developed in a reasonable period of time.

Nonradioactive and radioactive tests will be run to characterize the glass-ceramic materials and to verify the acceptable range of compositions for the most promising formulations. The results of the tests will be used to develop waste form specifications and to establish criteria for pilot-scale tests. Nonradioactive and radioactive unit operations tests will be run to determine feasibility and criteria for full scale design.

Calcine retrieval component tests are required to verify new technologies in pneumatic transport and robotics application areas. Glass-ceramic component tests are required in all of the unit operations in the process, including calcine-additive blending, grinding, transport vessel filling, remote vessel welding, densification of calcine-additive mixture to form a glass-ceramic, and packaging and decontamination of the waste container for disposal. The component testing will be carried out in existing ICPP pilot plants and the Multifunctional Pilot Plant Facility. The results of the these tests will be used to select the process components and to design integrated pilot plant demonstration tests to support construction and operation of the full-scale production facility.

The overall program schedule assuming glass-ceramic technology shows a record of decision for the full scale immobilization plant in the year 2003 and hot start-up of a product facility in 2014. Key FY-93 milestones include updating the ICPP waste form specifications and reporting the development progress of aluminum-silica based glass-ceramic waste forms. Designs for an intermediate-scale calcine grinder and small-scale calcine mixing will be completed and procurement/fabrication efforts for both unit operations will be undertaken. Evaluations of promising separation technologies to minimize the volume of high-level radioactive waste requiring permanent disposal will be initiated. Key equipment options required for calcine retrieval will be identified and component evaluation testing will be initiated.

SODIUM-BEARING RADIOACTIVE LIQUID WASTE

Sodium-bearing radioactive wastes were produced from decontamination and solvent recovery operations at ICPP, resulting in approximately 1.5 million gallons currently in storage. This waste is currently stored in seven different stainless-steel tanks in concrete vaults of nominal 300,000 gallon (1,100 m^3) capacity per tank with each tank being contained in a concrete vault. Under LDR, this waste must be processed with the best demonstrated available technology (BDAT) prior to disposal. Five of the tanks do not meet current seismic codes, and none of the tanks meet the RCRA requirements for secondary containment. As a result, the Consent Order to the State of Idaho's Notice of Noncompliance (NON) requires that the sodium-bearing waste be depleted by 2009 from the five tanks which do not meet current seismic codes and by 2015 from the remaining two tanks.

The sodium-bearing waste is acidic and has an average composition as shown in Table II. Because of the acidic nature, the waste does not have metal precipitates as found in other DOE waste tanks which have been neutralized. Although sodium-bearing waste may not fit the legal description of a HLW, the composition of some of the radionuclides will likely be greater than the Class C LLW and TRU waste limits. Past processing of the sodium waste was accomplished by calcining as a blend with acidic HLLW from reprocessing operations. Because of the low melting range of alkali oxides and resulting particle agglomeration, the sodium-bearing waste cannot be calcined directly in the New Waste Calcining Facility (NWCF) but must be blended with aluminum nitrate. Although this flowsheet appears to be feasible and is considered a baseline case, the waste volumes will likely be higher than other options. Other processing options under evaluation include separation processes to concentrate the radionuclides to reduce the volume requiring disposal.

The overall program schedule for developing processes to treat and immobilize sodium-bearing wastes shows a record of decision for the full-scale plant in 2004 and hot startup in 2012, assuming the requested funding level. Key FY-1993 milestones include identification of alternative processing methods, characterizing existing sodium-bearing waste, and completing preliminary scoping tests.

SPENT FUELS

The ICPP currently stores many different types of fuel, including naval propulsion (approximately 10 metric tons uranium), graphite and special fuels. This paper will address primarily graphite and special fuels.

Special Fuels

In addition to the sodium-bearing radioactive liquid and the calcined HLW, the US DOE has approximately 730 metric tons of material labelled as "special fuel" because no specific processing technique or recycle facility is available. There are over 90 identified types of special nuclear fuel at INEL and over 100 types in the DOE complex. Approximately 474 metric tons of special fuels occupying 60 m^3 is stored at the INEL.

