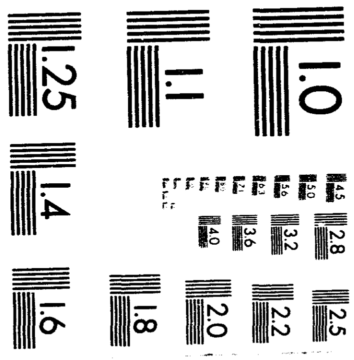
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TECHNOLOGY DEVELOPMENT PROGRAM FOR IDAHO CHEMICAL PROCESSING PLANT SPENT FUEL AND WASTE MANAGEMENT

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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, and describes the Spent Fuel and HLW Technology program in more detail.

INTRODUCTION

Irradiated nuclear fuel has been reprocessed at the Idaho Chemical Processing Plant (ICPP), which is a part of the Idaho National Engineering Laboratory (INEL), 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 in concrete vaults (Berreth, 1989). Residual HLW and radioactive sodium-bearing waste are stored in stainless-steel underground tanks contained in concrete vaults. Several different types of unprocessed irradiated DOE-owned fuels are also stored at INEL (U.S. DOE, 1992). In April, 1992, DOE announced that spent fuel would no longer be reprocessed to recover enriched uranium.

As a result of the decision to curtail reprocessing the ICPP Spent Fuel and Waste Management Technology Development plan has been implemented to identify acceptable options for disposing of the (1) sodium-bearing liquid radioactive waste, (2) radioactive calcine, and (3) irradiated spent fuel stored at the INEL. The plan was developed jointly by DOE and Westinghouse Idaho Nuclear Company, Inc.,(WINCO).

Simply storing spent fuel and high level waste for an indefinite period is not a viable option. High level waste is subject to regulation under the Resource Conservation and Recovery Act (RCRA) and must be disposed of utilizing the Best Demonstrated Available Technology (BDAT). The BDAT for high level liquid waste has been established by the EPA to be vitrification (U.S. EPA, 1990). For ICPP high level waste which has been converted into a granular solid, the proposed BDAT is a glass-ceramic process. The Federal Facilities Compliance Act will require agreements with regulating state agencies to dispose of RCRA wastes on a negotiated compliance schedule. The indefinite storage of spent fuel, while technically feasible, is unlikely to receive public and state acceptance.

Probably the most challenging part of the ICPP Technology Development Plan is the development of technologies for the dispositioning of spent fuel. The Nuclear Waste Policy Act of 1982, including the 1987 amendments, provides for the development of a geologic repository for the placement of commercial spent fuel and defense-generated, solidified, high level waste. The DOE is also required to assess the need for a second repository. The ICPP program will undertake development of repository acceptance criteria and dispositioning techniques to make currently stored fuels ready for storage at a repository. Changes in repository strategies or the possibility of introduction of a defense-related Monitored Retrieval Storage facility will require integration and coordination on a national basis to assure suitable technologies are developed.

The removal of reprocessing as the basis for management of spent fuel and resulting high level waste has required a new approach to management of this material. The ICPP Spent Fuel and Waste Management Technology Development Program is one of the means to developing a new technical basis for high level waste management. The plan proposes design and construction of new facilities during the early part of the next century. After a brief description of the existing ICPP HLW calcine, radioactive sodium-bearing liquid waste, and spent fuels, this paper presents the objectives and scope of the new Spent Fuel and Waste Management Technology Development program.

CALCINED HLW

The calcining process operates by feeding an acidic HLLW 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) (Schindler, 1977). 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 HLLW resulting from reprocessing a more recent fluorinel fuel and older 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 (Berreth, 1988).

Calcine is also a mixed hazardous waste, and the treatment process for calcine immobilization must meet LDR. The EPA Third Thirds Rulemaking (U.S. EPA, 1990) 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,

TABLE I. COMPOSITION OF ICPP CALCINE

Type of Calcine and Composition, wt%

Component	Alumina	Zirconia	Fluorinel -Na Blend[a]
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

a Contains additional nitrate at 10-15 wt%

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 (Berreth, 1988).

