
Fiscal Years 2007 and 2008

Nuclear Engineering Division
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U.S. Department of Energy and Nuclear Regulatory Commission
– Advanced Fuel Cycle Research and Development Seminar Series

Fiscal Years 2007 and 2008

Compiled by
Christopher Grandy
Nuclear Engineering Division
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August 2008
Overview

In fiscal year 2007, the Advanced Burner Reactor project initiated an educational seminar series for the Department of Energy (DOE) and Nuclear Regulatory Commission (NRC) personnel on various aspects of fast reactor fuel cycle closure technologies. This important work was initiated to inform DOE and NRC personnel on initial details of sodium-cooled fast reactor, separations, waste form, and safeguard technologies being considered for the Advanced Fuel Cycle Research and Development program, and to learn the important lesson from the licensing process for the Clinch River Breeder Reactor Plant that educating the NRC staff early in the regulatory process is very important and critical to a project success.

The following was the initial list of proposed training seminars from which DOE and NRC personnel could make a selection:

1. **Overview and Sodium Technology Background**
   a. Introduction to LMR Technology; LMR experience, designs, performance, operational issues, safety issues, US and international, status. US R&D program and experience.
   b. Sodium technology, chemistry, thermal-hydraulic performance, corrosion of steel, oxidation.

2. **Physics and Thermal-Hydraulics Performance**
   a. Fast reactor physics performance; breeding, burning, mass loadings/inventory, coolant void worth, and reactivity coefficients.
   b. Fast reactor thermal-hydraulic performance; temperatures, heat fluxes, forced convection, natural circulation, etc.

3. **Fuel Performance Experience**
   a. Fuel performance - general; metal and oxide
   b. Fuel irradiation experience and results; EBR-II, FFTF
   c. TREAT testing results for metal and oxide fuel
   d. Fabrication & standards history/current capabilities

4. **Structures, Systems, and Components #1**
   a. Major sodium component technology; pumps, heat exchangers, vessels, refueling systems design and performance, etc.
   b. Structural materials for SFR technology and their performance
   c. I&C, fire protection, sodium leak detection, etc.
   d. Prior and current operating experience (availability?)

5. **Structures, Systems, and Components #2**
   a. Control and protection systems designs and performance
   b. Containment systems designs and performance
   c. Seismic isolation systems and reactor applications
   d. Prior and current operating experience (availability?)

6. **Safety Issues #1**
   a. Safety issues; DBA, BDBA, severe accident historically (coolant void, recriticality, FCI), passive safety
   b. Safety analysis past results (FFTF, CRBRP, SAFR, PRISM)
c. Safety analysis methods; reactor, structural, coolant aerosols, containment

7. Safety Issues #2
   b. Inherent passive safety characteristics of sodium fast reactor systems (with respect to loss of flow without scram, etc.). Inherent reactivity shutdown, natural circulation decay heat removal
   c. Safety testing results, EBR-II shutdown heat removal test, FFTF unprotected loss of flow (Gas Expansion Modules); TREAT

8. Codes, Modeling, Analyses
   a. Computer Codes Used for Transient and Accident Analyses of Liquid Metal Reactors
   b. Mechanistic Source Term Modeling for Liquid Metal Reactors
   c. General – status of other codes, modeling, analyses; need for updating or new applications; access to data and codes, etc.

9. Licensing Strategy/Framework
   a. DOE – Applicant/Licensee
      • Relationship
      • Responsibilities
      • Contractual Requirements
   b. Applicability of DOE Orders
   c. NRC Regulatory Requirements
      • Technical Requirements
      • Material, SSC Specifications
      • Safety Program
      • QA Program
      • Fuel Technical Criteria and Qualification
   d. Recycle/Fabrication/Reactor Operations/Safety/Quality Interactions

10. Nuclear Fuel Separations Processes
    a. Introduction to nuclear fuel separations and chemical processes
    b. Aqueous separations processes for spent nuclear fuel treatment
       • PUREX and its modifications
       • UREX+ family of processes
    c. Electro-chemical processing of spent nuclear fuel
    d. International and domestic experience in processing of spent nuclear fuel

11. Advanced Modeling and Simulation - Overview
    a. GNEP approach to advanced modeling and simulation
    b. Modeling and Simulation Campaign
    c. Status of FY07 work on modeling and simulation
    d. Future directions in advanced modeling and simulation

12. Reactor – Advanced Modeling and Simulation
    a. Advanced Modeling and Simulation – T/H
b. Advanced Modeling and Simulation – Structural
 c. Reactor code framework
 d. Neutronics modeling

13. Advanced fuel cycle safeguards:
   a. Introduction and current technical basis
   b. Reprocessing
   c. Fuel Fabrication
   d. Fast Reactor

For each of the advanced fuel cycle technologies the current state of the art in safeguards approach and technical means is discussed, followed by challenges presented by AFCI/GNEP (both physics based and rule based). Presentation would provide a) fundamentals of technology (current and needed) and b) areas where NRC would need to address shortcomings from laboratory technical perspective (including DOE perspective). The balance between technical content and safeguards approaches can be tailored to their needs and interest.

14. Waste Forms
   a. Integrated Waste Management Strategy
   b. Candidate Waste/Storage Forms from Aqueous Processing
   c. Candidate Waste/Storage Forms from Electrochemical Processing
   d. Current Work in Technology Development
   e. Long-Term S&T Plan and Role of Modeling and Simulation

Given the above list of seminar topics, DOE and NRC personnel selected a number of seminars that were presented to both DOE and NRC personnel in Fiscal Years 2007 and 2008. The topics and dates of the presentations are given in Table 1.

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1 The dates shown in this table are the dates the presentations were given to NRC personnel. In general, the presentations were given to DOE personnel the day prior to the NRC seminar date.
<table>
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<th>Tag#</th>
<th>Topic</th>
<th>Presenter</th>
<th>Date</th>
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<td>Introduction to Fast Reactor Technology – An Overview(^2)</td>
<td>Chris Grandy / James Cahalan</td>
<td>March 21, 2007</td>
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<tr>
<td>2</td>
<td>Sodium as a Fast Reactor Coolant</td>
<td>Thomas Fanning</td>
<td>May 3, 2007</td>
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<tr>
<td>4</td>
<td>Sodium Fast Reactor – Safety Issues #1</td>
<td>Jim Cahalan</td>
<td>June 21, 2007</td>
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<td>5</td>
<td>Sodium Fast Reactor – Safety Issues #2</td>
<td>Jim Cahalan</td>
<td>October 31, 2007</td>
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<td>6</td>
<td>SFR Fuel Performance and Approach to Qualification</td>
<td>Steven Hayes and Douglas Porter</td>
<td>November 28, 2007</td>
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<td>7</td>
<td>Spent Nuclear Fuel Reprocessing</td>
<td>Terry Todd</td>
<td>March 25, 2008</td>
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<tr>
<td>8</td>
<td>UREX+ Process Overview</td>
<td>Candido Pereira</td>
<td>March 26, 2008</td>
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<td>9</td>
<td>Electrochemical Processing of Spent Nuclear Fuel</td>
<td>Michael Goff</td>
<td>March 26, 2008</td>
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<td>10</td>
<td>Overview of Systems Analysis and Waste Form Campaigns</td>
<td>Kathryn McCarthy</td>
<td>April 23, 2008</td>
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<td>12</td>
<td>GNEP Waste Form Technology Development</td>
<td>John Vienna</td>
<td>April 23, 2008</td>
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<td>13</td>
<td>Advanced Fuel Cycle Safeguards</td>
<td>Michael Miller</td>
<td>June 10, 2008</td>
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\(^2\) This presentation was removed from this report to aid in the publication clearance process.
Acknowledgement

The participants would like to thank the U.S. Department of Energy’s Global Nuclear Energy Partnership’s Advanced Fuel Cycle Research and Development program for sponsoring and supporting this important work. Specifically, the seminars were hosted and sponsored by the Federal Project Director, Fast Reactors and his staff for which we are truly grateful.

In addition, we would like to thank the hard work and dedication of all the presenters who spent evenings and weekends preparing their presentation materials for this seminar series. All of the presentations were very well received by all participants.
Sodium as a Fast Reactor Coolant

Presented by

Thomas H. Fanning
Nuclear Engineering Division

U.S. Department of Energy
U.S. Nuclear Regulatory Commission
Topical Seminar Series on Sodium Fast Reactors
May 3, 2007
Background

- Unless new sources of energy are found, the development of nuclear power will necessarily depend on fast reactors.
  - Fast reactors are not a new technology.
  - Fast reactors have achieved well over 300 reactor-years of operation.

- To support effective actinide management (as envisioned by GNEP) a fast reactor must have a compact core with a minimum of materials which absorb or moderate fast neutrons. This places a significant heat transfer requirement on the coolant.

- The choice of coolant determines the main design approaches and the technical and economic characteristics of a nuclear power plant.
  - Historically, this requirement has been met by the use of sodium in nearly all fast reactors constructed and operated.
  - The Generation IV International Forum (GIF) identified three fast reactor concepts for potential future development: SFR, LFR, and GFR.
Objectives

- Identify important differences between sodium and other fast reactor coolants (lead/LBE and helium) and why sodium is preferred.
- Characterize the nature of sodium interactions with air and water.
- Understand how differences between sodium and water result in broad design differences between a sodium-cooled fast reactor (SFR) and a light-water reactor.
Topics to be Covered

- **Historical Perspective on Reactor Coolants**
  - Coolants that have been used in the past
  - Coolants for future fast reactors: sodium, lead, helium

- **Comparison of Sodium with other Fast Reactor Coolant Options**
  - Thermophysical properties
  - Material properties
  - Neutronic properties
  - Safety
  - Cost
  - Other issues

- **Sodium Reactivity with Air and Water**

- **Impact of Coolant on SFR and LWR Differences**
  - Operating Pressure
  - Thermal Margins
  - Plant Design

- **Summary**
Historical Perspective on Reactor Coolants
Historical Perspective on Reactor Coolants

In the 1950s and 1960s, scientists and engineers considered (and in many cases built) nearly everything imaginable at the time:

- Water (light, heavy)
- Liquid-metal (NaK, sodium, lithium, mercury, rubidium, lead, bismuth, lead-bismuth, gallium, tin, etc. and numerous other alloys)
- Gas (air, argon, carbon dioxide, helium, hydrogen, nitrogen)
- Fluid Fuel (aqueous: UO$_2$/phosphoric acid, U(SO$_4$)$_2$, UO$_2$SO$_4$/ThO$_2$; molten salt: NaF-BeF$_2$-UF$_4$, LiF-BeF$_2$-ZrF$_4$-UF$_4$/FLiBe; liquid metal: U-Bi)
- Organic (polyphenyls/terphenyls, kerosene, Santowax)

Combinations of coolant and moderator were also studied:

- Sodium-cooled, graphite moderated (SRE, Hallam)
- Organic-cooled, heavy water moderated (Whiteshell 1, ESSOR)
Historical Perspective on Fast Reactors

- Data from IAEA Fast Reactor Database. Does not include submarine (S1G/S2G) or space reactors (SNAP).

- More recent fast reactors under construction:
  - CEFR (China) 2008 25 MWe
  - PFBR (India) 2010 500 MWe
  - BN-800 (Russia)

- China has an ambitious vision to deploy 200 GWe of sodium-cooled fast breeder reactors by 2050 (12% of projected capacity).

<table>
<thead>
<tr>
<th>Facility</th>
<th>Country</th>
<th>First Critical</th>
<th>Coolant</th>
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<tbody>
<tr>
<td>Clementine</td>
<td>USA</td>
<td>1946</td>
<td>Mercury</td>
</tr>
<tr>
<td>EBR-I</td>
<td>USA</td>
<td>1951</td>
<td>NaK</td>
</tr>
<tr>
<td>BR-2</td>
<td>Russia</td>
<td>1956</td>
<td>Mercury</td>
</tr>
<tr>
<td>BR-5/BR-10</td>
<td>Russia</td>
<td>1958</td>
<td>Sodium</td>
</tr>
<tr>
<td>DFR</td>
<td>UK</td>
<td>1959</td>
<td>NaK</td>
</tr>
<tr>
<td>Fermi</td>
<td>USA</td>
<td>1963</td>
<td>Sodium</td>
</tr>
<tr>
<td>EBR-II</td>
<td>USA</td>
<td>1963</td>
<td>Sodium</td>
</tr>
<tr>
<td>Rapsodie</td>
<td>France</td>
<td>1967</td>
<td>Sodium</td>
</tr>
<tr>
<td>BOR-60</td>
<td>Russia</td>
<td>1968</td>
<td>Sodium</td>
</tr>
<tr>
<td>SEFOR</td>
<td>USA</td>
<td>1969</td>
<td>Sodium</td>
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<tr>
<td>KNK-II</td>
<td>Germany</td>
<td>1972</td>
<td>Sodium</td>
</tr>
<tr>
<td>BN-350</td>
<td>Kazakhstan</td>
<td>1972</td>
<td>Sodium</td>
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<tr>
<td>Phenix</td>
<td>France</td>
<td>1973</td>
<td>Sodium</td>
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<tr>
<td>PFR</td>
<td>UK</td>
<td>1974</td>
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<td>FFTF</td>
<td>USA</td>
<td>1980</td>
<td>Sodium</td>
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<tr>
<td>BN-600</td>
<td>Russia</td>
<td>1980</td>
<td>Sodium</td>
</tr>
<tr>
<td>JOYO</td>
<td>Japan</td>
<td>1982</td>
<td>Sodium</td>
</tr>
<tr>
<td>FBTR</td>
<td>India</td>
<td>1985</td>
<td>Sodium</td>
</tr>
<tr>
<td>Super-Phenix</td>
<td>France</td>
<td>1985</td>
<td>Sodium</td>
</tr>
<tr>
<td>MONJU</td>
<td>Japan</td>
<td>1995</td>
<td>Sodium</td>
</tr>
</tbody>
</table>
Historical Perspective on Sodium

- Liquid metals in general have received the most attention for fast reactor applications because of their high thermal conductivity, indifference to radiation, and useful temperature range at low pressure.
  

- Sodium received so much additional attention, that a supplement to the Liquid-Metals Handbook was published in 1955


- Numerous additional reports and monographs have been published since. More recently, during the IFR program:
  
Historical Perspective on LBE

- In the U.S., use of lead/LBE was dismissed early mainly due to high pumping power requirements.

- A program for studying compatibility of lead, bismuth, and their alloys with structural materials existed at BNL between 1950 and 1962 as part of the Liquid Metal Fuel Reactor program.
  - Focus was on development/testing of a natural U-Bi liquid metal fuel.
  - An in-pile liquid-metal fuel loop was constructed and tested in the 1950s.

- Recent interest in lead and LBE developed as part of their application to accelerator-driven systems as target/coolant materials.

- The only LBE-cooled reactors are those developed for the Soviet Project 705 (Alfa-class) submarines
  - Modified November-class (Project 645) commissioned in 1963 to test reactors intended for Alfa-class submarines.
  - Initial prototype (Project 661/Papa Class) commissioned in 1969.
Historical Perspective on LBE (continued)

- Two different reactor designs were used in the Alfa-class submarines: OK-550 and BM-40A.
  - Significant effort invested to solve the problem of LBE corrosion.
  - Detailed information on the results of Russian LBE testing is difficult to find in the literature.
- First vessel was decommissioned in 1987, with four more by 1992. At least one vessel was refitted with a VM-4 PWR and used for training.
- Reactors were single-use designs with HEU and a long core lifetime (up to 15 years)
  - Coastal facilities were constructed to provide superheated steam during shutdown and maintenance to keep the LBE molten. These proved to be unreliable and completely broke down in the 1980s.
  - Reactors were kept running even while in harbor, and significant maintenance became impossible.
  - Reactor failures and coolant leaks led to a number of fatalities.
Historical Perspective on Helium

- No gas-cooled fast reactors have ever been constructed
  - GA led efforts to develop a gas-cooled fast breeder reactor in the 1970s.
- Most early gas-cooled reactors were CO\textsubscript{2} cooled, graphite moderated, natural uranium reactors developed primarily for plutonium production (mainly in UK and France)
- Vast majority of currently-operating gas-cooled reactors use CO\textsubscript{2} for coolant (UK: 14 AGR, 8 Magnox)
- Helium-cooled thermal power reactors (using enriched uranium) include:
  - China: HTR-10 (2000/3 – Present)
Properties of Fast Reactor Coolants: Sodium, Lead/LBE, and Helium
**Coolant Criteria**

- **Thermophysical Properties:**
  - Excellent heat transfer
  - Low vapor pressure
  - High boiling point
  - Low melting point

- **Material Properties:**
  - Thermal stability
  - Radiation stability
  - Material compatibility

- **Neutronic Properties:**
  - Low neutron absorption
  - Minimal induced radioactivity
  - Negligible moderation

- **Support Passive Safety**

- **Cost:**
  - Initial inventory
  - Make up inventory
  - Low pumping power

- **Hazards:**
  - Non-toxic
  - Non-reactive
## Thermophysical Properties for Fast Reactor Coolants

<table>
<thead>
<tr>
<th>Property</th>
<th>Na</th>
<th>Pb</th>
<th>LBE</th>
<th>He</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atomic Weight</td>
<td>22.997</td>
<td>207.21</td>
<td>208</td>
<td>4</td>
</tr>
<tr>
<td>Melting Point (°C)</td>
<td>97.8</td>
<td>327.4</td>
<td>123.5</td>
<td>n/a</td>
</tr>
<tr>
<td>Boiling Point (°C)</td>
<td>892</td>
<td>1737</td>
<td>1670</td>
<td>-267</td>
</tr>
<tr>
<td>Density (kg/m³)</td>
<td>880</td>
<td>10500</td>
<td>10300</td>
<td>0.178</td>
</tr>
<tr>
<td>Specific Heat (J/kg-K)</td>
<td>1300</td>
<td>160</td>
<td>146</td>
<td>5200</td>
</tr>
<tr>
<td>Heat Capacity (MJ/m³-K)</td>
<td>1.14</td>
<td>1.68</td>
<td>1.50</td>
<td>0.0009</td>
</tr>
<tr>
<td>Thermal Conductivity (W/m-K)</td>
<td>76</td>
<td>16</td>
<td>11</td>
<td>0.152</td>
</tr>
<tr>
<td>Viscosity (cP)</td>
<td>0.34</td>
<td>2.0-2.5</td>
<td>1.7</td>
<td>0.018</td>
</tr>
</tbody>
</table>

Values at STP. *Italic = Evaluated at ~300°C*
**Thermophysical Properties**

- Despite very different densities and specific heat, sodium and LBE have similar volumetric heat capacities.
  - Similar volumetric flow rates
  - For identical flow geometries, similar flow velocity. But LBE will have a significantly higher pressure drop and pumping requirements.
  - LBE will develop slightly higher natural circulation velocity, but at the expense of somewhat higher cladding temperatures.

