Treatment of Spent Nuclear Fuel with Molten Salts

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Also Know As

- Electrochemical technology
- Electrometallurgical treatment
- Pyroprocessing
- Pyrochemical processing
- Pyrometallurgical processing
- Dry processing
- Etc.
“Pyrochemical reprocessing methods use high temperature oxidation–reduction reactions in non-aqueous media to separate the actinides, U and Pu from the fission products. These methods exploit the differences in the volatilities or thermodynamic stabilities of the compounds of actinides and fission products to achieve the separation. Pyrochemical separations can be achieved by using electrochemical methods instead of chemical equilibrations.”

From PYROCHEMICAL Reprocessing of Fast Reactor Fuels on IGCAR Website
Work being performed in the United States is focused on an electrochemical process involving molten salts and liquid metals.

Work is performed primarily at Idaho National Laboratory and Argonne National Laboratory.

This talk will focus on that technology and its development.
Electrochemical Processing Background

- Present generation of technology for recycling or treating spent fuel started in the 1980s.
- Electrochemical processes were developed for the fast reactor fuel cycle.
  - The fast reactor fuel does not require a high degree of decontamination.
- Reasons technology pursued include:
  - Capable of low purity products
  - Potential compactness (co-location with reactor)
  - Resistance to radiation effects (short-cooled fuel can be processed)
  - Criticality control benefits
  - Compatibility with advanced fuel types
- Process was integrated with remote fuel fabrication for fast reactors.
Melt Refining Performed Prior to Electrochemical Processing

Technology deployed in the late 1960s

- EBR-II Reactor
- Spent Fuel Assembly
- New Fuel Assembly

Fuel Cycle Facility

- Clean Assembly (Remove Sodium)
- Disassemble & Remove Elements

- Re-cast pins, separate VYCOR and inspect
- Clad Fuel, Leak Test & Bond Test
- Construct Assembly and Test

- Melt Refining Furnace (1400 °C)
- Rejected Elements

- Scrap Fuel Alloy
- VYCOR scrap

- 235U to enrich
- Skull Oxidation

- U & Pu from oxide
- Crucible & Fission Products to Waste

- New Fuel Assembly
- Scrap Fuel Alloy

- Skilled & Bond Test
- Clean Assembly (Remove Sodium)

- VYCOR scrap
Chemical Bases for Electrochemical Processing

(Kcal per equivalent)

-90
-80
-70
-60
-50
-40
-30
-20
-10

Fe, Cd
Zr

U
Pu

Metal Waste
Uranium Product
Ceramic Waste

Hg, Mo, W, Ni, H, Zn, Cr, V, Mn, Be, Np, Sc, Mg, Ce, Pr, La, Ca, Na, Sr, Li, K, Ba, Cs, Rb
Electrorefining

Electrorefiner Cutaway

Fuel Dissolution Basket (Anode)

Uranium Deposit (Cathode)
Cathode Processing

- Salt must be separated from recovered actinides.
- Salt is distilled and recycled back to electrorefiners.

Process Crucibles

Cathode Processor
Electrochemical Treatment of EBR-II Spent Fuel

- **EBR-II used a sodium-bonded metallic fuel.**
  - Reactive materials (sodium and uranium metal)
  - Highly enriched uranium in driver fuel (53-78% U-235)
  - Approximately 1% plutonium in blanket fuel

- **Technology was formally demonstrated with spent fuel between 1996 and 1999.**

- **Demonstration was reviewed for the National Research Council.**
  - *Finding:* The Committee finds that ANL has met all of the criteria developed for judging the success of its electrometallurgical demonstration project.
Mission of Electrochemical Processing

- Until 1994, focus was on demonstration of closed fuel cycle with a fast reactor.
  - Recycle of fast reactor fuel
  - Limited work on production of fast reactor feed material from LWRs
- In 1994, activities were redirected to treatment for disposal.
  - Engineering-scale experience was gained with spent fuel.
- With the formation of the Advanced Fuel Cycle Initiative (AFCI) in 2002 and the Global Nuclear Energy Partnership (GNEP) in 2006, recycle focus was renewed and stressed.
The Fuel Conditioning Facility (FCF) is a one-of-a-kind electrochemical processing facility. FCF is used for treatment of fuel and demonstration of electrochemical process operations. Work includes development, qualification, and production of high-level waste. High-level waste work is performed in the Hot Fuel Examination Facility (HFEF).
Processing Lines in Fuel Conditioning Facility
Electrochemical Treatment of Spent Fuel

- Electrochemical processing has been performed on the engineering-scale with irradiated fuel since 1996.
- Approximately 3.5 MTHM of fuel, including highly enriched uranium fuels, have been treated.
- Installed process equipment could support throughputs between 3 and 5 MTHM per year.
- Advancements in the technology are a major focus of Advanced Fuel Cycle Initiative.
Throughout development of electrochemical processing, electrorefining has been scaled significantly for hot cell application.