ICPP 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 (U.S. DOE, 1992). 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. Under current Resource Conservation and Recovery Act (RCRA) Land Disposal Restrictions (LDR) regulations, this waste must be processed with the Best Demonstrated Available Technology 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 (Berreth, 1988). 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.

SPENT FUEL

The DOE currently stores "special" fuel, graphite fuel, and Naval fuel at the INEL (U.S. DOE, 1992). There are over 90 identified types of special nuclear fuel at the INEL and over 100 types in the DOE complex. About 156 metric tons (MT) of graphite based fuels, 240 MT of Naval propulsion fuels, and 474 MT of various (zirconium, aluminum, and stainless steel based) special fuels are stored at the INEL. The fuel characteristics are summarized in Table III.

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	0-0.009
Boron (B^{3+})	0.018	0.007-0.024
Calcium (Ca^{2+})	0.04	0-0.07
Chloride (Cl^-)	0.02	0.008-0.043
Iron ($Fe^{2+,4+}$)	0.03	0.01-0.05
Chromium ($Cr^{2+,3+,6+}$)	0.006	0.002-0.013
Cadmium (Cd^{2+})	0.002	0-0.004
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, g/cm^3	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 condition of fuel cladding also varies with some fuel intact and capable of continued storage as is and some fuel reduced to debris in buckets. Enrichments and burn-ups also vary widely.

PROGRAM OBJECTIVES

The principal objectives of the ICPP Spent Fuel and Waste Management Technology Development Program are:

1. Investigate direct disposal of spent fuel, striving for one waste form.

2. Determine the best treatment processes for liquid and calcine wastes to minimize HLW and LLW.

3. Demonstrate the integrated operability and maintainability of selected treatment and immobilization processes.

4. Assure that the final implementation is environmentally acceptable, ensures public and worker safety, and is economically feasible.

The strategy to obtain these objectives utilizes a systems approach during development which will take into account all of the factors which may impact final disposition of waste and spent fuels and capitalize on all available technology both national and international by benchmarking.

The ICPP technology development program resulting from the decision to curtail fuel reprocessing includes the following key elements:

1. Systems Analysis

2. Sodium Bearing Liquid Waste Processing

3. Calcine Immobilization

4. Spent Graphite Fuel Conditioning

5. Special Fuel Conditioning

6. Metal Recycle/Waste Minimization

SYSTEMS ANALYSIS

The objective of the Systems Analysis initiative is to develop a logical and consistent approach in executing the Spent Fuel and Waste Management Technology Development Program, looking at all aspects and developing the basis for integrated, strategic decision making using a structured systems method. Decisions will be made and program priorities will be set based on regulatory compliance, reduced risk, reduced cost, increased safety, public acceptance, waste minimization, and other key issues.

A preliminary performance assessment, including an analytical model which will simulate confinement of waste forms in postulated repository situations, will be developed. The assessment will initially be based on general physical knowledge such as rock type and characteristics, water depth and movement, volcanic and seismic activity and the likelihood of migration of the stored waste out of the repository. Characteristics of the waste form, repository location, and repository design will be integrated into the analysis as they become available. The performance assessment will also identify any obvious shortcomings with the candidate waste forms, container design, and repository definitions. As appropriate, the results of the performance assessment will be used to further refine the goals and priorities of the technology development.

SPENT FUEL CONDITIONING

The objective of the Spent Fuel Conditioning program is to characterize all the special fuel at the INEL and to develop technology for conditioning that spent fuel, striving for a single waste form, for dispositioning in a geologic repository. Fuel will be identified for subsequent inspection and characterization. Inspection issues will be evaluated and development of fuel inspection criteria will be developed. A high percentage of the fuel types have not been fully characterized. Characterization of each fuel type needs to be accomplished to determine handling and packaging methods and whether the fuel will be suitable for direct dispositioning or require conditioning.