- Relatively low thermal conductivity of LBE (combined with low flow velocity) affects heat transfer from cladding to coolant.

- Specific heat of helium is high, but density is very low. Requires a high pressure system (~85 atm) and high coolant velocity (~100 m/s). Introduces risk of flow-induced vibrations.

- Low thermal conductivity of helium results in poor heat transfer even at high coolant velocity. Cladding surfaces can be roughened to improve heat transfer (4x), but it is still 8-9x lower than for sodium.
Material Properties

- Sodium, LBE, helium have excellent thermal and radiation stability.
- Compatibility of coolants with available structural, heat exchanger, and pump materials is a key criteria for selection.
  - Helium is chemically inert, but low molecular weight leads to diffusive losses at seals and valves. Impurities (especially moisture) can lead to corrosion.
  - Liquid metals may selectively deplete constituents from, and impurities may chemically react with, structural materials
  - Chemistry control will be required for all coolants.

- Standards for sodium:
Material Properties (Sodium)

- Primary issue with sodium is its chemical reactivity in air and water.
- Sodium is inherently compatible with stainless steels, requiring no special corrosion protection measures
  - Oxygen preferentially reacts with sodium, forming Na₂O.
  - Oxygen impurity can readily be maintained well below 10 ppm using a cold trap.
  - No difference between high-purity sodium and helium when comparing strain-rate and creep-rupture data for austenitic steels. No indication of liquid metal embrittlement.
  - Based on long-term corrosion testing in BR-10, “the operating service life of sodium equipment in fast reactors can be increased to 60 yr and longer.”

- Fuel-coolant interactions are benign for metallic fuel.
- Many fission products are soluble in sodium and can be filtered out in the cold trap.
- Database for sodium compatibility is extensive, with information available in the open literature.
Material Properties (LBE)

- Prior to 1998, material database relied mainly on anecdotal information from Russian sources.
- Solubility of steels in LBE is generally higher than in lead (due to Bi). Solubility of Ni is of particular concern (Ni ~37000 ppm at 600°C).
- Russian approach is to maintain a protective oxide layer on structural components (particularly cladding) and minimize coolant flow velocities.
  - Applicable to ferritic-martensitic steels.
  - Above 550°C, oxide layer can become thick and unstable
  - Oxygen control is a dynamic equilibrium: thickness of oxide layer stabilizes, but structural (cladding) weight loss continues.
  - Coolant velocity and high shear stress (viscosity) results in erosion.
  - Oxide layer impacts heat transfer from cladding to coolant.

Oxide control:

- Dissolution: Fe₃O₄, FeCr₂O₄
- Protective Oxide Layer: PbO
- PbO Sludge

Oxygen in LBE
Oxide Control in LBE Systems

- Oxygen is controlled by constant addition (e.g. flow over PbO “balls”) and subtraction (e.g. bubbling hydrogen gas in a helium carrier to form water).

- Problems with oxygen control:
  - Non-homogeneous oxygen distribution in real systems results in non-uniform coatings. Crevice corrosion and dissolution of occlusions can occur.
  - Magnetite (Fe₃O₄) undergoes a phase transformation at 570°C.

- Newer solution is to prefer SiO₂ and Al₂O₃ based oxide layers.
  - Oxide layer forms at lower oxygen concentrations.
  - No phase transformation.
  - EP-823 (Russian steel similar to HT-9 but with higher Si content).
  - Silicon degrades mechanical properties (ductility) and reduces irradiation performance.
  - Some interest in oxide dispersion strengthened (ODS) steels. Irradiation performance and cost are not well known.
Neutronic Characteristics

- Sodium, Lead, LBE, and Helium are all suitable for fast reactor applications
  - Sodium and LBE both introduce a small amount of parasitic absorption.
  - Reduced neutron leakage in an LBE system allows for a higher coolant volume fraction.
  - Helium is transparent to neutrons, and the relatively low density leads to negligible moderation, but this leads to a higher neutron leakage fraction.
Sensitivity of Neutron Flux to Varying Parameters

Average Experimental Fast Flux, Center

Equivalent Core Diameter (cm)

He, Standard (U)  He, Standard (Pu)  He, Compact (U)  LBE, 61-Pin (U)
Na, 91-Pin (U)  Na, 61-Pin (U)  Na, 91-Pin (Pu)  LBE, 91-Pin (U)

$D = 119 \text{ cm}$
Sensitivity of Enrichment to Varying Parameters

Equivalent Core Diameter (cm)

Fissile Enrichment (%)

- He, Standard (U)
- He, Standard (Pu)
- He, Compact (U)
- LBE, 61-Pin (U)
- Na, 91-Pin (U)
- Na, 61-Pin (U)
- Na, 91-Pin (Pu)
- LBE, 91-Pin (U)
Scaling to Higher Power

- When comparing sodium and LBE designs with identical geometries, LBE:
  - Requires a slightly lower enrichment
  - Can produce a higher fast flux
  - Results in a lower fissile inventory.

- Use of identical geometries does not account for optimization based on individual coolant properties

- 250 MW\textsubscript{th} options for sodium and LBE were evaluated to assess more realistic core designs.
  - Additional constraint on fissile uranium enrichment: < 20\% U-235 (LEU)
  - Coolant velocity limit (2 m/s) imposed on LBE to limit erosion.
  - Core designs were adjusted to achieve similar experimental flux levels.
Scaling to Higher Power

- Sodium coolant results in a smaller core with higher power density, lower enrichment, fewer driver assemblies, and lower heavy metal inventory.

<table>
<thead>
<tr>
<th></th>
<th>Sodium</th>
<th>LBE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal Power (MW)</td>
<td>250</td>
<td>250</td>
</tr>
<tr>
<td>Coolant Velocity (m/s)</td>
<td>8.3</td>
<td>2.0</td>
</tr>
<tr>
<td>Average Experimental Fast Flux ($10^{15}$ n/cm²/s)</td>
<td>1.83</td>
<td>1.79</td>
</tr>
<tr>
<td>Number of Driver Assemblies</td>
<td>135</td>
<td>237</td>
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<tr>
<td>Pins per Assembly</td>
<td>91</td>
<td>61</td>
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<tr>
<td>Equivalent Core Diameter (cm)</td>
<td>119</td>
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</tr>
<tr>
<td>Active Height (cm)</td>
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<td>100</td>
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<tr>
<td>Fissile Enrichment (%)</td>
<td>18.3</td>
<td>20.0</td>
</tr>
<tr>
<td>Heavy Metal Inventory (kgHM)</td>
<td>5138</td>
<td>5716</td>
</tr>
</tbody>
</table>

- Sodium case still has room for further optimization.
Coolant Activation

- Coolant activation results in radioactive isotopes circulating through the primary loop.
  - Helium yields no activation products unless impurities are present.
  - Sodium: \( ^{23}\text{Na}(n,\gamma)^{24}\text{N} \) (\( t_{1/2} = 15 \) hours)
  - LBE: \( ^{209}\text{Bi}(n,\gamma)^{210}\text{Bi}, \quad ^{210}\text{Bi} \rightarrow ^{210}\text{Po} \) (\( t_{1/2} = 138 \) days)

- Operationally, this requires shielding for primary components.
- Po-210 represents a significant biological hazard, requiring a leak-proof system.
  - \( \text{PbPo}(s) + \text{H}_2\text{O} = \text{PbO} + \text{H}_2\text{Po}(g) \) (volatile alpha-emitting aerosol)

- Estimates of cool-down time to meet the IAEA “exemption” criteria (to be freely used for other industrial purposes):
  - Sodium (pure): ~7 years
  - Sodium (with impurities): 50-100 years
  - LBE: ~100,000 years

- Experience exists for processing large quantities of primary sodium coolant for disposal (EBR-II).
## Passive Safety (Liquid Metals)

- Heat capacity and high thermal conductivity of liquid-metal coolants provides large thermal inertia against system heating during loss-of-flow accidents.
  - Combined with favorable reactivity feedback (core design/fuel choice) sufficient cooling is available to support passive shutdown.
  - Full-power unprotected loss of flow and loss of heat sink accidents demonstrated in EBR-II in 1986. Similar tests (at 50% power) demonstrated passive response in FFTF.

- **Response of lead/LBE systems to seismic events will pose design challenges.**
  - For medium-sized plant, one can expect primary coolant alone to weigh ~10,000 metric tons.
  - Structural design of primary system becomes a significant challenge.
**Passive Safety (Demonstrated in EBR-II in 1986)**

- Unprotected (no scram) Loss of Flow (ULOF) results in brief temperature rise which is terminated by negative reactivity feedback effects.

- Unprotected Loss of Heat Sink (ULOHS) causes increase in inlet temperature which introduces negative reactivity feedback effects.
Passive Safety (Helium-Cooled Fast Reactor)

- In gas-cooled thermal reactors, graphite moderator provides a large heat capacity against excessive temperature rises. Not present in a fast reactor.
- Low thermal conductivity of helium coolant minimizes thermal shock during transients.
  - Thermal transients do not propagate to structural materials rapidly. Heat is deposited in the fuel.
  - Despite minimal thermal shock, power-to-flow mismatches may be a significant safety concern for gas-cooled fast reactors.
- GFR relies on secondary vessel/containment to limit pressure loss.
  - May be able to support decay heat removal, but not full-power transients.
  - Adequate decay heat removal may not be possible under loss of pressure scenarios.
Coolant Cost

■ Direct cost
  – Sodium: On a volume basis, it is generally the cheapest of all metals, and is among the most abundant elements in the earth’s crust. 2006 price from DuPont (reactor grade/Niapure™) is $1.58/lb ($3400/m³).
  – Lead: 2005 USGS, 43 – 61 ¢/lb ($15,000/m³).
  – Bismuth: U.S. ceased domestic production in 1997 and is highly dependent on imports. By the end of 2005, USGS-tracked price had increased to >$4/pound ($86,000/m³).
  – The Helium Privatization Act of 1996 mandates the price of helium. In 2005, Government price was 1.965 $/m³. Private prices ranged from 2.42 to 2.63 $/m³. At 85 atm, this is roughly $220/m³.

■ Indirect costs
  – Coolant chemistry/purification, inventory makeup (helium).
  – Pumping requirements.
  – Core and primary system size.
  – Component lifetime.
  – Passive safety and safety-related systems.
Other Considerations

- Component Technology R&D
  - Regardless of coolant, component testing will be required.
  - 50+ years of sodium component development, testing, and operation.

- Fuel handling
  - Opaqueness of sodium and lead/LBE affects fuel handling.
  - Need to maintain pressure boundary with helium also affects fuel handling.
  - Must be able to remove residual coolant from spent fuel or test assemblies.
  - Significant experience exists for “washing” sodium from spent fuel with steam or moist air.
  - Removing LBE from spent fuel may require an acidic “brew” or boiling in glycerin. Sodium may be used, but it will also remove surface oxides.
Sodium as a Fast Reactor Coolant

- Thermophysical and thermal-hydraulic properties of sodium are superior to lead or helium.
  - Smaller core with higher power density, lower enrichment, and lower heavy metal inventory. Demonstrated passive safety performance.
  - Use of sodium codified in ASTM standards.

- Extensive testing of coolants lead to nearly all (land-based) fast reactors constructed during the last 50 years using sodium as the primary coolant.
  - Current fast reactor construction projects use sodium as the primary coolant.
  - LBE-cooled reactors limited to Russian Alfa-class submarine experience.
  - No He-cooled (or gas-cooled) fast reactors.

<table>
<thead>
<tr>
<th>Thermophysical Properties:</th>
</tr>
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<tbody>
<tr>
<td>Excellent Heat Transfer ✓+</td>
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<tr>
<td>Low Vapor Pressure ✓+</td>
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<tr>
<td>High Boiling Point ✓+</td>
</tr>
<tr>
<td>Low Melting Point ✓</td>
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<table>
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<tr>
<th>Material Properties:</th>
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<tr>
<td>Thermal Stability ✓+</td>
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<tr>
<td>Radiation Stability ✓+</td>
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<td>Material Compatibility ✓+</td>
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<tr>
<th>Neutronic Properties:</th>
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<tr>
<td>Low Neutron Absorption ✓+</td>
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<tr>
<td>Minimal Activation ✓</td>
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<tr>
<td>Negligible Moderation ✓+</td>
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<table>
<thead>
<tr>
<th>Supports Passive Safety ✓+</th>
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<table>
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<tr>
<th>Cost:</th>
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<tbody>
<tr>
<td>Initial Inventory ✓+</td>
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<tr>
<td>Make-Up Inventory ✓+</td>
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<tr>
<td>Low Pumping Power ✓+</td>
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<table>
<thead>
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<th>Hazards:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sodium reacts with air and water (next)</td>
</tr>
</tbody>
</table>
Sodium Reactivity with Air and Water
Sodium Reactivity with Air

- Burning reaction is characterized by a zone of small (few cm) flames at the sodium-air interface, formation of Na₂O on the surface, and vigorous emission of oxide fumes.

- Sodium reaction with air is slow and causes relatively low heat release
  - Sodium has a high latent heat of vaporization and high boiling temperature.
    - *This results in a low evaporation rate during combustion.*
    - *Gasoline will burn approximately 4 times faster than sodium*
  - Sodium-air heat of reaction is one-fourth that of burning gasoline.
    - *Energy released during sodium burning is approximately a factor of 15 lower than in the case of gasoline.*
  - In one series of tests, temperature 1 meter above a 1 square meter pool of burning sodium was less than 100°C.
  - For gasoline, the flame zone extended as high as 4 meters and the average temperature 2 meters above the pool exceeded 600°C.
**Sodium Reactivity with Air**

- Sodium aerosols (NaO and Na$_2$O) react with air (including water vapor and CO$_2$) to produce NaOH, and Na$_2$CO$_3$.
  - Production of NaOH takes a few seconds after particle formation.
  - Na$_2$CO$_3$ formation can take several minutes.

- **Aerosols deposit on the floor, walls, and ceiling.**
  - Aerosol particles can cause equipment damage (electrical and instrumentation)
  - Highly toxic

- **Sodium leaks can be detected through gas sampling techniques**
  - Ability for detection means leaks can be readily identified.
Sodium Leaks

- Most sodium leaks have been small and were the result of design and/or fabrication deficiencies.

- E.g. leak from the secondary cold trap at the Fast Breeder Test Reactor:
  - During initial commissioning, preheating of the cold trap led to over-pressurization of the NaK “jacket” due to inadequate expansion space.
  - 2.5 liters of NaK leaked through a spark-plug type high-level probe.
  - A 20 liter expansion “pot” was added to the argon supply line to prevent overpressurization.

- Of the 27 leaks at BN-600, the main causes have been identified as
  - Inadequate valve design.
  - Insufficient thermal compensation and manufacturing faults.
  - Loss of leak-tightness in sodium reception from tanker cars.

- Causes for leaks are known.
Significant Sodium Leaks *

- **BN-600**
  - 1981: Flange joint failure in SG valve seal (300 kg)
  - 1990: Manufacturing defect in SG drain pipeline (600 kg)
  - 1993: Thermal expansion induced failure in primary sodium purification system (1000 kg, ~10 Ci)
  - 1994: Staff error, pipeline cutting before sodium was frozen (650 kg)

- **Monju (8th December, 1995) secondary sodium leak (~640 kg)**
  - 19:47 – Off-scale sodium temperature alarm at IHX outlet, followed by fire alarm, and sodium leak alarm
  - 20:00 – Reactor power-down begins
  - 21:20 – Reactor manually tripped due to observation of increased fumes
  - 22:55 – Began draining secondary sodium circuit
  - 00:15 – Draining operations completed

- **No adverse effects were reported for operating personnel or surrounding environment**

* IAEA-TECDOC-1180: Unusual Occurrences During LMFR Operation, October 2000
Consequences of the Sodium Leak at Monju

Source: Figure 2 from A. Miyakawa, et al., “Sodium Leakage Experience at the Prototype FBR Monju,” Unusual Occurrences During LMFR Operation, IAEA-TECDOC-1180, October 2000. Used with permission.
Sodium Reactivity with Water

- Sodium-water chemical interactions take place in two stages.
- In the first stage, the reaction proceeds at a high rate with release of gaseous hydrogen:
  \[ \text{Na} + \text{H}_2\text{O} = \text{NaOH} + \frac{1}{2} \text{H}_2 + 140 \text{ kJ/mole} \]

- In the second stage, chemical interaction takes place between the products of the first stage and excess sodium:
  \[ 2 \text{Na} + \text{NaOH} = \text{Na}_2\text{O} + \text{NaH} \]
  \[ \text{Na} + \frac{1}{2} \text{H}_2 = \text{NaH} \]

- Hydrogen detection plays a key role in leak detection
  - Diffusion method uses a metal membrane permeable to hydrogen.
  - E.g. diffusion of hydrogen through a nickel membrane into a vacuum cavity coupled to a magnetic discharge pump.
  - Capable of detecting 10 – 30 gram water leak into 100 tons of secondary sodium.