TABLE II. Chemical Composition of Sodium-Bearing Waste

COMPONENT	AVG. COMPOSITION (moles/liter)	RANGE (moles/liter)
Acid (H^+)	1.45	0.43 - 1.92
Nitrate (NO_3^-)	4.36	2.93 - 5.79
Aluminum (Al^{3+})	0.55	0.21 - 0.81
Sodium (Na^+)	1.26	0.78 - 2.00
Potassium (K^+)	0.15	0.10 - 0.23
Fluoride (F^-)	0.07	0.04 - 0.17
Zirconium (Zr^{4+})	0.003	0.000 - 0.009
Boron (B^{3+})	0.018	0.007 - 0.024
Calcium (Ca^{2+})	0.04	0.00 - 0.07
Chloride (Cl^-)	0.02	0.008 - 0.043
Iron ($Fe^{2+,3+}$)	0.03	0.01 - 0.05
Chromium ($Cr^{2+,3+,6+}$)	0.006	0.002 - 0.013
Cadmium (Cd^{2+})	0.002	0.000 - 0.004
Lead ($Pb^{2+,4+}$)	0.001	0.001 - 0.002
Mercury ($Hg^{1+,2+}$)	0.002	0.001 - 0.003
Manganese ($Mn^{2+,3+,4+,7+}$)	0.01	0.01 - 0.02
Phosphate (PO_4^{3-})	0.009	0.002 - 0.023
Sulfate (SO_4^{2-})	0.04	0.01 - 0.07
Specific Gravity	1.22	1.15 - 1.26

The special fuel varies widely in characteristics. There are individual rods in buckets, fuel assemblies, canned fuel, fuel test assemblies, etc. The length varies from 15 to 406 cm and effective diameter from 1 to 46 cm. Enrichments and burn-ups also vary widely. Table III summarizes the characteristics of special fuels based on enrichment, fuel type, burn-up, cladding, other materials, hazardous constituents, and leachability.

Graphite Fuels

There are two basic types of HTGR (High Temperature Gas Reactor) or graphite fuel at ICPP. The largest quantity is from the Fort St. Vrain Reactor (FSV) in Colorado and the rest comes from the Peach Bottom Reactor. The Peach Bottom fuel is a different configuration then the FSV Fuel but the properties are similar to the FSV Fuel. The total amount of graphite fuel in inventory is about 340 metric tons.

The FSV fuel element consists of a 280-lb hexagonal graphite block, 14.2-in. across the flats and 31.2-in. high. Each graphite fuel block contains 108 coolant channels and 210 fuel holes, all drilled from the top face of the element. The fuel holes occupy alternating positions with the coolant channels in a triangular array within the element structure and contain the nuclear fuel. The Peach Bottom fuel utilized a 12-ft-long cylindrical fuel element 3.4 inches in diameter composed largely of graphite, containing about 1.8 kg of uranium and thorium. These heavy metals are present as carbon-coated particles that were formed (compacts) by addition and sintering of carbonaceous materials.

SPECIAL FUEL DISPOSITIONING PROGRAM

The objective of the special fuel program is to characterize all the special fuel at the INEL and to develop processes for conditioning spent fuel for dispositioning in a geologic repository.

This program will develop and demonstrate methods for dispositioning of special fuel. This program will also develop fuel inspection criteria for a variety of special fuel at the INEL. Fuel will be identified for subsequent inspection and characterization. Inspection issues will be evaluated and development of fuel inspection criteria will be initiated. The program will establish how the fuel can be conditioned to meet preliminary limiting conditions and repository criteria for long-term, direct dispositioning of special fuel. Alternative direct conditioning methods will be investigated.

TABLE III. FUEL CHARACTERISTICS FOR WASTE TYPE

Form Cat	U-235 enrich	fuel type	fuel matrix	burnup	clad mater	other mater[2]	hazards[3]	leach[4]
	H High > 20% L Low 1-20% D Depleted < 1%	oxide alloy metal hydride unknown	SST Al. Mo BeO.MgO ZrO2,CaO ThO2 none ternary unknown	H 40-50.000 M 10-40.000 L 1-10.000 neg <1 unknown (Mwd/MTHM)[1]	Al SST Zr ceramic none unknown	Pu C etc unknown	Na met Cr Pu hydride unknown	M medium L low U unknown
1.	H	hydride	none	L	Mix	C,Pu,Mo	Cr,Pu	M
2.	H	oxide	SST	M	SST	Ti.Pu	Pu.Cr	L
3.	H	alloy	Al	H	Al	Pu	Pu	M
4.	H	oxide	BeO.MgO	L	none	Be.Mg.Y ceramic	unknown	L
5.	H	oxide	ZrO2	H	Zr	Pu.B	Pu	M
6	H	oxide	ZrO2.CaO	H	Zr	ZrO2.CaO epoxy	Pu	M
7.	L	alloy	none ThO2	U	SST	Th.Na.Mo U-233	Pu.Na	L
8.	L	oxide	ThO2.CaO ZrO2	U	Zr	Th.CaO.Pu U-233	Pu,	L
9.	H	alloy	none	L	Zr	Na met,Pu	Pu.Na	L
10.	H	oxide	SST	U	Zr	B4C. thermal-	unknown	L
11.	H	metal	none	H	SST	Pu.Na	Pu.Na	L
12.	L	metal	Mo	U	SST	Pu	Pu	L
13.	H	oxide	none	neg	none	unknown	unknown	M
14.	L	oxide	none	U	Zr	Be.Pu	Pu	U
15.	L	oxide	unknown	U	Zr,SS	Pu	Cr,Pu	U
16.	H	oxide	nicrome	U	unknown	unknown	unknown	U
17.	D	unknown	unknown	U	unknown	unknown	unknown	U
18.	L	oxide	unknown	U	unknown	Pu	Pu	U
19.	L	oxide	ternary	U	SST	Pu	Pu	U
20.	L	alloy	Mo	neg	SST	unknown	Na	M