Alternative conditioning methods will be investigated. The three major dispositioning options consist of direct disposal, mechanical disassembly and encapsulation of the HLW, and chemical processing. Direct disposal of the fuel is the simplest alternative since it only involves handling, packaging and transportation to a repository. However, criticality control is a major issue which must be resolved for this to be a viable alternative. Mechanical disassembly consists of removing bulk cladding materials for disposal in shallow land burial and encapsulation of the HLW material (fission products, actinides) for disposal in a repository. This alternative holds potential for significant minimization of the amount of HLW sent to a repository. Chemical processing would involve dissolution or burning of the fuel, perhaps separation of the HLW, and immobilization for disposal in a repository. This alternative holds the greatest potential for achieving a single waste form for dispositioning. A combination of these alternatives may be the most attractive disposal option as determined by the progression of development.

Table III. SPENT FUEL CHARACTERISTICS

Fuel Type	Fuel Matrix	Clad Material	Other Material	U-235 Enrich	Burnup
oxide	SST	Al	Pu	high	H 40-50
alloy	Al	SST	C	low	M 10-40
metal	BeO, MgO	Zr	etc	deplete	L 1-10
hydride	ZrO_2, CaO	none			neg <1
	ThO_2				u unknown
	none				
hydride	none	mix	C, Pu, Mo	H	L
oxide	SST	SST	Ti, Pu	H	M
alloy	Al	Al	Pu	H	H
oxide	BeO, MgO	none	Be, Mg, Y	H	L
			ceramic		
oxide	ZrO_2	Zr	Pu, B	H	H
oxide	ZrO_2, CaO	Zr	ZrO_2, CaO	H	H
			epoxy		
alloy	none	SST	Th, Na, Mo	L	U
	ThO_2		U-233		
oxide	ThO_2, CaO	Zr	Th, CaO, Pu	L	U
	ZrO_2		U-233		
alloy	none	Zr	Na met, Pu	H	L
oxide	SST	Zr	B_4C	H	U
			thermal-		
metal	none	SST	Pu, Na	H	H
metal	Mo	SST	Pu	L	U
oxide	none	none		H	neg
oxide	none	Zr	Be, Pu	L	U
oxide		Zr, SS	Pu	L	U
oxide	Nichrome			H	U
oxide			Pu	L	U
oxide		SST	Pu	L	U

Laboratory and component testing of valid candidate processes will be performed to establish technology feasibility. Pilot plant and hot integrated testing of selected process(es) which meet health and safety, cost effectiveness and waste minimization criteria will be carried out to verify the process(es) prior to a full scale facility being placed in operation.

SODIUM-BEARING LIQUID WASTE TECHNOLOGY

Process operations and decontamination activities at the ICPP have resulted in the accumulation of approximately 1.5 million gallons of radioactively contaminated liquid waste. The chemical composition of these sodium-bearing wastes are given in Table II. The waste is currently stored in stainless steel tanks which are contained in underground concrete vaults. These tanks do not meet either new seismic codes or RCRA requirements for secondary containment. A Consent Order to the State of Idaho's Notice of Noncompliance requires that the waste be depleted from all of the tanks by 2015.

The previous method for disposing of sodium-bearing waste was to calcine it with a blend of radioactive waste from spent fuel reprocessing to dilute the sodium concentration. Sodium-bearing waste cannot be calcined by itself due to the low melting points of sodium and potassium salts relative to the calcination temperature (500°C). However, with the recent decision to curtail fuel reprocessing, this waste will no longer be available for blending with the sodium-bearing waste. It is also possible to calcine this material via the addition of nonradioactive aluminum nitrate, but results in significantly increased calcine waste volumes.

The objective of this program is to develop a sodium-bearing waste processing method which would separate the waste into a sodium-rich low-level fraction and a high-level fraction containing actinides, fission products, and hazardous components. The technology should minimize the volume of high-level waste requiring ultimate immobilization and disposal, as well as being environmentally acceptable and assuring the safety and health of the general public.