- Acoustic detection techniques are also being developed.
**Sodium Reactivity with Water**

- Potential for sodium/water interactions requires separation of steam cycle from primary system: an intermediate sodium loop is used.
- Lack of fabrication experience in early sodium/water steam generators resulted in a number of large and small water leaks into the sodium.
  - 33 gas-space leaks in PFR SGs were all associated with cracking of the tube-to-tubeplate welds due to lack of post-weld heat treatment.
  - Large ingress of water does not lead to catastrophic consequences. (One incident at BN-350 leaked 800 kg of water into secondary sodium.)
  - Components remained repairable.
- Steam generators can be designed to minimize the impact of leaks on the operation of the plant as a whole.
  - Minimize weld joints and prevent flow-induced vibrations
  - Proper selection of tube materials, thickness, spacing
  - Use of expansion volumes, rupture disks
  - Accommodate leak detection

E BR-II Steam Generator Experience

- No tube leaks occurred during the 30 years of operation of EBR-II.
- No sodium leaks occurred at the tube-to-sodium tube sheet welds.
- One water/steam leak occurred at the tube-to-steam tube sheet.
  - Steam leaked into the space between the two tube sheets.
  - This weld was repaired (only one tube-to-tube sheet weld was involved) and the unit was returned to service.
  - Repaired unit operated satisfactorily for the life of the plant.
- The EBR-II objective was achieved: sodium and water never came in contact during the operating lifetime of the plant.

Source: Figure C-30 from L. J. Koch, Experimental Breeder Reactor-II (EBR-II) An Integrated Experimental Fast Reactor Nuclear Power Station, © 2008 by the American Nuclear Society, La Grange Park, IL. Used with permission.
Impact of Coolant on SFR and LWR Differences
Coolant Choice Affects Design Differences

- The choice of sodium has broad consequences in reactor design
  - Absence of high pressure in the primary system
  - Higher operating temperatures with significant thermal margins
  - More compact fuel assembly designs
  - Incorporation of an intermediate loop
  - Different arrangements for fuel handling
## Selected Properties of Sodium and Water

<table>
<thead>
<tr>
<th>Property</th>
<th>Sodium</th>
<th>Water</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atomic Weight</td>
<td>22.997</td>
<td>18</td>
</tr>
<tr>
<td>Optical Properties</td>
<td>Opaque</td>
<td>Transparent</td>
</tr>
<tr>
<td>Melting Point (°C)</td>
<td>97.8</td>
<td>0</td>
</tr>
<tr>
<td>Boiling Point (°C)</td>
<td>892</td>
<td>100</td>
</tr>
<tr>
<td>Density (kg/m³)</td>
<td>880</td>
<td>713</td>
</tr>
<tr>
<td>Specific Heat (J/kg-K)</td>
<td>1300</td>
<td>5600</td>
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<tr>
<td>Heat Capacity (MJ/m³-K)</td>
<td>1.14</td>
<td>4.00</td>
</tr>
<tr>
<td>Thermal Conductivity (W/m-K)</td>
<td>76</td>
<td>0.54</td>
</tr>
<tr>
<td>Viscosity (cP)</td>
<td>0.34</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Values at STP. *Italic = Evaluated at ~300°C (and 2000 psi for water)*
Sodium Allows Operation at Low System Pressure

- While PWRs operate at system pressures in excess of 2000 psi, the inert cover gas in a sodium-cooled fast reactor is near atmospheric pressure.
- This difference impacts several design features:
  - Vessel Thickness: PWR ~10 inches, SFR ~ 1 inch
  - No need for pressurization of SFR fuel pins.
- Low system pressure offers advantages in terms of safety:
  - Minimal pressure loading on the coolant boundary.
  - In a high-pressure system, coolant pipe breaks are a concern.
  - In a low-pressure system, coolant leaks are unlikely to propagate to a large-scale failure.
  - No need for high-pressure injection cooling.
Some nucleate boiling in a PWR is desirable, and the margin to boiling does not represent a real limit. Instead, the limit is defined by the departure from nucleate boiling, which can result in cladding burnout.

Boiling in an SFR significantly impairs heat transfer and must be avoided.
**Sodium Possesses High Thermal Conductivity**

- In sodium, the thickness of the thermal boundary layer is significantly larger than the thickness of the hydrodynamic boundary layer.
  - Heat transfer coefficient for sodium is independent of viscosity (which is temperature dependent).

- In turbulent flow, there is a high resistance to heat transfer in the laminar and buffer sublayers for ordinary fluids (Pr ~ 1).
  - Critical heat flux determines minimum pin diameter.
  - Moderator-to-fuel ratio impacts core size.

- For sodium (Pr < 0.01) resistance to heat transfer is distributed throughout the flow channel.
  - No thermal limit on pin diameter.

---

**FIGURE 9-15.** Comparison of thermal ($\delta_t$) and hydrodynamic ($\delta_h$) boundary layers for ordinary fluids vs liquid metals.

**Source:** Figure 9-15 (upper right) and Figure 9-16 (above) from A. E. Waltar and A. B. Reynolds, *Fast Breeder Reactors*, Pergamon Press, New York, 1981. Used with permission.
Thermal Hydraulics and Neutronics Affect Assembly Design

Typical PWR Assembly (289 pin locations)
Pin Diameter = 9.4 mm
Pin Pitch = 12.5 mm

“Typical” SFR Assembly (271 pins)
Pin Diameter = 7.4 mm
Pin Pitch = 8.9 mm
Sodium Coolant Needs an Intermediate Coolant Loop

- Because of coolant activation, the potential for sodium/water interactions between high-pressure steam and a low-pressure sodium loop, an intermediate coolant loop is used.
- This leads to two fundamental design choices: Pool vs. Loop

**Opaqueness of Sodium**

- The opaqueness of sodium has never been an issue for the safe operation of sodium fast reactors.
- Fuel handling requires careful tracking and positioning since visual inspections cannot be made under sodium.
- Dimensional gauging: A probe (or the fuel handling mechanism) can be used to identify and measure predefined index points to check the integrity of reactor internal structures.
- Under-sodium viewing (USV):
  - Ultrasonic imaging technique
  - Developed as early as the late 1960s
  - Can suffer from specular reflection effects
  - Presently limited to a research area. Not essential for operation.

Source: Figure 6.20 from Status of Liquid Metal Cooled Fast Reactor Technology, IAEA-TECDOC-1083, April 1999. Used with permission.
Summary

- Extensive testing of a wide variety of coolants in the 1950s and 1960s.
- Nearly all fast reactors constructed have used sodium coolant.
  - 50+ years of sodium component development, testing, and operation.
  - Current fast reactor construction projects also use sodium.
  - Very limited experience with LBE coolant, and no experience with lead or helium-cooled fast reactors.

- Thermophysical and thermal-hydraulic properties of sodium are superior to lead or helium.
  - Smaller core with higher power density, lower enrichment, and lower heavy metal inventory.
  - Use of sodium codified in ASTM standards.

- Issues of sodium reactivity must be addressed through proper component design, fabrication, and testing.

- There are important differences in reactor design introduced by the use of sodium.
  - Low system pressure, high thermal conductivity, large safety margins.
  - Demonstrated capability for passive shutdown and decay heat removal.
Fast Reactor Physics and Core Design

NRC Topical Seminar on Sodium Fast Reactors
Two White Flint, Rockville, MD
May 3, 2007

Robert N. Hill
Department Head – Nuclear Systems Analysis
Nuclear Engineering Division
Argonne National Laboratory
Outline

■ Fast Reactor Physics
  - Contrast LWR physics and different fast reactor types
  - Important phenomena and modeling/data challenges
  - Impact on fuel cycle performance
  - Brief overview of existing methods and codes

■ Sodium-cooled Fast Reactor (SFR) Core Design
  - Contrast to LWR design parameters
  - Typical reactor configuration
  - Typical reactor performance

■ SFR Reactivity Coefficients
  - Identification of physics for each feedback
  - Brief discussion of safety implications
Comparison of LWR and SFR Spectra

- In LWR, most fissions occur in the 0.1 eV thermal “peak”
- In SFR, moderation is avoided – no thermal neutrons
Neutron Moderation Comparison

- Significant elastic scattering of the neutrons in both spectra

- In FRs, neutron moderation is avoided by using high A materials
  - Sodium is most moderating

- In LWRs, neutrons are moderated primarily by hydrogen
- Oxygen in water and fuel also slows down the neutrons

- Slowing-down power in FR is ~1% that observed for typical LWR
- Thus, neutrons are either absorbed or leak from the reactor before they can reach thermal energies

<table>
<thead>
<tr>
<th></th>
<th>$\sigma_s$ (barn)</th>
<th>$N$ (#/barn-cm)</th>
<th>$\xi \Sigma_s$ (cm$^{-1}$)</th>
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<tbody>
<tr>
<td>TRU</td>
<td>4.0</td>
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<td>1.1E-04</td>
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<td>U</td>
<td>5.6</td>
<td>5.6E-03</td>
<td>2.7E-04</td>
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<td>Zr</td>
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<td>2.6E-03</td>
<td>4.6E-04</td>
</tr>
<tr>
<td>Fe</td>
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<td>2.3E-03</td>
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<tr>
<td>Na</td>
<td>3.8</td>
<td>8.2E-03</td>
<td>2.7E-03</td>
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<tr>
<td>H</td>
<td>11.9</td>
<td>2.9E-02</td>
<td>3.5E-01</td>
</tr>
</tbody>
</table>
Also, spectral differences between fast reactor concepts
- At high energy (>1 MeV) lead is effective inelastic scattering material
- Low energy tail caused by moderating materials (next viewgraph)
Fast Reactor Moderating Materials

- Lead has highest scattering of the neutrons, but little moderation
- Oxide fuel in SFR leads to a significant moderation effect – resonances also observed
- LFR designs also utilize nitride or oxide fuel forms
- In modern GFR designs, SiC matrix for fuel is utilized, with most moderation of the FR cases
- Net result is that the SFR-metal has the hardest neutron spectrum
  - SFR-oxide and LFR similar with slightly more moderation
- GFR (with silicon-carbide matrix) has significant low energy tail

<table>
<thead>
<tr>
<th></th>
<th>$\sigma_s$ (barn)</th>
<th>N (#/barn⋅cm)</th>
<th>$\xi \Sigma_s$ (cm$^{-1}$)</th>
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<tr>
<td>TRU</td>
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<td>Pb</td>
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<td>C</td>
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<tr>
<td>He</td>
<td>1.7</td>
<td>3.1E-04</td>
<td>2.3E-04</td>
</tr>
</tbody>
</table>
Spectral Variation of Neutron Cross Sections Pu-239

- Fission and capture cross section >100X higher in thermal range
- Sharp decrease in capture cross section at high energy
Spectral Variation of Neutron Cross Sections U-238

- Much smaller thermal increase in capture (~10X)
- Unresolved resonance range begins at ~10 keV
- Threshold fission at ~1 MeV
Spectral Variation of Neutron Cross Sections Fe and Na

- Capture cross sections much higher in thermal range
- Significant scattering resonance structure throughout fast range
**Implications of Fast Spectrum Physics**

- Combination of increased fission/absorption and increased number of neutrons/fission yields **more excess neutrons** from Pu-239
  - Enables “breeding” of fissile material
- In a fast spectrum, U-238 capture is more prominent
  - **Higher enrichment** (TRU/HM) is required (next viewgraph)
  - Enhances internal conversion
- Reduced parasitic capture and improved neutron balance
  - Allows the use of conventional stainless steel structures
  - Slow loss of reactivity with burnup
    - **Less fission product capture and more internal conversion**
- The lower absorption cross section of all materials leads to a much longer neutron diffusion length (10-20 cm, as compared to 2 cm in LWR)
  - Neutron leakage is increased (>20% in typical designs)
  - Reflector effects are more important
  - Heterogeneity effects are relatively unimportant
**Impact of Energy Spectrum on Enrichment and Depletion Behavior**

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Thermal Concepts</th>
<th>Fast Concepts</th>
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<tbody>
<tr>
<td></td>
<td>PWR</td>
<td>VHTR</td>
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<tr>
<td>U238c</td>
<td>0.91</td>
<td>4.80</td>
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<tr>
<td>Pu239f</td>
<td>89.2</td>
<td>164.5</td>
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<tr>
<td>P239f/U238c</td>
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<tr>
<td>Fe</td>
<td>0.4</td>
<td></td>
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<tr>
<td>Fission Prod.</td>
<td>90</td>
<td></td>
</tr>
</tbody>
</table>

- Generation-IV fast systems have similar characteristics
- One-group XS are significantly reduced in fast system
- However, U-238 capture is much more prominent (low P239f/U238c)
  - *A much higher enrichment is required to achieve criticality*
- The parasitic capture cross section of fission products and conventional structures is much higher in a thermal spectrum (next viewgraph)
# Neutron Balance

<table>
<thead>
<tr>
<th>Source</th>
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<tbody>
<tr>
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<td>33.3</td>
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<tr>
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<td></td>
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<tr>
<td>fission</td>
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<td>99.8%</td>
<td>99.9%</td>
</tr>
<tr>
<td>(n,2n)</td>
<td>0.2%</td>
<td>0.1%</td>
<td></td>
</tr>
<tr>
<td>Leakage</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>radial</td>
<td>3.0%</td>
<td>12.3%</td>
<td>16.6%</td>
</tr>
<tr>
<td>axial</td>
<td>0.4%</td>
<td>10.6%</td>
<td>12.1%</td>
</tr>
<tr>
<td>Absorption</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>fuel</td>
<td>96.5%</td>
<td>77.1%</td>
<td>71.3%</td>
</tr>
<tr>
<td>(U-238 capture)</td>
<td>(27.2%)</td>
<td>(31.6%)</td>
<td>(17.1%)</td>
</tr>
<tr>
<td>coolant</td>
<td>3.4%</td>
<td>0.1%</td>
<td>0.1%</td>
</tr>
<tr>
<td>structure</td>
<td>0.6%</td>
<td>3.7%</td>
<td>3.7%</td>
</tr>
<tr>
<td>fission product</td>
<td>6.8%</td>
<td>1.5%</td>
<td>2.4%</td>
</tr>
<tr>
<td>control</td>
<td>9.0%</td>
<td>0.0%</td>
<td>2.9%</td>
</tr>
</tbody>
</table>

- Conversion ratio defined as ratio of TRU production/TRU destruction
  - Slightly different than traditional breeding ration with fissile focus
Impact of Energy Spectrum on Fuel Cycle (Transmutation) Performance

- Fissile isotopes are likely to fission in both thermal/fast spectrum
  - Fission fraction is higher in fast spectrum
- Significant (up to 50%) fission of fertile isotopes in fast spectrum

Net result is more excess neutrons and less higher actinide generation in FR
Spectral Comparison of Isotopic D-factors

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Thermal Concepts</th>
<th>Fast Concepts</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PWR</td>
<td>VHTR</td>
</tr>
<tr>
<td>U-235</td>
<td>0.65</td>
<td>0.53</td>
</tr>
<tr>
<td>U-238</td>
<td>0.02</td>
<td>-0.26</td>
</tr>
<tr>
<td>Np-237</td>
<td>-0.96</td>
<td>-1.11</td>
</tr>
<tr>
<td>Pu-239</td>
<td>0.83</td>
<td>0.72</td>
</tr>
<tr>
<td>Pu-240</td>
<td>-0.04</td>
<td>-0.12</td>
</tr>
<tr>
<td>Pu-241</td>
<td>0.95</td>
<td>0.88</td>
</tr>
</tbody>
</table>

- D-factor measures the neutron balance to completely fission a given isotope
  - Positive value indicates excess neutrons are generated
- Fast systems have favorable neutron balance for all TRU isotopes
  - Thermal reactor only for the fissile isotopes
- Thus, fast systems can efficiently convert U-238 and consume the actinides, while a fissile source is required to sustain the thermal conversion
Equilibrium Composition in Fast and Thermal Spectra