Classification rationale

1. Enrichment affects fission product spectrum and Pu quantities
2. Fuel type affects potential chemical reactions. leachability
3. Burnup affects fission product inventory, Pu quantity, actinides
4. Cladding affects leachability until breached
5. Other material affects potential hazards, leach rate, chemical reactions
6. Hazards affect potential storage problems
7. Leachability affects storage facility design

Notes:

1. Mwd/MTHM = MegaWatt day/metric ton heavy metal
2. Materials that may react, generate gas, be hazardous, used in construction.
3. Materials known to represent waste hazard.
4. Leachability: the potential that the material will leach
   Low: intact cladding in a can
   Medium: scrap in a can or intact cladding but no can
   High: cladding breached not in a can
   Also dependent on the chemical form of the material. Right now all materials are assumed to be in the oxide form.

The special fuel potential conditioning technologies include:

- Can the fuel directly - this option include the addition of neutron poison, glass, ceramic, metal, corrosion barrier, or other additive.
- Dilute enriched uranium with depleted uranium - this option includes dissolve, blend and convert to solid, or shred and blend concepts.
- Shred and mix with other material in geometrically safe can.
- Chloride volatility treatment to separate components.
- Reprocess the spent fuel, calcine waste, convert to glass or ceramic.
- Cut and package in geometrically safe can.
- Recycle metal such as diluting the high-enriched uranium (HEU) for use in commercial reactors.
- Others to be identified.

The overall program schedule assuming the required funding level shows a record of decision by ? 04 and hot operation by 2013. Key FY-93 milestones include fuel inspection and characterization, criteria development, identify and evaluate candidate technology, and laboratory testing. Future milestone include the design, construction, and test of an integrated cold pilot plant.

GRAPHITE SPENT FUEL CONDITIONING PROGRAM

The objective of the graphite spent fuel conditioning program is to establish disposal criteria, and to perform development, engineering, and demonstration of waste conditioning methods for fort St. Vrain and other spent graphite fuels.

Options for waste management and disposal of spent graphite fuel elements include the following methods:

- Dispose of the fuel elements directly by placing the fuel directly into canisters with little or no conditioning of the fuel and then sending the canisters directly to the geologic repository.
- Mechanically remove fuel rods and package into a canister with a glass that fills the voids for repository disposal.
- Burn the bulk graphite to the SiC layer for the FSVR and Peach Bottom Core 2 fuel. The Peach Bottom Core 1 elements are assumed to be burned oxide ash. Dispose of the ash in a glass.
- Shred the graphite blocks, burn the bulk graphite, grind the fuel particles, burn the carbon layers, and then encapsulate (glass) the ash and remaining oxides and SiC.
- Shred the graphite blocks, burn the bulk graphite,grind the fuel particles, burn the carbon layers, and then separate the fissile material. The fissile material separation would reduce the chances of a criticality in the repository and the material could be recycled for future use. The remaining waste could then be placed in the repository.

The overall program schedule assuming the required funding level shows a record of decision by 1999 and hot startup by 2007. Key FY-93 milestones include criteria development, identify and evaluate candidate technology, and laboratory testing. Future milestone include the design, construction, and test of an integrated cold pilot plant.

WASTE TO REPOSITORY SUMMARY

The materials planned for repository disposal will include the spent fuel, immobilized calcine, and possibly a small amount of material generated from the Na-bearing waste by separation/concentration processes. The amounts of spent fuel, Na-bearing and calcined HLW material were given in previous sections. The volume of immobilized calcine for repository disposal includes approximately 1,930 canisters of glass-ceramic or 4,920 canisters of glass, using 1.2 m^3 canisters similar as planned for commercial fuel disposal. The disposal volume resulting from the current inventory of 474 metric tons (60 m^3) special fuels, 350 metric tons (260 m^3) graphite fuels, and approximately 10 tons naval fuels will be determined as treatment and packaging options are developed in the Spent Fuel and Waste Management Development Program.

END

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