- First hot cell electrorefiner has a current capacity of 3.5 amps.
- Mark IV electrorefiner has a current capacity of 200 amps.
- Mark V electrorefiner has a current capacity of 2400 amps.
- Technology has been scaled in cell by three orders of magnitude.
Electrochemical Processing Accomplishments

- Group transuranic recovery tests have been successfully performed at laboratory and engineering-scales.
  - Recoveries of more than 1 kg of transuranics have been obtained.
  - Purity requirements for fast reactor fuel have been demonstrated at both scales.

- Engineering-scale electrorefining operations continue to demonstrate high dissolution of actinides (99.7%).
Electrochemical Processing Accomplishments

- Electrochemical recovery of zirconium has been demonstrated.

- Improved crucible materials for high temperature processing have been successfully tested at engineering-scale.
  - Crucibles are reusable, which supports increased throughput.
  - Very little dross forms in new crucibles.
Metallic Fuels

- Process development work has focused on metallic fuels.
- Metallic fuels are amendable to remote fabrication.
- Metallic fuels are capable of high burnups.

Injection Casting of Fuel

Cast Fuel Pins

Cast Pin with Transuranics
Electrolytic Reduction Process Scale-Up

- Pre-conceptual layouts of an engineering-scale electrolytic reduction vessel (20 to 40 kg oxide fuel batch size) based on a planar electrode configuration have been generated.

- A pre-conceptual design of a facility to treat spent oxide fuel has been prepared to help focus research on critical activities.
### Chemical Bases for Electrochemical Processing

#### (Kcal per equivalent)

- **Metal Waste**
  - Fe
  - Cd
  - Zr

- **Uranium Product**
  - U
  - Pu

- **Ceramic Waste**
  - Hg
  - Mo
  - W
  - Ni
  - H
  - Zn, Cr
  - V
  - Mn, Be
  - Np
  - Sc
  - Mg
  - Cr, Am
  - Nd
  - Ce
  - Pr, La
  - Ca, Na
  - Sr
  - Li
  - K
  - Ba, Cs, Rb
Two high-level wastes are produced from electrochemical processing.

A zeolite-based ceramic waste stabilizes fission products that form chlorides.

A stainless-steel-15% zirconium metal waste stabilizes the cladding hulls and more noble fission products.
Ceramic Waste Process

1. **Electrorefiner**
   - **Electrorefiner Salt**
   - **Crushed Salt**

2. **Salt Crusher**
   - **Ground Salt**

3. **Heated V-Mixer**
   - **Dried Zeolite**
   - **Glass and Salt-Loaded Zeolite**

4. **Mill/Classifier**
   - **Milled Zeolite**

5. **Zeolite Mill**

6. **Furnace**
   - **Ceramic Waste**
     - Glass added after producing salt-loaded zeolite
Installation was completed in March 2005.
Process development testing is ongoing.
Maximum operating temperature is 1025 °C.
Dimensions of the internal cylindrical cavity are:
   - Diameter of 0.6 m (2 ft)
   - Length of 3 m (10 ft)
System is capable of processing waste forms in excess of 320 kg.
Metal Waste Form Process

- Cladding Hulls
- Induction Furnace
- Salt
- Electrorefiner
- Ceramic Waste
- Zirconium
- Ingot
Metal Waste Development Activities

- The Prototype Metal Waste Furnace is installed in a large inerted glovebox for processing testing.
- A series of test runs were completed to show equivalency of the Prototype to the Production Metal Waste Furnace in mockup.
- The Prototype furnace is being used to:
  - Finalize the full-scale production process (≤90 kg) which will be used in cell
  - Demonstrate the reliability of each process step, including sodium oxidation, salt distillation, and ingot formation prior to installation of the Production Furnace in a hot cell
  - Validate acceptability of the produced ingots through 5 qualification runs
Waste Form Qualification

- Waste forms were extensively characterized for qualification.
- Waste form degradation models were developed for each waste form.
- Repository performance assessment calculations to assess the impact of the waste forms on the repository were performed.
- Report prepared for US Congress addressing the disposal of the waste forms.
  - DOE-NE, DOE-RW (Office of Civilian and Radioactive Waste Management), and DOE-EM (Environmental Management) concurred on disposal plans.
- Waste forms are now classified formally as high-level wastes in DOE orders.
HLW Data Package

A data package was compiled for the ceramic and metal waste forms. This document included 71 reports containing data and analyses needed to support acceptance into Yucca Mountain. Issues addressed in the reports include:

- waste form specifications
- waste form characteristics and degradation data,
- production information
- radiation effects
- product consistency
- impact of processing on waste characteristics
- leach behavior
- radionuclide distribution
- modeling of degradation behavior
International Collaborations

- International collaborations on aspects of electrochemical processing have occurred with
  - Japan
  - Korea
  - France
  - United Kingdom
  - Australia
Summary

- Electrochemical fuel cycle can be used for recycle of fast reactor spent fuel and treatment of thermal reactor spent fuel to produce a feed for fast reactors.
- Electrochemical processing of spent metallic nuclear fuel has been successfully scaled by more than three orders of magnitude in a hot cell.
- Critical infrastructure exits domestically to support development and demonstration of technology.
- Work activities are focused on research, development, and demonstration in support of Advanced Fuel Cycle Initiative.
- Development and qualification of high-level wastes are integrated with overall process development.