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Once-Through</th>
<th>Fast U-238</th>
<th>Thermal U-238</th>
</tr>
</thead>
<tbody>
<tr>
<td>Np237</td>
<td>0.048</td>
<td>0.008</td>
<td>0.002</td>
</tr>
<tr>
<td>Pu238</td>
<td>0.024</td>
<td>0.014</td>
<td>0.046</td>
</tr>
<tr>
<td>Pu239</td>
<td>0.476</td>
<td>0.666</td>
<td>0.388</td>
</tr>
<tr>
<td>Pu240</td>
<td>0.225</td>
<td>0.243</td>
<td>0.197</td>
</tr>
<tr>
<td>Pu241</td>
<td>0.106</td>
<td>0.021</td>
<td>0.111</td>
</tr>
<tr>
<td>Pu242</td>
<td>0.066</td>
<td>0.018</td>
<td>0.085</td>
</tr>
<tr>
<td>Am241</td>
<td>0.034</td>
<td>0.021</td>
<td>0.019</td>
</tr>
<tr>
<td>Am242m</td>
<td>0.000</td>
<td>0.001</td>
<td>0.001</td>
</tr>
<tr>
<td>Am243</td>
<td>0.015</td>
<td>0.005</td>
<td>0.033</td>
</tr>
<tr>
<td>Cm242</td>
<td>0.000</td>
<td>0.000</td>
<td>0.002</td>
</tr>
<tr>
<td>Cm244</td>
<td>0.005</td>
<td>0.002</td>
<td>0.055</td>
</tr>
<tr>
<td>Cm245</td>
<td>0.000</td>
<td>0.000</td>
<td>0.018</td>
</tr>
<tr>
<td>Cm246</td>
<td>0</td>
<td>0.000</td>
<td>0.031</td>
</tr>
<tr>
<td>Cm247</td>
<td>0</td>
<td>0.000</td>
<td>0.004</td>
</tr>
<tr>
<td>Cm248</td>
<td>0</td>
<td>0.000</td>
<td>0.006</td>
</tr>
</tbody>
</table>

- Equilibrium higher actinide content much lower in fast spectrum system
- Generation of Pu-241 (key waste decay chain) is suppressed
- However, if starting from once-through LWR composition (e.g., burner reactor) the higher actinide content will be higher than the U-238 equilibrium
Fuel Cycle Implications

The physics distinctions facilitate different fuel cycle strategies

- **Thermal reactors** are typically configured for once-through (open) fuel cycle
  - They can operate on low enriched uranium (LEU)
  - They require an external fissile feed (neutron balance)
  - Higher actinides must be managed to allow recycle
    - Separation of higher elements – still a disposal issue
    - Extended cooling time for curium decay

- **Fast reactors** are typically intended for closed fuel cycle with uranium conversion and resource extension
  - Higher actinide generation is suppressed
  - Neutron balance is favorable for recycled TRU
    - No external fissile material is required
    - Can enhance U-238 conversion for traditional breeding
    - Can limit U-238 conversion for burning
Computational Methods

- Many of the assumptions employed in traditional LWR methods do not apply
  - Lack of a 1/E type spectrum as a basis for the calculation of resonance absorption
    - $E \varphi(E)$ strongly decreases with decreasing energy in FR
  - Up-scattering resulting from the thermal motion of the scattering nuclei may be neglected
  - Inelastic, (n,2n), anisotropic scatterings are of great importance
  - Long mean free path implies global coupling
    - Local reactivity effects impact entire core
  - The energy range where neutrons induce fission and the energy range where the fission neutrons appear strongly overlap

- Other physics consideration have high priority in FR methods
  - Detailed energy modeling for resonance structure (core/reflectors)
  - Transport and anisotropy effects more important at high energy

- In general, a distinct set of physics analysis and core design tools with tailored assumptions was developed for fast reactor analysis
Comparison of LWR and SFR Spectra

- In LWR, most fissions occur in the 0.1 eV thermal “peak”
- In SFR, moderation is avoided – no thermal neutrons
Existing Sodium-Cooled Fast Reactor Code Suite

- ENDF/B Cross Section Data
- System Configuration, Operating Conditions
- Assembly Design Parameters
- Fuel Management Scheme
- Transient Scenario
- Plant Layout
- T-H and Structural Data, Eqs. of State
- ETOE2/MC^2-2/SDX (Cell Homogenization and Energy Condensation)
- Multigroup XS vs. Burnup and Temperature
- RCT Intra-Assembly Reconstruction
- Pin-wise Flux, Power, and Nuclide Densities
- DIF3D/VARIANT/TWODANT REBUS-3 (Whole Core Neutronics & Depletion)
- Assembly Power, Burnup, and Nuclide Densities
- VARI3D Perturbation Theory
- Reactivity Coeff. and Kinetics Parameters
- SE2-ANL (Steady State Thermal Hydraulics)
- Flow and Temperature Distributions
- System Dynamic Behavior
- SASSYS/SAS4A (Coupled Neutron Kinetics, Thermal Hydraulics, and Structural Mechanics)
Typical Neutronics Analysis Techniques

- Design calculations typically performed using ~33 energy groups
  - Cross sections self-shielded at ultrafine group (~2000+) level
  - Spatial collapse to form regional broad group cross sections

- Design calculations utilize nodal diffusion method
  - Nodal transport techniques also available
  - Continuous energy Monte Carlo for benchmarking
  - Advanced transport methods (generalized geometry and many energy groups) being developed in GNEP advanced simulation campaign

- Depletion calculations include extensive external cycle modeling
  - Several axial depletion regions within assembly (pin-wise for processing)
  - Conventional FR fuel management does not shuffle fuel

- Current tools are adequate to begin the design process
  - Extensive critical experiment and reactor operation database exists
  - Validation and capabilities have evolved in parallel
  - Formal and comprehensive documentation needs to be done
  - This activity should support evaluation of advanced methods
## Typical Design Specifications of LWR and SFR

<table>
<thead>
<tr>
<th></th>
<th>PWR</th>
<th>SFR</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>General</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Specific power (kWt/kgHM)</td>
<td>786 (U-235)</td>
<td>556 (Pu fissile)</td>
</tr>
<tr>
<td>Power density (MWt/m³)</td>
<td>102</td>
<td>300</td>
</tr>
<tr>
<td><strong>Fuel</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rod outer diameter (mm)</td>
<td>9.5</td>
<td>7.9</td>
</tr>
<tr>
<td>Clad thickness (mm)</td>
<td>0.57</td>
<td>0.36</td>
</tr>
<tr>
<td>Rod pitch-to-diameter ratio</td>
<td>1.33</td>
<td>1.15</td>
</tr>
<tr>
<td>Enrichment (%)</td>
<td>~4.0</td>
<td>~20 Pu/(Pu+U)</td>
</tr>
<tr>
<td>Average burnup (MWd/kg)</td>
<td>40</td>
<td>100</td>
</tr>
<tr>
<td><strong>Thermal Hydraulic</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Coolant</td>
<td></td>
<td></td>
</tr>
<tr>
<td>pressure (MPa)</td>
<td>15.5</td>
<td>0.1</td>
</tr>
<tr>
<td>inlet temp. (°C)</td>
<td>293</td>
<td>332</td>
</tr>
<tr>
<td>outlet temp. (°C)</td>
<td>329</td>
<td>499</td>
</tr>
<tr>
<td>reactor Δp (MPa)</td>
<td>0.345</td>
<td>0.827</td>
</tr>
<tr>
<td>Rod surface heat flux</td>
<td>average (MW/m²)</td>
<td>0.584</td>
</tr>
<tr>
<td></td>
<td>maximum (MW/m²)</td>
<td>1.46</td>
</tr>
<tr>
<td>Average linear heat rate (kW/m)</td>
<td>17.5</td>
<td>27.1</td>
</tr>
<tr>
<td>Steam</td>
<td></td>
<td></td>
</tr>
<tr>
<td>pressure (MPa)</td>
<td>7.58</td>
<td>15.2</td>
</tr>
<tr>
<td>temperature (°C)</td>
<td>296</td>
<td>455</td>
</tr>
<tr>
<td><strong>Control</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Control Rods</td>
<td>0.07 (10.8 $)</td>
<td>Primary/Secondary (10$/3$)</td>
</tr>
<tr>
<td>Chemical Shim</td>
<td>0.25 (38.5 $)</td>
<td></td>
</tr>
</tbody>
</table>
Sodium-Cooled Fast Reactor Primary Vessel and Fuel Assembly

Fuel Assembly (FFTF)
## Typical Core Design Volume Fractions

<table>
<thead>
<tr>
<th>Material</th>
<th>PWR</th>
<th>SFR</th>
<th>LFR</th>
<th>GFR</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>CR=1.0</td>
<td>CR=0.5</td>
<td></td>
</tr>
<tr>
<td>Fuel</td>
<td>30</td>
<td>40</td>
<td>30</td>
<td>34</td>
</tr>
<tr>
<td>Coolant</td>
<td>59</td>
<td>35</td>
<td>44</td>
<td>55</td>
</tr>
<tr>
<td>Structure</td>
<td>11</td>
<td>25</td>
<td>26</td>
<td>11</td>
</tr>
</tbody>
</table>

- For conventional fast reactor, fuel volume fraction (VF) maximized
  - Tightly packed pin lattice
  - High volume fraction blankets to introduce additional U-238
- For burner design, TRU production reduced by lower fuel VF
  - Smaller pins to yield increased coolant VF
- For LFR, higher coolant VF is required to reduce coolant velocity
  - Needed for oxygen control to prevent cladding erosion
- For GFR, higher coolant VF is required because of inferior heat transfer
  - Trade-off between pumping power and neutronic performance
Early Fast Reactors and Fuel Forms

Original choice was high density metal fuel (for breeding)
- First usable nuclear electricity—EBR-I in 1951
- EBR-II (1963), Fermi (1963), DFR (UK, 1959) all used metal fuel
- Early designs experienced severely limited fuel burnup because of fuel swelling (U-10Mo burnup of 3 GWd/MT for Fermi)

U.S. and international programs switched to oxide fuel in the late 1960s
- Low swelling and successful Navy oxide fuel experience → high burnup
- Fast Flux Test Facility (400 MWt) operated with oxide from 1980 to 1992
- EBR-II (20 MWe) continued metal fuel development from 1963 to 1994
  - Solved burnup limitation by allowing adequate space for fuel swelling
  - Demonstrated peak burnup comparable to oxide fuel (200 GWd/MT)
# Fast Reactor Fuel Options

<table>
<thead>
<tr>
<th>Fast Reactor Fuel Type Fresh Fuel Properties</th>
<th>Metal U-20Pu-10Zr</th>
<th>Oxide UO₂-20PuO₂</th>
<th>Nitride UN-20PuN</th>
<th>Carbide UC-20PuC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heavy Metal Density, g/cm³</td>
<td>14.1</td>
<td><strong>9.3</strong></td>
<td>13.1</td>
<td>12.4</td>
</tr>
<tr>
<td>Melting Temperature, °K</td>
<td><strong>1350</strong></td>
<td>3000</td>
<td>3035*</td>
<td>2575</td>
</tr>
<tr>
<td>Thermal Conductivity, W/cm-°K</td>
<td>0.16</td>
<td><strong>0.023</strong></td>
<td>0.26</td>
<td>0.20</td>
</tr>
<tr>
<td>Operating Centerline Temperature at 40 kW/m, °K, and (T/Tₘₐₑₙ)</td>
<td>1060 (0.8)</td>
<td>2360 (0.8)</td>
<td>1000 (0.3)</td>
<td>1030 (0.4)</td>
</tr>
<tr>
<td>Fuel-Cladding Solidus, °K</td>
<td><strong>1000</strong></td>
<td>1675</td>
<td>1400</td>
<td>1390</td>
</tr>
<tr>
<td>Thermal Expansion, 1/°K</td>
<td>17E-6</td>
<td>12E-6</td>
<td>10E-6</td>
<td>12E-6</td>
</tr>
<tr>
<td>Heat Capacity, J/g°K</td>
<td>0.17</td>
<td>0.34</td>
<td>0.26</td>
<td>0.26</td>
</tr>
<tr>
<td>Reactor Experience, Country</td>
<td>US, UK</td>
<td>RUS, FR, JAP US, UK</td>
<td>IND</td>
<td></td>
</tr>
<tr>
<td>Research &amp; Testing, Country</td>
<td>US, JAP, ROK, CHI</td>
<td>RUS, FR, JAP US, CHI</td>
<td>US, RUS, JAP</td>
<td>IND</td>
</tr>
</tbody>
</table>
Conventional 1000 MWt SuperPRISM (Metal Core)

- Internal and external blankets allocated
  - Result in conversion ratio of ~1
- Only 12 control rod locations with very low burnup reactivity losses
- Blanket, two row reflector, and boron carbide for radial shielding
Two enrichment zones to reduce radial power peaking
No blankets allocated for conversion ratio < 1
Additional (20) control rod locations for burnup reactivity losses
Similar radial shield configuration
### Core Performance and Mass Flow

<table>
<thead>
<tr>
<th></th>
<th>SPRISM – Metal</th>
<th>1000 MWt ABR</th>
<th>840 MWt LCFR</th>
<th>3500 MWt US-Eur</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Breakeven</td>
<td>Breeder</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cycle length, month</td>
<td>23</td>
<td>23</td>
<td>12</td>
<td>6</td>
</tr>
<tr>
<td>Number of batches</td>
<td>4</td>
<td>4</td>
<td>4</td>
<td>6</td>
</tr>
<tr>
<td>Average TRU enrichment, %</td>
<td>~11</td>
<td>~9</td>
<td>21.9</td>
<td>~50</td>
</tr>
<tr>
<td>Fissile/TRU conversion ratio</td>
<td>1.05 / -</td>
<td>1.22 / -</td>
<td>0.84 / 0.73</td>
<td>- / 0.25</td>
</tr>
<tr>
<td>HM/TRU inventory at BOC, MT</td>
<td>26.1 / 3.1</td>
<td>36.3 / 3.2</td>
<td>13.2 / 2.9</td>
<td>4.6 / 2.25</td>
</tr>
<tr>
<td>Average/peak burnup, MWd/kg</td>
<td>106 / 149</td>
<td>103 / 145</td>
<td>93 / 138</td>
<td>177 / 321</td>
</tr>
<tr>
<td>Peak Fast Fluence, 10^{23} n/cm^2</td>
<td>3.9</td>
<td>3.9</td>
<td>4.0</td>
<td>4.0</td>
</tr>
<tr>
<td>Burnup reactivity swing (%Δk)</td>
<td>0.12</td>
<td>-0.31</td>
<td>3.5</td>
<td>4.3</td>
</tr>
<tr>
<td>Average linear power, kW/m</td>
<td>18.9</td>
<td>18.3</td>
<td>23.3</td>
<td></td>
</tr>
<tr>
<td>Peak linear power, kW/m</td>
<td>30.4</td>
<td>29.8</td>
<td>37.2</td>
<td>45.4</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Mass flow (kg/year)</th>
<th>Uranium</th>
<th>Breakeven</th>
<th>Breeder</th>
<th>1000 MWt ABR</th>
<th>840 MWt LCFR</th>
<th>3500 MWt US-Eur</th>
</tr>
</thead>
<tbody>
<tr>
<td>TRU</td>
<td>33.6</td>
<td>-255.8</td>
<td>-75</td>
<td>-1216</td>
<td></td>
<td></td>
</tr>
<tr>
<td>MA</td>
<td></td>
<td>-6.5</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fissile Pu</td>
<td>19.3</td>
<td>69.9</td>
<td>-44.0</td>
<td>124</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

NRC Topical Seminar on SFR, May 3, 2007
Burner Design Challenges

- Radial blanket typically replaced by reflector
  - Many criticals (BFS-62, MUSE-4) exhibit problems in accurate prediction of reaction rates in the immediate reflector region
  - Spectral and directional transitions are hard to model
  - Important for shielding and bowing (safety) considerations
- High leakage configurations also challenge design methods
  - Transport effects are magnified
  - Key reactivity coefficients (e.g., void worth) impacted
- Current GNEP strategy keeps the grouped TRUs together
  - Minor actinides present in fresh fuel
  - If low conversion ratio, the TRU enrichment could be high
  - However, plutonium remains the dominant fission source (fission contributions on next viewgraph)
  - For dedicated minor actinide burners (or targets), uncertainty of the basic MA cross section data becomes important
## Isotopic Fission Fractions

<table>
<thead>
<tr>
<th>U-235 or TRU enrichment, %</th>
<th>PWR</th>
<th>SFR CR=1.0</th>
<th>SFR CR=0.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-235</td>
<td>4.2</td>
<td>13.9</td>
<td>33.3</td>
</tr>
<tr>
<td>U-238</td>
<td>59.8%</td>
<td>0.2%</td>
<td>0.2%</td>
</tr>
<tr>
<td>Np-237</td>
<td>6.2%</td>
<td>15.7%</td>
<td>8.3%</td>
</tr>
<tr>
<td>Pu-238</td>
<td>0.2%</td>
<td>0.8%</td>
<td>3.4%</td>
</tr>
<tr>
<td>Pu-239</td>
<td>29.8%</td>
<td>70.7%</td>
<td>57.0%</td>
</tr>
<tr>
<td>Pu-240</td>
<td>6.7%</td>
<td>11.4%</td>
<td></td>
</tr>
<tr>
<td>Pu-241</td>
<td>4.0%</td>
<td>4.5%</td>
<td>11.9%</td>
</tr>
<tr>
<td>Pu-242</td>
<td>0.4%</td>
<td>2.5%</td>
<td></td>
</tr>
<tr>
<td>Am241</td>
<td>0.3%</td>
<td>0.8%</td>
<td></td>
</tr>
<tr>
<td>Am-242m</td>
<td>0.2%</td>
<td>0.6%</td>
<td></td>
</tr>
<tr>
<td>Am-243</td>
<td>0.1%</td>
<td>0.7%</td>
<td></td>
</tr>
<tr>
<td>Cm-244</td>
<td>0.1%</td>
<td>0.9%</td>
<td></td>
</tr>
<tr>
<td>Cm-245</td>
<td>0.1%</td>
<td>1.2%</td>
<td></td>
</tr>
</tbody>
</table>
Batch-averaged Assembly Power