Preliminary studies have identified several technologies as possibilities for processing sodium-bearing wastes, including direct solidification methods, neutralization, precipitation, solvent extraction, ion exchange, electrohydrolysis, and freeze crystallization. Technologies applicable to processing sodium-bearing wastes will be identified and defined with respect to operating conditions and equipment requirements. Although these technologies are currently available, they may not be directly applicable for use at ICPP due to incompatibility with facilities and processes. Therefore, possible modifications to make them applicable will be examined. Laboratory tests will be performed for selected processing methods to scope out the technical feasibility of the concepts. Components/subsystems for acceptable options will be identified, procured and tested on a pilot scale. A radioactive demonstration of the selected processing method will be performed.

Since a major portion of the sodium-bearing waste was generated as a result of decontamination activities, this program will also consider alternative decontamination methods. Some potential options, identified through preliminary studies, include abrasion, electropolishing, ultrasonics, carbon dioxide blasting, light ablation, and alternative chemicals. It is intended to develop as many of these as practical to maturity.

CALCINE IMMOBILIZATION TECHNOLOGIES

The objective of the Calcine Immobilization Program is to develop and demonstrate a process to immobilize ICPP HLW calcine in an acceptable form and 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 pretreatment to separate radionuclides and hazardous components, pr[illegible]degassing, densification, robotic areas, and waste for[illegible]tions, and (5) testing of subsystem components in[illegible]grated pilot plant to provide operating parameters needed for full-scale design. The composition of ICPP calcine is given in Table I.

Several technologies have been identified to date that could immobilize calcine; these include vitrification (Nelson, 1991) and glass-ceramic processing (Bolon, 1991). Nonradioactive and radioactive laboratory tests have been carried out to develop glass waste forms for existing calcines (Staples, 1982). 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 glass (Knecht 1990; Vinjamuri, 1992). 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 (Bolon, 1991). Simplified, small-scale calcine retrieval mock-up tests have been run using calcium carbonate as a nonhazardous stimulant (Hendricks, 1981). The work, while not complete, provides confidence that acceptable processes can be developed in a reasonable period of time. If additional pretreatment steps are incorporated, similar waste forms would be used for the separated radionuclides.

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 acceptance preliminary specifications (WAPS) and to establish criteria for pilot scale tests. Non radioactive and radioactive tests will be run to establish feasibility and criteria for component tests.

Calcine retrieval component tests are required to verify new technologies in pneumatic and robotics 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 welding of vessel, densification of calcine-additive mixture to form a glass-ceramic, and packaging and decontamination of the waste form for disposal. The component testing will be carried out in ICPP pilot plants. The results of these tests will be used to select the process components and to design an integrated pilot plant for demonstration tests.

The overall program schedule could result in a record of decision for the full scale immobilization plant in the year 2003 and hot start-up of a production facility in 2014 if projected funding requirements are met.

METAL RECYCLE/WASTE MINIMIZATION

Based on very preliminary information, DOE estimates that about 1.5 million tons of radioactive scrap metal (RSM) are stored at various DOE facilities. There will be further amounts generated as DOE facilities are decontaminated and decommissioned. The current method of storing unsheltered piles of RSM on open ground may be restricted or discontinued in the future. The major options for future disposition of the RSM are beneficial reuse, engineered interim storage and LLW disposal.

The ICPP program addresses RSM management and disposition with emphasis on recycle and beneficial reuse; it includes support for coordination of RSM activities. Supporting program elements described in the ICPP plan include: (1) Source Compilations, (2) Regulations/Criteria, (3) Systems Analysis/Life Cycle Cost Estimates, (4) Decontamination, and (5) Industrial Interfaces/Technology Transfer. The Metal Recycle Program will also involve programs to demonstrate the restricted recycle of radioactive stainless steel (SS). Assessment of SS melting technologies and potential recycle of ICPP SS from the first phase of the Fuel Storage Reracking Project will be addressed.

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