Fresh Assembly Power

markdown

Power peaking factor (BOC/EOC) = 1.43 / 1.39
Power Profiles of 1000 MWt ABR – Startup Core

- Bottom skewed axial distribution (control assembly tip position at BOC = 57 cm)
- Axial power peaking factor at BOC/EOC = 1.20/1.16
Control Assembly
**Control System Requirements**

- Typical design utilizes ducted bundle of absorber pins within the coolant duct
- Sufficient reactivity worth to bring reactor
  - From any operating condition
    - *Overpower condition*
    - *Reactivity fault*
  - To cold sub-critical at refueling temperature
  - With most reactive control assembly stuck at full power operating condition
- Hold down excess reactivity for fuel cycle
  - Fuel burnup
  - Axial growth of metal fuel
- Accommodate reactivity uncertainties
  - Criticality and fissile loading
  - Temperature defect, burnup reactivity, and fuel growth
## Control Requirement and Shutdown Margin ($) of Primary System

<table>
<thead>
<tr>
<th></th>
<th>1000 MWe LMFBR a)</th>
<th>975 MWt CRBRP b)</th>
<th>840 MWt ALMR/95</th>
<th>1000 MWt ABR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of control assemblies</td>
<td>13</td>
<td>15</td>
<td>10</td>
<td>15</td>
</tr>
<tr>
<td>Total worth</td>
<td>20.0</td>
<td>31.8</td>
<td>20.0</td>
<td>39.5</td>
</tr>
<tr>
<td>Worth of 1 stuck rod</td>
<td>3.0</td>
<td>2.8</td>
<td>2.3</td>
<td>4.3</td>
</tr>
<tr>
<td>Reactivity worth available</td>
<td>17.0</td>
<td>29.0</td>
<td>17.7</td>
<td>35.2</td>
</tr>
<tr>
<td>Control requirement</td>
<td>12.0</td>
<td>26.5</td>
<td>12.7</td>
<td>17.9</td>
</tr>
<tr>
<td>- Temperature defect</td>
<td>2.4</td>
<td>3.2</td>
<td>1.1</td>
<td>1.4</td>
</tr>
<tr>
<td>- Burnup reactivity loss</td>
<td>5.0</td>
<td>18.2</td>
<td>7.2</td>
<td>10.4</td>
</tr>
<tr>
<td>- Fuel axial growth</td>
<td></td>
<td></td>
<td>0.5</td>
<td>1.2</td>
</tr>
<tr>
<td>- Overpower margin</td>
<td>0.3</td>
<td>0.2</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>- Reactivity fault</td>
<td>3.0</td>
<td>2.8</td>
<td></td>
<td>0.7</td>
</tr>
<tr>
<td>- Uncertainties</td>
<td>1.3</td>
<td>2.1</td>
<td>2.0</td>
<td>2.3</td>
</tr>
<tr>
<td>- Other margin c)</td>
<td></td>
<td></td>
<td>1.8</td>
<td>1.8</td>
</tr>
<tr>
<td>Shutdown margin</td>
<td>5.0</td>
<td>2.5</td>
<td>5.0</td>
<td>17.3</td>
</tr>
</tbody>
</table>

a) GEFR-00392 (1978)  
b) Early homogeneous design (1974)  
c) ALMR/95 requires additional margins such as ATWS reactivity and fast runback margin
Requirements of Secondary System

- Sufficient reactivity worth to shutdown reactor
  - From any operating condition
    - Including reactivity fault of one primary control assembly
  - To hot standby condition
  - With most reactive control assembly stuck at full power operating condition

- Not necessary to duplicate primary system capability
  - Hold down excess reactivity for fuel cycle
  - Reactivity uncertainties
## Control Requirement and Shutdown Margin ($) of Secondary System

<table>
<thead>
<tr>
<th></th>
<th>1000 MWe LMFBR a)</th>
<th>975 MWt CRBRP b)</th>
<th>840 MWt ALMR/95</th>
<th>1000 MWt ABR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of control assemblies</td>
<td>6</td>
<td>4</td>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>Total worth</td>
<td>8.0</td>
<td>8.4</td>
<td>9.4</td>
<td>9.5</td>
</tr>
<tr>
<td>Worth of 1 stuck rod</td>
<td>2.5</td>
<td>2.0</td>
<td>3.7</td>
<td>2.6</td>
</tr>
<tr>
<td>Reactivity worth available</td>
<td>5.5</td>
<td>6.4</td>
<td>5.7</td>
<td>6.9</td>
</tr>
<tr>
<td>Control requirement</td>
<td>5.0</td>
<td>6.3</td>
<td>1.7</td>
<td>1.9</td>
</tr>
<tr>
<td>- Temperature defect</td>
<td>1.6</td>
<td>2.4</td>
<td>1.1</td>
<td>1.4</td>
</tr>
<tr>
<td>- Overpower margin</td>
<td>0.3</td>
<td>0.2</td>
<td>0.3</td>
<td>0.2</td>
</tr>
<tr>
<td>- Temp defect uncertainty</td>
<td>0.3</td>
<td>0.9</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>- Reactivity fault or others</td>
<td>3.1</td>
<td>2.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shutdown margin</td>
<td>0.5</td>
<td>0.1</td>
<td>4.0</td>
<td>5.0</td>
</tr>
</tbody>
</table>

a) GEFR-00392 (1978)
b) Early homogeneous design (1974)
Reactivity Feedback Coefficients

- The reactivity coefficients further define the physics of system
  - Kinetics parameters
  - Response to a variety of perturbations
- Feedback coefficients are computed for a specific design (geometric and material) configuration
  - Typically evaluated for BOEC and EOEC composition
- Typical set of whole-core reactivity coefficients
  - Computed with perturbation theory for spatial distributions
    - Delayed neutron fraction and prompt lifetime
    - Sodium density coefficient and void worth
    - Fuel and structural Doppler coefficient
    - Fuel and structural worth distributions
  - Computed by eigenvalue difference ($\Delta k/k$)
    - Uniform axial expansion
    - Uniform radial expansion
    - Control rod driveline expansion
### Whole-Core Reactivity Coefficients for Different Powers

<table>
<thead>
<tr>
<th></th>
<th>250 MWt ABTR</th>
<th>1000 MWt ABR</th>
<th>3500 MWt US-Europe</th>
</tr>
</thead>
<tbody>
<tr>
<td>Effective delayed neutron fraction</td>
<td>0.0033</td>
<td>0.00334</td>
<td>0.0035</td>
</tr>
<tr>
<td>Prompt neutron lifetime</td>
<td>Ms</td>
<td>0.33</td>
<td>0.38</td>
</tr>
<tr>
<td>Radial expansion coefficient</td>
<td>ºC</td>
<td>-0.43</td>
<td>-0.38</td>
</tr>
<tr>
<td>Axial expansion coefficient</td>
<td>ºC</td>
<td>-0.05</td>
<td>-0.05</td>
</tr>
<tr>
<td>Sodium density coefficient</td>
<td>ºC</td>
<td>0.03</td>
<td>0.13</td>
</tr>
<tr>
<td>Doppler coefficient</td>
<td>ºC</td>
<td>-0.10</td>
<td>-0.13</td>
</tr>
<tr>
<td>Sodium void worth</td>
<td>$</td>
<td>1.10</td>
<td>4.93</td>
</tr>
<tr>
<td>Sodium voided Doppler coefficient</td>
<td>ºC</td>
<td>-0.07</td>
<td>-0.09</td>
</tr>
</tbody>
</table>

- **Power coefficient is quite negative**
  - More negative at smaller size because of radial expansion coefficient
  - Sodium density coefficient also more positive at larger size
- **Physics underlying each component will be explained**
  - Void worth will be addressed separately
**Delayed Neutron Fraction**

- Hummel and Okrent – *Reactivity Coefficients in Large Fast Power Reactors, ANS, 1970* is a good reference for underlying physics
- Delayed neutron fraction dominated by key fission isotopes
  - Low (0.2%) for Pu-239
  - High (1.5%) for U-238
  - Between 0.3-0.5% for higher plutonium isotopes
  - Particularly low (<0.2%) for minor actinides
- Net result is 0.3-0.4% for conventional compositions
  - Slightly lower for burner designs (next viewgraph)
- Much higher for U-235 enriched systems
  - Delayed neutron fraction for U-235 is ~0.67%
- Delayed neutron fraction is an indicator of sensitivity
  - At low values, response to small changes in the reactivity is magnified and power can change more quickly
  - Feedback effects can be favorable or not depending on the transient
Low Conversion Ratio Fast Reactor Analyses

Fast reactors with closed fuel cycle can effectively manage TRU

- Can be configured as modest breeders (CR≥1) to moderate burners (CR≥0.5) with conventional technology
- Low conversion ratio designs (CR<0.5) have been investigated for transmutation applications in AFCI
  - High enrichment fuels are required (~50% TRU/HM for CR=0.25)
  - Non-uranium fuel would be needed to achieve CR=0

<table>
<thead>
<tr>
<th>Conversion Ratio</th>
<th>TRU/HM Enrichment</th>
<th>Equilibrium Fraction of FR</th>
<th>Delayed Neutron Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1</td>
<td>11%</td>
<td>100%</td>
<td>0.0034</td>
</tr>
<tr>
<td>0.75</td>
<td>20%</td>
<td>51%</td>
<td>0.0033</td>
</tr>
<tr>
<td>0.5</td>
<td>30%</td>
<td>34%</td>
<td>0.0031</td>
</tr>
<tr>
<td>0.25</td>
<td>50%</td>
<td>26%</td>
<td>0.0028</td>
</tr>
<tr>
<td>0.0</td>
<td>100%</td>
<td>21%</td>
<td>0.0020</td>
</tr>
</tbody>
</table>
**Geometric Expansion Coefficients**

Whole-core coefficients are computed by eigenvalue difference for a small change in each dimension

- **Radial expansion** – uniform expansion of grid plate by 1%
  - Reduction of fuel/structure densities by 1%
  - This allows more axial leakage in particular

- **Axial expansion** – uniform expansion of fuel by 1%
  - Reduction of fuel density by 1%
  - Allows more radial leakage
  - Also, effectively inserts the control rods which remain stationary
  - In some cases, fuel assumed bound to clad for axial expansion

- These feedbacks are very important for fast reactor transient behavior
  - Tied to different material temperatures (grid plate, fuel)
  - Thus, timing will be different
Neutron Balances of Radial and Axial Expansions

<table>
<thead>
<tr>
<th></th>
<th>Base Case</th>
<th>Radial Expansion</th>
<th>Axial Expansion</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>balance</td>
<td>balance</td>
<td>Δρ (%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>balance</td>
</tr>
<tr>
<td>Fission source</td>
<td>100.00</td>
<td>100.00</td>
<td>100.00</td>
</tr>
<tr>
<td>(n,2n) source</td>
<td>0.18</td>
<td>0.18</td>
<td>0.18</td>
</tr>
<tr>
<td>Absorption</td>
<td>68.89</td>
<td>68.93</td>
<td>-0.04</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>68.93</td>
</tr>
<tr>
<td>Leakage</td>
<td>31.54</td>
<td>32.16</td>
<td>-0.63</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>31.61</td>
</tr>
<tr>
<td>Radial</td>
<td>17.49</td>
<td>17.72</td>
<td>-0.23</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>17.59</td>
</tr>
<tr>
<td>Axial</td>
<td>14.05</td>
<td>14.45</td>
<td><strong>-0.40</strong></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>14.02</td>
</tr>
<tr>
<td>Sum</td>
<td></td>
<td>-0.67</td>
<td>-0.12</td>
</tr>
</tbody>
</table>

- To first order, radial expansion is an axial leakage effect, and
- Axial expansion is a radial leakage effect
- Because the height is the short dimension (more axial than radial leakage), the radial expansion coefficient is more negative
- Absorption effect arises from control rod absorption increases
Coolant Density Coefficient

Coolant density coefficient computed by first-order perturbation theory to evaluate small density (temperature variation) impacts

- **Spectral effect**
  - Reduced moderation as sodium density decreases
  - In fast regime, this is a positive reactivity effect
    - *From Pu-239 excess neutrons and threshold fission effects*

- **Leakage effect**
  - Sodium density decrease allows more neutron leakage
  - This is a negative reactivity effect in the peripheral regions

- **Capture effect**
  - Sodium density decrease results in less sodium capture
  - This is a relatively minor effect

Void worth is evaluated using exact perturbation theory to account for shift in flux distribution and change in cross sections for voided condition

In general, 10% more positive than the first-order density worth
**Sodium Void Worth by Components ($)**

<table>
<thead>
<tr>
<th></th>
<th>Capture</th>
<th>Spectral</th>
<th>Leakage</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>1000 MWt ABR</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(startup metal core)</td>
<td>BOC</td>
<td>0.5</td>
<td>9.1</td>
<td>4.4</td>
</tr>
<tr>
<td></td>
<td>EOC</td>
<td>0.5</td>
<td>9.9</td>
<td>4.9</td>
</tr>
<tr>
<td><strong>250 MWt ABTR</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(startup metal core)</td>
<td>BOC</td>
<td>0.4</td>
<td>6.4</td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>EOC</td>
<td>0.4</td>
<td>6.6</td>
<td>1.1</td>
</tr>
</tbody>
</table>

- Flowing sodium completely voided in ALL active and above-core regions
- Void worth tends to increase with core size
- However, difficult to conceive transient situations that reach boiling
  - Low pressure system
  - >300°C margin to boiling
  - Other feedbacks are negative (see next viewgraphs)
Response to a ULOF in Large 3500 MWt SFR using Metal Fuel
Response to a ULOF in Large 3500 MWt SFR using Metal Fuel
Doppler Coefficient

- Doppler coefficient arises primarily from U-238 resonance broadening
  - Enhanced by high U-238 content
    - Reduced Doppler for high enrichment burner concepts
  - Self-shielding effect more pronounced at low energies (keV range)
    - Doppler enhance by spectral softening
    - Voided Doppler is smaller from spectral shift

- Temperature dependence in fast spectrum is different than LWR
  - Doppler range from $1/T^{1/2}$ for large to $1/T^{3/2}$ for small resonances
  - For typical FR, an approximate $1/T$ dependence observed

- There is also a structural Doppler reactivity effect (~1/3 fuel Doppler)
  - However, tied to temperature of steel, not fuel (different timing)

- Doppler feedback is not helpful in all transients
  - For example, when trying to cool the fuel to shutdown condition (e.g., ULOF), it is a positive feedback
  - Conversely prompt negative feedback in UTOP transient
Passive Safety Behavior


Brief synopsis by Cahalan at last topical seminar

The fast reactor reactivity balance can be written as follows:

\[
\delta \rho = [P(t)-1] A + [P(t)/F(t) -1] B + [\delta \rho_{in}(t)] C + \delta \rho_{\text{external}}
\]

where

- \( P(t) \) = normalized reactor power
- \( F(t) \) = normalized core coolant flow
- \( \delta \rho_{in}(t) \) = change in coolant temperature at the core inlet
- \( \delta \rho_{\text{external}} \) = externally applied change in reactivity (control rods, etc.)

- the relative importance of each of these terms is determined by the grouped reactivity feedback parameters, \( A, B, \text{ and } C \)
Reactivity Feedback Coefficients

The reactivity feedback coefficients that form the three parameters A, B, and C are associated with the reactor core, and depend on fuel type, fuel volume fraction, coolant volume fraction, etc.,

\[
A = \alpha_{\text{Doppler}} \Delta T_{\text{FC}}(t=0)
\]

\[
B = \left[ \alpha_{\text{Doppler}} + \alpha_{\text{NaCoolant}} + \alpha_{\text{AxialExp.}} + a_1 \alpha_{\text{RadialExp}} + a_2 \alpha_{\text{ControlRod}} \right] \Delta T_{\text{C}}(t=0)/2
\]

\[
C = \left[ \alpha_{\text{Doppler}} + \alpha_{\text{NaCoolant}} + \alpha_{\text{AxialExp.}} + b_1 \alpha_{\text{RadialExp}} + b_2 \alpha_{\text{ControlRod}} \right] \Delta T_{\text{C}}(t=0)/2
\]

where \(\alpha_{\text{Doppler}}\) = Doppler coefficient

\(\alpha_{\text{NaCoolant}}\) = Sodium coolant density coefficient

\(\alpha_{\text{AxialExp.}}\) = Fuel axial expansion coefficient

\(\alpha_{\text{RadialExp}}\) = Core radial expansion coefficient

\(\alpha_{\text{ControlRod}}\) = Control rod driveline expansion coefficient

\(\Delta T_{\text{FC}}(t=0)\) = Steady-state temperature difference, fuel to coolant

\(\Delta T_{\text{C}}(t=0)\) = Steady-state coolant temperature rise, inlet to outlet

\(a_1, a_2, b_1, b_2\) = geometric parameters
**Passive Safety Criteria**

Criteria established for acceptable asymptotic core outlet temperatures for possible unprotected accident scenarios

- \( \frac{A}{B} < 1 \) for passive control of pump and BOP-induced accident scenarios

- \( 1 < \frac{C \Delta T_c}{B} < 2 \) for LOF, pump overspeed and chilled inlet

- \( \delta \rho_{TOP}/B < 1 \) for TOP performance

- Comparison of the whole-core reactivity coefficients to these criteria gives and indicated that the concept has favorable passive safety features
  - Detailed safety analyses required to confirm performance and margins
Summary and Conclusions

- Fast reactor physics are quite different from thermal reactor behavior
  - Better neutron balance (flexible actinide management)
  - Higher enrichment is required to compensate U-238 capture
  - Neutron leakage is increased

- Typical fast reactor core designs were reviewed
  - Traditional blanketed breeder, moderate burner with no blanket, low
    conversion ratio option (high fuel enrichment) configurations
  - Reactor performance, high power density, burnup, fluence
  - Reactivity compensation and control rod requirements

- Reactivity coefficients were discussed
  - Expansion coefficients prominent because of high leakage
  - Positive sodium density (and void) coefficient
  - Overall favorable passive performance for complete set of feedbacks
Sodium Fast Reactor

Safety #1

DOE/HQ June 20, 2007

NRC/White Flint June 21, 2007

(Rev. 1, October, 2008)

Presented by: Jim Cahalan
Notice

- This presentation has been revised to remove citations to and information from Applied Technology documents.
**FFTF References**

- C. P. Cabell, A Summary Description of the Fast Flux Test Facility, HEDL-400, December, 1980.
CRBRP References


SAFR and PRISM References

General References

- 10CFR50 Appendix A, General Design Criteria for Nuclear Power Plants
- DOE O 5480.30, Nuclear Reactor Safety Design Criteria
Outline

- Safety Issues #1 (Today)
  - Safety issues; DBA, BDBA, severe accident historically (coolant void, recriticality, FCI), passive safety
  - Safety analysis past results (FFTF, CRBRP, SAFR, PRISM)
  - Safety analysis methods; reactor, structural, coolant aerosols, containment

- Safety Issues #2 (Future)
  - Licensing issues in FFTF, CRBRP, SAFR, PRISM. International experience in perspective, high profile issues in Phenix, SuperPhenix, MONJU, BN-350, EBR-1, FERMI explained and lessons learned.
  - Inherent passive safety characteristics of sodium fast reactor systems (wrt loss of flow without scram, etc.). Inherent reactivity shutdown, natural circulation decay heat removal
  - Safety testing results, EBR-II SHRT, FFTF ULOF (GEMS); TREAT
Basic Safety Objectives

- Protect public health and safety
  - Reduce the risk from releases of radioactivity to acceptable levels
  - Comply with regulatory requirements and provide additional margin
- Protect plant worker health and safety
  - Provide a safe working environment and reduce risk of injury
  - Comply with regulatory requirements and promote worker protection
- Protect the environment
  - Provide a design that complies with all federal, state, and local requirements
  - Build, operate, and decommission the plant in a way that preserves environmental quality
- Protect the plant investment
  - Provide plant designs, equipment, and operating/maintenance practices to preserve investor equity and return reward
  - Maintain product quality and reliability
Radiation Protection Mechanisms

- **Barriers**
  - Contain radioactive materials and prevent human exposure or release to the environment

- **Distance**
  - Provide spatial margins to reduce the intensity of radiation exposure

- **Time**
  - Isolate radioactive material until it has decayed to a stable or less harmful state
Safety by Design

- By design, the plant and all its systems are configured and constructed in a manner that assures safe, stable, and reliable operation while preserving protection mechanisms.

- Engineered safety systems are provided to *prevent* development of conditions that can defeat the basic radiation protection mechanisms, and to *mitigate* the consequences of equipment failure or inappropriate operator actions.
  - Inherent protection margins (No operator action or equipment activation needed) can be provided by selection of materials and arrangement of components.

- The design principle of *defense in depth* is applied for important safety-related functions:
  - Containment
  - Reactor shutdown
  - Residual heat removal
Defense in Depth

- Plant functions that are important to preservation of safety protection mechanisms are designed according to the defense in depth principle, which provides multiple layers of safety assurance
  - Level 1: Provide a conservative design with large safety margins that can be constructed and operated normally without challenges to safety limits
  - Level 2: Provide additional design features that protect against a single, unlikely fault (~once in the plant lifetime). (Limited damage, minor repair).
  - Level 3: Provide additional design features that protect against a single, extremely unlikely fault (not expected in the plant lifetime). (Extensive damage, major repair)
- Foreseen events at Levels 1, 2, and 3 are within the plant safety design basis, and the most demanding events are usually identified as Design Basis Accidents (DBAs)
Defense in Depth Safety Design Features

- For key safety functions (maintain barriers, keep radioactivity at a distance, and provide time for recovery)
  - Multiple, diverse, and independent structures, systems, or components, each of which is capable of achieving the defined safety function
- Redundancy, diversity, and independence assure that not all safety function can be lost due to a single failure
- **Safety grade** systems, components, and systems are designed and maintained to criteria that assure their reliable operation
  - Quality assurance, inspection, testing, repair, ...
  - Seismic, electrical supply, ...
Safety Design Features

- Containment of radioactive material by multiple physical barriers
  - Fuel cladding
  - Primary coolant system boundary
  - Containment structure

- Reactor shutdown by multiple reactor control and protection systems
  - Primary scram system for startup, shutdown, reactor power changes, and power distribution management
  - Secondary scram system, always available for activation

- Residual heat removal by multiple heat transport paths and systems
  - Normal heat removal system (SG, condenser)
  - Dedicated shutdown heat removal systems (HXs, forced or natural circulation, from primary or secondary coolant, through vessel wall)
Sodium Fast Reactor Characteristics

- Fast spectrum, compact core design (400 MW FFTF example)
  - High power density (~400 kW/liter, PWR 100 kW/liter)
  - High coolant velocity (~20 ft/sec)
  - High linear pin power (~8 kW/ft, PWR 5 kW/ft), short core height (~3 ft, PWR 12 ft)
  - Small fuel pins (~0.23 in OD, PWR 0.37 in) on a tight triangular pitch (P/D~1.2, PWR ~ 1.5 sq) grouped in hexagonal ducted fuel assemblies
  - Oxide (FFTF) or metallic (EBR-II) fuel with a high fissile content (~20%, PWR ~3%)
  - Core coolant rise ~300°F (PWR ~63°F), outlet ~980°F (PWR 620°F) (Na NBP ~1638°F)

- Low pressure (~atmospheric) coolant
  - Large temperature margin to boiling (~4X Power/Flow Ratio in typical reactor designs for flow transients at core outlet pressure)
  - Pool (EBR-II) or loop (FFTF) primary system design, with backup guard vessel and piping to assure core cooling if the primary vessel leaks
  - Intermediate heat exchanger isolates radioactive sodium within containment, transfers heat to second sodium loop which transports heat to steam generator (or for FFTF, air dump heat exchangers) outside containment
Fast Flux Test Facility (FFTF) Chronology

- Conceptual design studies 1966-1969
- PSAR submitted by HEDL in September 1970
- Initial construction authorization in September 1971; full construction authorization in March 1972
- ACRS letter in May 1973
- FSAR submitted by HEDL in December 1975
- Construction complete/Na fill 1978
- Criticality February 1980, full power October 1980
- Research operations April 1982 to April 1992
- DOE Shutdown order December 1993
**FFTF Design**

- **Mission:** Provide a prototypic LMFBR operating environment for testing and development of fuels, materials, and components
  - Secondary: Develop design and construction experience
- **400 MWt, MOX fuel (22% and 27% Pu), three loop primary system, three intermediate sodium loops to air dump heat exchangers**
- **Reactor core:** 73 fuel assemblies, 9 control assemblies, 9 test assemblies
- **Coolant inlet 680°F (360°C), outlet 980°F (527°C)**
- **10psi steel containment**
- **Two independent reactor shutdown systems (both by moveable rods)**
- **Natural convection decay heat removal through three independent loops**
  - Pony motors on primary and secondary pumps
- **Core physics and structural design for inherent negative power and temperature reactivity feedbacks**
FFTF Site – Hanford, Washington
FFTTF Containment Building View
FFTFT Loop-Type Primary and Secondary Systems
FFTFR Reactor and Vessel Design
FFT F Construction View

- Simulated core assemblies
- Two of three instrument trees in place
FFTTF Driver Fuel Pin
FFT F Driver Fuel Assembly

- Duct
- Above Core Load Pad
- Handling Socket
- Fuel Pin Bundle Assembly
- Shield-Orifice Block
- Floating Collar (Top Load Pad)
- Nozzle Assembly
- Coolant Inlet Ports
- Piston Ring
FFT Core Layout

INSTRUMENT TREE #1 COVERAGE

SECTOR 1

IVHM #1 COVERAGE

300°

180°

IVHM #3 COVERAGE

180°

IVHM #2 COVERAGE

SECTOR 2

INSTRUMENT TREE #3 COVERAGE

SECTOR 3

0°

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Argonne National Laboratory
Clinch River Breeder Reactor Plant (CRBRP) Chronology

- March 1972. Two not-for-profit organizations established. Project Management Corporation (PMC) for project management, and Breeder Reactor Corporation (BRC) for utility industry liaison.
- November 1972. Westinghouse-Advanced Reactor Division (W-ARD) selected as lead reactor manufacturer, Burns and Roe (B&R) as A/E.
- February 1973. Initial work authorization. AI and GE added to team.
- PSAR submitted April 1975 (Updated through Rev. 77 May 1983)
- Licensing suspended: commercial reprocessing banned March 1977
- Licensing resumed September 1981
- Funding stopped October 1983
CRBRP Design

- **Mission**: Demonstrate the safe and reliable operation of an LMFBR in a utility environment. Demonstrate LMFBR economics, and the transition from technology development to commercial operation
- 975 MWt, 380 MWe (gross), MOX fuel (22% and 32% Pu), three loop primary system, three intermediate sodium loops to steam generators
- Reactor core: 198 fuel assemblies (108 inner/90 outer), 19 control assemblies (15 primary/4 secondary), 150 radial blanket assemblies
- Coolant inlet 730°F (388°C), outlet 995°F (535°C)
- 10psi steel containment
- Two independent reactor shutdown systems (both by movable rods)
- Decay heat removal through three independent loops
  - Pony motors on primary and secondary pumps
  - Auxiliary decay heat removal through water side of SG
- Direct decay heat removal system independent of HTS loops
- Core physics and structural design for inherent negative power and temperature reactivity feedbacks
Clinch River Breeder Reactor Plant (CRBRP)
SAFR and PRISM Background and Chronology

- Beginning in the early/mid 1980’s, DOE funded conceptual design studies for modular advanced liquid metal reactor plants
  - At Rockwell International, the Sodium Advanced Fast Reactor (SAFR)
  - At General Electric, the Power Reactor - Inherently Safe Module (PRISM)
- Preliminary Safety Information Documents for SAFR and PRISM were submitted to NRC in November, 1986
- The initial SAFR and PRISM concepts focused on innovative design approaches for economics and safety
  - Design simplification based on passive safety performance
- DOE subsequently (ca 1988?) selected the PRISM concept for continued development in the Advanced Liquid Metal Reactor (ALMR) program
- NRC issued pre-application SERs for SAFR (1991) and PRISM (1994)
- DOE support for PRISM design studies ceased with the cancellation of the ALMR program in 1994
**SAFR Design**

- Multiple (4) power units per site co-located with a spent fuel processing facility
- 900 MWt reactor, U-Pu-Zr metallic fuel, pool type primary system, two intermediate loops
- 60 year plant life
- Reactor core: 96 driver fuel assemblies, 46 internal and 48 radial blanket assemblies, 6 control assemblies, 3 safety assemblies
- Coolant outlet 950°F (510°C), inlet 675°F (357°C)
- Inherent response for emergency reactor shutdown
  - Inherent reactivity feedbacks in temperature and flow transients
  - Self-Actuated Shutdown System (SASS); thermally-activated magnetic latch release
- Two natural circulation decay heat removal systems
  - Direct heat removal from hot sodium pool (DRACS)
  - Ambient air cooling of guard vessel (RACS)
- Compact containment building design
**PRISM Design**

- Multiple power modules (6) co-located with a spent fuel reprocessing facility (Module reactor size grew as the concept evolved, to 840 MWt in 1995. The SER looks at the 471 MWt design)
- 840 MWt, U-Pu-Zr metallic fuel, pool-type primary system, two intermediate loops
- Reactor core (burner): 192 fuel assemblies, 10 control assemblies, 3 safety assemblies
- Coolant outlet 930°F (500°C), inlet 680°F (360°C)
- One safety grade automatic reactor protection system, with a manually operated safety grade ultimate shutdown system (3 safety rods)
  - Inherent accommodation of ATWS transients without core melt, significant reactivity addition, or large radiological release
- Shutdown cooling by turbine bypass with emergency removal systems
  - Air cooling system (ACS) on the steam generator shell
  - Primary sodium auxiliary cooling system (PSACS); reqs. valve action
  - Reactor vessel air cooling system (RVACS)
- Compact containment shell design
**Special Safety Considerations for SFRs -- 1**

- **Low pressure, chemically reactive coolant**
  - For pool-type design, all radioactive sodium contained within reactor vessel, with secondary guard vessel for leak protection
  - For loop-type design, guard vessels and double piping; equipment in steel-lined, inerted cells within containment
  - Guard vessels and double pipes fulfill ECCS role for core coverage

- **Reactor shutdown and residual heat removal requirements**
  - Diverse, redundant, independent, safety grade systems
  - Potential for natural circulation designs due to large thermal heat capacity of coolant systems for time margin, excellent sodium heat transfer and buoyancy properties for natural convection

- **Containment loading mechanism for design basis**
  - Low pressure coolant, leak-before-break
  - Sodium oxidation with air; heating, aerosol generation, pressurization
Special Safety Considerations for SFRs -- 2

- Low pressure, liquid metal boiling
  - Only relevant for beyond design basis power, flow, or heat removal accident sequences
  - Flow regime: low pressure and high heat flux leads to rapid vapor formation, large vapor bubbles that fill the hexcan radially and move upwards due to buoyancy or forced liquid flow
  - Vapor bubbles condense in unheated, above-core region
  - Moving vapor bubbles leave behind a relative thick liquid film (~15% of original vol. frac.); sodium wets stainless steel at high temperature
  - Liquid film absorbs heat from cladding, and evaporates; can also be stripped by high vapor velocities
  - High two-phase pressure drop with boiling can stop assembly liquid flow (parallel flow system)
  - Boiling can remove low power (< 1% ?), but higher heat fluxes lead to coolant voiding, followed by cladding dryout, temperature increase, and cladding failure

- Coolant voiding may introduce significant positive reactivity, depending on the core design
Sodium Boiling Model Features (SAS4A)
Special Safety Considerations for SFRs -- 3

- **Fuel melting and relocation**
  - Only relevant for beyond design basis power, flow, or heat removal accident sequences (Unprotected = Without reactor scram)
  - Without cooling, cladding failure and fuel melting will occur rapidly, within seconds for full power levels
  - Modes of cladding and fuel melting and relocation depend on reactor power and coolant flow conditions, and type of fuel (oxide or metal)
    - *Unprotected transient overpower accident (UTOP)*
    - *Unprotected loss-of-flow accident (ULOF)*
    - *Unprotected loss-of-heat-sink accident (ULOHS)*

- **Fuel compaction in a fast reactor will introduce significant positive reactivity**
  - Fast reactors have a fuel inventory of several/many critical masses, depending on design
  - The reactivity state of the reactor is sensitive to small (thermal expansion) fuel relocations
  - Gross fuel motion in accidents can yield energetic power excursions
Special Safety Considerations for SFRs -- 4

- **UTOP Accident Sequence**
  - External reactivity insertion (i.e. control rod withdrawal) without scram; time scale depends on insertion rate
  - Reactor power increases with full coolant flow; inherent reactivity feedback mechanisms may limit power increase
  - At around 4X normal power, cladding will rupture locally, and molten fuel will discharge through the cladding failure

- **ULOF Accident Sequence**
  - Coolant flow coastdown without scram; time scale depends on coastdown rate
  - Outlet coolant temperature rises with flow coastdown; inherent reactivity feedback mechanisms may reduce power
  - At around 4X Power/Flow, coolant boils, and voiding leads to loss of fuel cooling and reactivity increase
  - As fuel and coolant temperatures increase, melting and relocation will occur
Special Safety Considerations for SFRs -- 5

- **ULOHS Accident Sequence**
  - Loss of heat removal (e.g. SG feedwater) propagates a temperature increase through the coolant systems that eventually results in a reactor inlet temperature increase (pumps still running); slow time scale (minutes) due to large coolant systems heat capacity
  - Coolant temperature increases with inlet temperature rise; inherent reactivity feedbacks may reduce reactor power
  - As coolant temperature increases to boiling, bubbles formed are swept out of the core by pump head, until local boiling pressure drop increase overcomes pump (parallel flow system)
  - Coolant void formation can lead to fuel heating and melting

- **Fuel relocation implications**
  - Initially compactive motion will yield positive reactivity; dispersive motion will yield negative reactivity
  - Extended ex-core fuel motion will yield large shutdown reactivity
  - If the fuel is coolable, relocation will cease
  - If sufficient fuel collects ex-core, recriticality may occur
Initiating Phase Fuel Relocation Model Features (SAS4A)
**Inherent Protection in Unprotected SFR Accident Sequences**

- After the FFTF and CRBRP era, continued R&D identified the potential for SFR design options that provide inherent reactivity feedback mechanisms to reduce fission power and natural circulation cooling options to remove heat in ULOF, ULOHS, and UTOP accident sequences
  - Metallic fuel; high thermal conductivity, lower operating temperature, less ‘stored’ reactivity to overcome for power reduction to decay heat
  - Sodium coolant natural circulation decay heat removal

- EBR-II tests of ULOF and ULOHS accident sequences from 100% power and flow initial conditions demonstrated inherent protection potential with metallic fuel and sodium coolant

- FFTF ULOF tests from 50% power with GEMS

- EBR-II and FFTF test results covered in future presentation

- SAFR and PRISM designs incorporated inherent protection design features to eliminate coolant boiling, cladding failures, and fuel melting in ULOF, UTOP, and ULOHS accident sequences
Regulatory Reviews for Safety Design Assurance

- Sodium fast reactors have design features to prevent severe accident progression, and to mitigate the consequences of extremely unlikely accidents
  - Safety by design, Defense in Depth
  - Reactor scram, shutdown heat removal, containment

- Past regulatory reviews of SFR projects
  - FFTF: SAR, SER, ACRS letter
  - CRBRP: SAR, SER, “met CP requirements”
  - SAFR: PSID, Pre-application SER
  - PRISM: PSID, Pre-application SER

- Safety review basis provided by the SAR
  - Design development and safety review proceeds from the General Design Criteria
General Design Criteria - 1

- 10CFR50 Appendix A lists criteria in six areas for an LWR
  - Overall Requirements (1 through 5)
  - Protection by Multiple Fission Product Barriers (10 through 19)
  - Protection and Reactivity Control Systems (20 through 29)
  - Fluid Systems (30 through 46)
  - Reactor Containment (50 through 57)
  - Fuel and Radioactivity Control (60 through 64)

- ANS 54.1 lists criteria in the same areas for a liquid metal-cooled reactor (LMR)
  - Some criteria deleted/added/changed compared to 10CFR50 because of generic design differences

- DOE O 5480.30 lists safety design criteria for a DOE reactor
  - Criteria written to apply to all DOE reactors

- Cross comparison of individual criteria provided in Appendix A
General Design Criteria - 2

- **LMR Design Characteristics (compared to LWR)**
  - Low pressure coolant; “leak before break”; reduces DBA focus on pipe break and loss-of-coolant; eliminates ECCS requirement by supplying secondary guard pipes and vessels to maintain core coverage in the event of leaks
    - *Loop vs. Pool arrangement*
  - Chemically reactive reactor coolant; adds requirement for leakage detection and multiple barriers to mitigate consequences; cell liners and inert cell atmosphere for loop-type primary system; fire suppression systems; steam generator instrumentation and pressure relief
  - Sodium heating to prevent freezing
  - Enhanced potential for natural circulation residual heat removal
  - Enhanced potential for inherent protection against reactivity and cooling faults
Top level safety requirements for an LMR are the same as for an LWR

- Diverse, independent, redundant reactor shutdown systems
  - *Normally, two safety-grade reactivity shutdown systems; independent instrumentation and power supplies*
  - *Role for inherent reactivity feedback protection?*

- Diverse, independent, redundant residual heat removal systems
  - *Normally, two safety-grade decay heat removal systems; independent instrumentation and power supplies*
  - *For diversity and reliability, one of the systems may be a natural circulation system*

- Multiple barriers for release of radioactivity
  - *Fuel cladding, primary coolant boundary, containment building*
  - *Role for reduced volume containment, or alternative confinement system?*
Domestic and international experience has demonstrated the feasibility of successful LMR licensing
- Design flexibility provided by sodium coolant properties in normal operation

Potential severe accident safety issues associated with positive coolant voiding reactivity or core melt scenarios have been successfully treated as beyond-design-basis phenomena
- Severe accident probability reduced through conservative design, large safety margins, and adherence to defense-in-depth design philosophy
- Analysis of severe accidents with “best estimate” (not conservative) modeling assumptions to provide measure of thermal and structural margins beyond the design basis; significant resources applied
Is there a design basis role for LMR inherent safety characteristics?

- With sodium cooling, reactor thermal safety margins are already large
- Emphasis of self-protection design features can utilize natural safety margins to provide enhanced reactivity feedback and reactor cooling mechanisms to further reduce the probability of severe accidents for faulted initiators
- Enhanced natural circulation decay heat removal at high temperatures
- Inherent protection in under-cooling (loss-of-coolant-flow or loss-of-heat-sink) or over-power (reactivity insertion) accidents without scram
SAR Chapter 15 Accident Analysis Example: Clinch River Breeder Reactor Plant (CRBRP) Safety Design Approach and Design Basis Accident (DBA) Classification

- Design Level 1: Normal Operation. Prevention of accidents by intrinsic features of design, construction, and operation, including quality assurance, redundancy, testability, and maintainability.
- Design Level 2. Anticipated Faults and Unlikely Faults. Provide design features to protect against errors or malfunctions.
- Design Level 3. Extremely Unlikely Faults. Provide extra protection in the design basis to assure public safety for events not expected to occur in the life of the plant.

In Chapter 15, accident sequences classified as “Anticipated”, “Unlikely”, or “Extremely Unlikely” are defined and analyzed as design basis events

- Accident analysis techniques include consideration of uncertainties
Chapter 15 Accident Analysis (cont.)

- The CRBRP DBAs are listed in Appendix B. 17 Anticipated Events, 21 Unlikely Events, 23 Extremely Unlikely Events
- The events are design specific
- Classification of events taken from RDT Standard C-16-1
- Anticipated: Faulted event that may occur once or more in the plant life, no damage, return to operation after fault correction
- Unlikely: Faulted event not individually expected during the plant life, but one or more events in whole category may occur in the plant life, some damage requiring repair for return to operation
- Extremely unlikely: Faulted event with such a low probability that no events in the whole category are expected in the plant life, damage sufficient preclude resumption of operation but no loss of safety function
**Safety Documentation for Licensing - 3**

- DBA acceptance criteria are established for affected systems, structures, and components
  - Criteria based on performance requirement for a class of events, usually expressed as a limiting temperature or mechanical limit
  - For some structures and components, may use ASME Code

- CRBRP acceptance criteria example for fuel pins
  - Basis:
    - *maintain cladding integrity in Anticipated and Unlikely faults*
    - *maintain coolable geometry in Extremely Unlikely faults*
    - *limiting criteria based on enveloping thermal/mechanical analysis*
  - Anticipated fault: no fuel melting and cladding temperature < 1500°F
    - *Fuel melting ~ 4800°F (MOX fuel)*
    - *Cladding melting ~ 2500°F (316 SS)*
  - Unlikely fault: no fuel melting and cladding temperature < 1600°F
  - Extremely unlikely fault: no cladding melting or coolant boiling
    - *Coolant boiling ~ 1800°F (Reactor outlet pressure)*
In the SAR Chapter 15 presentation, it is demonstrated that the selected set of design basis accidents result in reactor and plant design compliance with the acceptance criteria.

- In a successful regulatory review, both applicant and regulatory agree that the set of DBAs envelope and characterize the satisfactory performance of the safety design, in compliance with the general design criteria and applicable regulations.
Beyond Design Basis -- Core Disruption Accidents

- The role of core disruption accidents in licensing was addressed by Fauske, 1976, during the CRBRP regulatory review
  - The consensus view (industry, regulator, and R&D) was that core disruption accidents (CDAs) should be considered and analyzed in the design review and licensing process, but that their probability of occurrence should (and must) be reduced to a level that justifies exclusion from the design basis.
  - Consequence assessment should be made on a best-estimate basis using ‘realistic’ data and methods rather than arbitrarily conservative assumptions.

- In earlier analyses for FFTF (See Appendix A of the FFTF FSAR) the case had been made for a ‘conservative upper bound HCDA case’
  - Consequence assessment was used to estimate barrier challenge, containment performance, and radiological consequences

- For CRBRP, beyond design basis structural and thermal margins were assessed in relation to calculated accident energetics
FFTF Core Disruption Accident Analysis -- 1

- Documented in FSAR Appendix A
- Analyzed bounding UTOP and ULOF accident sequences for estimation of consequences and demonstration of the ability of the containment to provide protection of public health and safety
  - UTOP initiators: 0.03 $/s (max. by design) to 0.5 $/s without scram
  - ULOF initiators: complete loss of primary pumping without scram
- Initiating phase accident analyses tracked with detailed modeling to the point of initial fuel melting and dispersal
  - Reactor power reduction, but for the ULOF sequence, the reactor state may not be coolable, leading to further melting and transition to whole core melting
- Whole core disruption phase analyzed with scoping tools to assess modes and rates of fuel relocation, assessment of associated reactivity transients, and resulting energy deposited in the fuel
FFTFT Core Disruption Accident Analysis -- 2

- Work-energy analysis to assess conversion of fuel energy to pressure loading of reactor structures
- Mechanical consequences assessment to determine damage to structures and reactor vessel
- Post-accident heat removal assessment to scope final core debris stability and coolability
- Radiological assessment to scope radioactivity release

Conclusions
- Neither the UTOP nor the ULOF lead to significant public risk
- UTOP: Initial cladding failures, fuel sweep-out and dispersal, remainder of core is coolable in place, fuel within intact reactor vessel
- ULOF: Whole core disruption and dispersal, moderate pressure transient causes minor coolant (“~300 lbs”) and fuel (“<1%”) loss from vessel, containment structure remains intact (0.1 % per day leak rate)
- 30 day dose at site boundary is >100X below guidelines for reactor siting
FFTF Core Disruption Accident Analysis Scenarios
CRBRP Core Disruption Accident Analyses --1

Initially document in PSAR Appendix D
- PSAR Appendix D was deleted in Amendment 24, July 1976
- Core Disruption Accidents were excluded from the design basis, see papers by Strawbridge and Slember
- The limiting design basis accident for containment challenge was PSAR 15.6.1.1 Primary Sodium In-Containment Storage Tank Failure During Maintenance (See Appendix B)

However, the subject of core disruptive accidents and their consequences were extensively analyzed and discussed by the project and NRC. (At one point, the CDA issue determined the critical path for the licensing schedule)
- NRC discussion in SER Appendix A
- Project Reports:
  - CRBR-3, Hypothetical Core Disruptive Accident Consideration in CRBRP, Volumes 1 and 2, Revision 4, 1982
Extensive analyses were performed by General Electric (using Argonne codes and data) and by Argonne Natl. Lab.
CRBRP Core Disruption Accident Analyses --3

- Compared to FFTF, the issue in CRBRP was the larger core size (975 MWt vs 400 MWt), and the implications for energetics
  - ~2.5X initial core power in the same diameter reactor vessel
  - ~3.5 $ coolant void worth compared to ~0 in FFTF

- The role of the larger coolant void reactivity worth altered the ULOF scenario
  - In FFTF, coolant boiling and voiding had little reactivity effect, so initial fuel disruption occurred at near nominal power in a ‘dry’ core
  - In CRBRP, coolant boiling and voiding introduced positive reactivity and raised the power level, so cladding failure and initial fuel disruption occurred at elevated power (~10X P₀) in a core with significant liquid sodium (and corresponding reactivity) still present
  - CDA attention was focused on the ULOF and the potential for initiating phase energetics

- Attention on the CDA issue was emphasized by the role of CRBRP as a prototype demonstration design
Fauske (1976) Illustration of Work Energy Estimates
Example CRBRP ULOF Analysis Results

- Nominal base case results for best-estimate phenomenological assumptions (similar to FFTF analysis assumptions) yields a non-energetic initiating phase
  - Peak power $\sim 12 \times P_0$, peak reactivity $\sim 0.8 \beta$

- Bounding case results
  - Neglect of reactivity feedback from axial thermal expansion
  - “Pessimistic” modeling assumptions for initial fuel dispersal
  - “Pessimistic” modeling of cladding failure and subsequent fuel motion in low power fuel that fails late into unvoided or partially voided coolant channels (‘Fuel-coolant interaction’ or ‘LOF-driven-TOP’)
  - Peak power $\sim 2750 \times P_0$, peak reactivity $\sim 1.06 \beta$, peak fuel temperature $\sim 6000 \text{ K}$, average core fuel temperature $\sim 4480 \text{ K}$, isentropic fuel expansion work energy $\sim 680 \text{ MJ}$ (CRBRP structural margin design basis $\sim 661 \text{ MJ}$)
Resolution of CRBRP CDA Issues for Licensing

- See 1985 papers by Strawbridge and Giitter, and NUREG-0968 Appendix A Section A.6
- NRC approved the project position on CDA energetics
  - Contingent on changes to the fuel pin design, the rotating plugs design, and analysis and testing results for cell liner performance
  - NRC sponsored independent analyses at LANL
  - Concluded that containment would not be challenged by missiles or spray fires, and radiological consequences would depend on the beyond-design-basis containment thermal margins to deal with sodium spills and hydrogen burns. NRC concluded that the design could adequately deal with such challenges, and that the risk from a CDA in CRBRP was not significantly different from the risks from typical LWRs.
SAFR and PRISM Safety Analyses

By mission, the SAFR and PRISM conceptual designs were developed under a mandate for innovation, modularity, and reduced costs.

This mission prompted new approaches to interpretation of the General Design Criteria and the Defense-in-Depth principle:
- Reliance on inherent mechanism in place of active systems for reactor shutdown and residual heat removal.
- With large initial (fuel and coolant) safety margins and very reliable inherent safety mechanisms, the probability of CDAs was dramatically reduced, and containment designs were adjusted accordingly.

The SAFR and PRISM projects engaged NRC and a dialog on begun. However, the designs did not reach full maturity, and the projects terminated with issues unresolved:
- See the SAFR and PRISM SERs, and the GE paper by Pat Magee.
SAFR and PRISM GDC, DBA, and BDBA

- The SAFR and PRISM PSIDs present the designer proposals for the GDCs, and the SERs document the NRC responses
  - Mostly in agreement, with some divergences
- The SAFR and PRISM projects supplied only minimal DBA analyses, because of the immaturity of the designs.
  - Designs were evolving, in part due to additional mission requirements and in part due to interactions with NRC
- Considerable emphasis was placed on passive safety accommodation of the ULOF, UTOP, and ULOHS accident sequences while maintaining substantial margins to prevent CDA progression and severe accident consequences
  - The combination sodium cooling, a pool-type primary system, and metallic fuel provides the potential for self protection mechanisms to prevent severe accident consequences
SAFR and PRISM Passive Safety Mechanisms

- **Pool-Type primary system**
  - All primary coolant confined to a single vessel with no external piping. Eliminates pipe break (leak) accident scenario (with appropriate redundancy in pump-to-inlet plenum connection)

- **Sodium coolant**
  - Low operating pressure eliminates pipe or vessel rupture possibility
  - With oxygen control, no corrosion of steel structural material
  - Superior heat removal properties due to high thermal conductivity, volumetric heat capacity, and thermal expansion for buoyancy (natural convection)

- **Metallic fuel**
  - High thermal conductivity, low operating temperature, low invested cold-to-hot reactivity to compensate for shutdown
  - Hard spectrum and high density promotes low burnup reactivity swings, minimizing control rod worths and external reactivity
Reactivity Swing for Power Reduction

Oxide Fuel
(Doppler Coeff. = -0.005)

~1.5 $

892°C BP

Metallic Fuel
(Doppler Coeff. = -0.003)

~0.3 $

510°C Outlet

360°C Inlet
Small Fast Reactor ULOF Power and Flow

- TOTAL POWER
- CHANNEL 3 FLOW
- DECAY POWER

NORMALIZED POWER OR FLOW

TIME, SEC.
Small Fast Reactor ULOF Reactivities

![Diagram showing small fast reactor ULOF reactivities](image-url)
Small Fast Reactor ULOF Temperatures

CHANNEL 3 TEMPERATURE, C

TIME, SEC.

Argonne National Laboratory
CDA Accommodation

- A sodium-cooled, pool-type, metallic fuelled reactor design can be configured to survive all ULOF, UTOP, and ULOHS accident initiators with substantial margin to coolant boiling, cladding failure, or fuel melting
  - In FFTF and CRBRP, these initiators lead to core disruption and the potential for energetics and containment challenge
  - In SAFR, PRISM, and similar designs, unprotected accident initiators result in reduced accident consequences, short of coolant boiling and fuel disruption

- Progression to severe accident consequences requires assumption of very low probability initiators (triple fault?)
  - Assessment of severe accident consequences may best be carried out within the PRA framework, not necessarily related to a deterministic accident sequence
Safety Analysis Methods

- See paper by Fauske (1976) for LMFBR models and codes
- FFTF and early CRBRP core disruption analysis was performed with the SAS3A (SAS3D) computer code at Argonne
  - HEDL used the MELT code for UTOP analysis (ULOF at ANL)
- The SAS4A code was developed at Argonne with modeling appropriate for initiating phase core disruption analysis of higher void worth core designs
  - Mainly one-dimension modeling appropriate until melting of hexcans
- The SIMMER code development began at about the same time at LANL for analysis of the ‘transition’ phase of the core disruptive accident; whole-core two dimensional model
  - The VENUS code had been developed earlier at ANL for analysis of the disassembly phase
- The SASSYS-1 code was developed later for whole-plant analysis
- Focus today on Argonne SAS4A and SASSYS-1 codes
SAS4A/SASSYS-1 Computer Code Background

- Developed at ANL for transient analysis of liquid metal reactor core disruption and systems analysis
  - Originally developed to support CRBRP licensing
  - Safety margin assessment in design basis accidents (DBA) and anticipated transients without scram (ATWS); SASSYS-1
  - Consequence assessment in severe accidents; SAS4A
- Development continued during the Integral Fast Reactor (IFR) program (metallic fuel)
- Heavy liquid metal coolant options added during the Advanced Accelerator Applications (AAA) program
  - Water versions developed in NPR and international programs
- Modeling validated by applications to testing data from TREAT, EBR-II, and FFTF
- World-wide standard code for LMR safety analysis; Germany, Japan, France, Italy, Russia; additional validation with CABRI test data
SAS4A/SASSYS-1 Modeling Features

- Single and multiple pin subassembly thermal hydraulics; multiple subassemblies for whole-core simulation
- Two-dimensional pin heat transfer
- Single and two-phase coolant dynamics (coolant boiling, fission gas release upon pin failure)
- Liquid metal coolants: Na, NaK, Pb, Pb-Bi
- Oxide and metallic fuel and cladding mechanics
- Reactor point and spatial kinetics with reactivity feedbacks; nodal diffusion and nodal transport options
- Primary and intermediate coolant systems thermal hydraulics with components (pumps, pipes, plena, valves, heat exchangers, steam generators)
- Steam power cycle with components (turbine, condenser, pumps, heaters, steam generators)
- Reactor and plant control systems
SAS4A Models for Initiating Phase Core Disruption Analysis

- **SAS4A: Simulation of initiating phase CDA**
  - Developed for higher-void-worth core applications (CRBRP); fuel disruption at above nominal power levels
  - Modeling validated with TREAT test data
  - Exported to Japan, France, and Germany; validation with CABRI test data

- **SAS4A Fuel Disruption Models**
  - Single-pin, multiple channel thermal hydraulics core model
  - DEFORM-4 (oxide) fuel pin mechanics model
  - Multiple slug/bubble coolant boiling model
  - CLAP cladding melting and relocation model
  - PLUTO2 fuel/coolant interaction model
  - LEVITATE fuel pin disruption model
SAS4A/SASSYS-1 Single Pin Model
SAS4A/SASSYS-1 Single-Pin Channel Model
SAS4A Fuel Pin Heat Transfer Model

- One-dimensional, radial heat transfer at many axial locations
- Radial conduction through the fuel and cladding; conduction and radiation across the fuel/cladding gap
- Convective boundary condition to the axially flowing coolant from the cladding and the structure
- Initial, steady state conditions for the coolant mass flow and the axially-dependent channel power
- Steady state channel pressure drop in all channels adjusted to match the peak channel pressure drop with addition of a channel-dependent inlet orifice coefficient
- Transient channel flows computed from time-dependent inlet-to-outlet plena pressure drop
Channel Radial Heat Transfer Mesh
DEFORM-4 Model

- Mechanistic simulation of oxide fuel and stainless steel cladding behavior during irradiation and in a transient
- Porosity migration, grain growth, fission gas release, fuel cracking with crack healing, fission-gas-induced swelling, irradiation-induced steel swelling, gas plenum pressurization, fuel-cladding gap conductance
- Fuel and cladding mechanical behavior, thermal expansion, molten fission gas release and pressurization, cladding failure
- Original ANL model was modified at FZK (Germany) to be more applicable to European reactor materials and designs
DEFORM-4 Fuel/Cladding Deformation Model
SAS4A Sodium Boiling Model

- Single and two-phase sodium coolant flow in pin bundles
- Low-pressure liquid sodium boiling: multiple slug/bubble flow regime
- Cladding wetting: liquid sodium film on cladding and hexcan wall
- Simulates bubble initiation and growth, bubble motion, void formation, film evaporation and stripping, liquid flow reversal and chugging
- Loss of heat removal and reactivity effect of voiding
- Model validated by analysis of laboratory and reactor tests (e.g. R5 TREAT)
SAS4A Sodium Boiling Model
CLAP Cladding Melting and Relocation Model

- Cladding melting and relocation following dryout of sodium film
- Motion due to coolant vapor flow (pressure gradient and shear forces) and gravity
- Molten cladding freezing and blockage formation
- Heat capacity loss with cladding motion
- Reactivity effect of steel removal from core
CLAP Model
PLUTO-2 Fuel Coolant Interaction Model

- Model for fuel motion following cladding failure with flowing liquid coolant
- Fuel (in- and ex-pin), fission gas, and coolant dynamics following cladding failure in a channel with flowing liquid sodium
- Detailed material dynamics (two-fluid) with particulate of continuous flow regimes
- Fuel freezing and plate-out
- Cladding rip elongation
- Model validated by comparison to TREAT tests
- Modifications performed by JNC for CABRI test analysis
PLUTO-2 Modeling
LEVITATE Fuel Pin Disruption Model

- Model for fuel pin disruption in voided coolant channels
- Fuel (in- and ex-pin), fission gas, cladding, and coolant vapor dynamics following cladding failure and disruption in a channel voided of coolant
- Detailed material dynamics (two-fluid) with multiple flow regimes
- Solid fuel fragmentation ("chunk" formation) and relocation
- Molten fuel ejection and freezing
- Modifications performed by JNC for CABRI test analysis
LEVITATE Modeling
SASSYS-1 Models -- 1

- SASSYS-1 modeling was originally developed for analysis of shutdown heat removal transients
  - Reactor and heat transport systems models for design basis and accident analysis
- Scope of analysis soon grew to cover the steam generators and balance-of-plant, and the control systems
- During the ANL Integral Fast Reactor program (1984-94) metallic fuel modeling was added, and SASSYS-1 was used to analyze ATWS events to assess passive safety performance
  - Extensive applications to EBR-II Shutdown Heat Removal Tests and FFTF Inherent Safety Tests (circa 1986)
  - EBR-II real time simulator
  - Concept evaluations in the IFR program, support to SAFR and PRISM
SASSYS-1 Models -- 2

- SASSYS-1 models include
  - Multiple-pin subassembly model
  - PRIMAR-4 coolant systems model
  - DEFORM-5 metallic fuel cladding failure model
  - SSCOMP metallic fuel properties model
  - FPIN-2 metallic fuel pin behavior model
SASSYS-1 Multiple Pin Subassembly Model

Steady-state and transient calculation of fuel, cladding, coolant, and structure temperatures and coolant flow rates

- Basic model applicable to sodium, lead, lead-bismuth, helium, and water coolants given appropriate constituent data
- Triangular pitch/hexagonal geometry or square pitch/square geometry.
- With or without subassembly wrapper can
- Coolant flow parallel to pins and between sub-channels driven by pressure differences and flow sweeping due to wire wrap
  - Forced convection and buoyancy-driven natural circulation
- Solve mass, energy, and momentum equations for coolant
  - Constitutive relations for cross-sub-channel mass flow and energy flow (turbulent mixing and thermal conduction)
- Solve conduction energy equation in fuel, cladding, and structure
SASSYS-1 Multiple-Pin Subassembly Model

EBR-II XX09 Instrumented Subassembly

SASSYS-1 Model of XX09
SASSYS-1 Multiple-Pin Subassembly Coolant Temperature Results
**PRIMAR-4 Model**

- Thermal and hydraulic model for primary and intermediate sodium coolant loops
- General arrangement of volumes connected by segments
- Volumes are perfectly mixed, compressible liquid with or without cover gas
- Flow segments divided into elements for hydraulics simulation and into temperature groups for heat transfer
- Special models for pumps, valves, heat exchangers, steam generators
- Explicit core by-pass channels for control, blanket, and reflector assemblies
- Component-to-component heat transfer
- Explicit decay heat removal simulation: DRACS, RVACS, etc.
- Annular flow elements for IHX bypass flow
PRIMAR-4 Model of a Small Pool-Type Reactor
DEFORM-5 Metallic Fuel Cladding Failure Model

- For stainless steel clad metallic fuel pins, predicts
  - Margins to cladding failure in DBA and BDBA transients
  - Cladding failure time and location in severe accident transients
- Simulates transient cladding mechanical response taking into account
  - Fuel and cladding temperatures
  - Fission gas pressure loading
  - Eutectic thinning of cladding at fuel/cladding interface
- Failure prediction based on plastic cladding strain and strain rate
  - Cladding damage based on evaluated life fraction correlation used in FPIN-M
  - Calculated cladding failure predictions show excellent agreement with TREAT test data
SSCOMP Metallic Fuel Properties Model

- Correlation of Integral Fast Reactor (IFR) program metallic fuel properties database
  - Enthalpy, specific heat, density, thermal expansion, and thermal conductivity for binary (U-Zr) and ternary (U-Pu-Zr) metallic fuel
  - Burnup/porosity corrections and uncertainties for thermal conductivity

- Automated data interpolation among experimental data for composition corrections

- Integrated implementation in SASSYS-1 fuel heat transfer and constituent models as an alternative to oxide correlations and table input for properties

- Basis for integration of future metallic fuels data and phenomenological models
FPIN-2 Metallic Fuel Pin Behavior Model

- Validated model for fuel pin fuel and cladding mechanical behavior developed by John Kramer and Tom Hughes
  - Integrated with SASSYS-1 by Tanju Sofu
  - Driven by SASSYS-1 calculated fuel and cladding temperatures
- Models elastic, plastic, thermal, and swelling phenomena
  - Rigorous mechanical model based on force-displacement formulation
  - Equilibrium equations derived from equations for virtual work
  - Implicit finite element solution method using linear shape functions
- Additional metallic fuel models for fission gas generation and release, molten cavity formation, gas plenum, fuel-cladding eutectic formation
- Cladding rupture predicted using a life fraction formulation based on measured data stress-strain data
In closing…

- Liquid sodium was initially selected as the coolant for fast reactors on the basis of 1) low pumping power requirement, 2) high volumetric heat capacity, and 3) high thermal conductivity.
  - Other favorable properties include low neutron absorption and very good compatibility (negligible corrosion) with structural materials.

- Many reactor years (~300) of experience in the US and internationally have demonstrated the effectiveness of the liquid metal fast reactor concept for electricity generation and fuel production, as originally conceived by Fermi, Szilard, and Wigner.

- The US reactor development program has also demonstrated that liquid metal cooling contributes to excellent inherent safety reactor performance, through
  - Natural circulation decay heat removal
  - Passive reactor power reduction in beyond-design-basis accidents

- Based on several decades of liquid metal fast reactor research and development, the concept has achieved the level of technical maturity necessary for commercial deployment.
Appendix A
General Design Criteria Cross-Reference between
10CFR50 Appendix A, ANSI/ANS 54.1, and
DOE Order 5480.30
## Appendix A GDC Comparison - 1

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# Appendix A GDC Comparison - 2

## II. Protection by Multiple Fission Product Barriers

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Appendix B
Listing of Design Basis Accidents for The Clinch River Breeder Reactor Plant
PSAR Chapter 15
## Appendix B  CRBRP PSAR Table 15.1.3-2 Accident Events - 1

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Sodium Fast Reactor

Safety #2

DOE/HQ October 31, 2007

NRC/White Flint November 1, 2007

(Rev. 1, October, 2008)

Presented by: Jim Cahalan
Notice

- This presentation has been revised to remove citations to and information from Applied Technology documents.
FFTFT References

- C. P. Cabell, A Summary Description of the Fast Flux Test Facility, HEDL-400, December, 1980.
CRBRP References

SAFR and PRISM References


EBR-I, Fermi References


BN-350, Phenix References

SuperPhenix, Monju References

EBR-II. FFTF References


General References

- 10CFR50 Appendix A, General Design Criteria for Nuclear Power Plants
- DOE O 5480.30, Nuclear Reactor Safety Design Criteria
Outline

Safety Issues #1 (Last Time)
- Safety issues; DBA, BDBA, severe accident historically (coolant void, recriticality, FCI), passive safety
- Safety analysis past results (FFTF, CRBRP, SAFR, PRISM)
- Safety analysis methods; reactor, structural, coolant aerosols, containment

Safety Issues #2 (Today)
- Licensing issues in FFTF, CRBRP, SAFR, PRISM.
- Domestic and international experience in perspective, high profile events in EBR-1, FERMI, BN-350, Phenix, SuperPhenix, and MONJU explained and lessons learned.
- Safety testing results, EBR-II SHRT, FFTF ULOF (GEMS)
- Inherent passive safety characteristics of sodium fast reactor systems (wrt loss of flow without scram, etc.). Inherent reactivity shutdown, natural circulation decay heat removal
Licensing issues in FFTF, CRBRP, SAFR, PRISM
FFTFA Chronology

- Conceptual design studies 1966-1969
- PSAR submitted by HEDL in September 1970
- Initial construction authorization in September 1971; full construction authorization in March 1972
- ACRS letter in May 1973
- FSAR submitted by HEDL in December 1975
- Construction complete/Na fill 1978
- Criticality February 1980, full power October 1980
- Research operations April 1982 to April 1992
- DOE Shutdown order December 1993
**FFTFT Design**

- **Mission:** Provide a prototypic LMFBR operating environment for testing and development of fuels, materials, and components
  - Secondary: Develop design and construction experience
- **400 MWt, MOX fuel (22% and 27% Pu), three loop primary system, three intermediate sodium loops to air dump heat exchangers**
- **Reactor core:** 73 fuel assemblies, 9 control assemblies, 9 test assemblies
- **Coolant inlet 680°F (360°C), outlet 980°F (527°C)**
- **10psi steel containment**
- **Two independent reactor shutdown systems (both by moveable rods)**
- **Forced and natural convection decay heat removal through three independent loops**
  - Pony motors on primary and secondary pumps
- **Core physics and structural design for inherent negative power and temperature reactivity feedbacks**
FFTF Site – Hanford, Washington
FFT Containment Building View
FFTFT Loop-Type Primary and Secondary Systems
FFTFR Reactor and Vessel Design
FFTFT Regulatory Review -- 1

- As an AEC project, FFTF did not require licensing as for commercial LWR plants, but technical review by NRC was required
  - The depth and detail of the NRC review was similar to full licensing
  - Construction and operation permission; ACRS letters


- Site issues: seismic (0.25g) and tornado (150 mph rotation)
  - Studies and analyses submitted to NRC and ACRS for review

- Hypothetical Core Disruptive Accidents (HCDA) received the greatest regulatory attention and review emphasis
  - Basis for evaluation of containment margins (10CFR100 offsite dose)
  - Project position: HCDA was not a design basis (150 MW-s margin evaluation basis)
  - NRC requested further study; response by HEDL and ANL
A major part of the LMFBR safety base program was oriented to support FFTF regulatory review

At HEDL
- Transient Overpower (TOP) accident analysis (MELT computer code)
- TOP fuel testing (TREAT)

At ANL
- Loss-of-Flow (LOF) accident analysis (SAS3A computer code)
- LOF fuel testing (TREAT)
- Post-accident Heat Removal (PAHR) analyses and experiments
- Structural dynamics analysis and testing
- Fuel Element Failure Propagation (FEFP) studies and experiments
- Coolant dynamics analyses and experiments
- Fuel dynamics analyses and experiments (OPERA)
- High temperature materials properties
- Fuel coolant interactions (FCI) analyses and experiments (OPERA)
LMFBFR safety base program activities also performed at ORNL, AI, GE, and W-ARD
- Activities coordinated under HEDL technical direction

ANL provided direct support to FFTF licensing
- Preparation of technical reports of analyses, experiments, and tests for use as FSAR support documents
- Participation in meetings with NRC staff and ACRS

Regulatory review for construction nominally concluded with the May 1973 ACRS letter, but open issues continued to receive attention
- HCDA energetics
- Design fallbacks, including sealing the reactor head compartment and ex-vessel core melt retention
- Piping integrity; provision for pipe break mediation design, and surveillance and in-service inspection
- Natural convection cooling and emergency power
Through 1976, HEDL and ANL continued to meet with NRC staff and ACRS.

ANL supplied technical support for resolution of the HCDA energetics and core melt retention issues.

- In 1974, NRC concurred with the ANL assessment that HCDA energetics would not exceed FFTF capability. Also, NRC concurred that sealing of the head compartment would not significantly improve containment margins.
- In 1975, NRC recommended that construction could be completed without addition of an ex-vessel core catcher.

The FFTF FSAR was issued in March, 1976, followed by an NRC staff review.

The NRC staff review continued, and the Final Safety Evaluation Report was issued in August 1978. The SER stated that the major unresolved issues were natural convection verification, control room habitability, piping integrity, and containment margins.

Natural convection verification testing was performed during start-up.
A safety-grade system to provide control room isolation upon detection of unacceptable levels of sodium aerosols or radioactivity was added.

The piping integrity and containment margin issues were resolved without design changes by additional analyses and information submittals to NRC.

ACRS concurred with NRC findings in a November 1978 letter.

Coolant filling was accomplished in 1979, and fuel loading began.

First criticality was in February 1980, and power operation began in October 1980.
Clinch River Breeder Reactor Plant (CRBRP) Chronology

- March 1972. Two not-for-profit organizations established. Project Management Corporation (PMC) for project management, and Breeder Reactor Corporation (BRC) for utility industry liaison.
- November 1972. Westinghouse-Advanced Reactor Division (W-ARD) selected as lead reactor manufacturer, Burns and Roe (B&R) as A/E.
- February 1973. Initial work authorization. AI and GE added to team.
- PSAR submitted April 1975 (Updated through Rev. 77 May 1983)
- Licensing suspended: commercial reprocessing banned March 1977
- Licensing resumed September 1981
- Funding stopped October 1983
CRBRP Design

- **Mission:** Demonstrate the safe and reliable operation of an LMFBR in a utility environment. Demonstrate LMFBR economics, and the transition from technology development to commercial operation
- **975 MWt, 380 MWe (gross), MOX fuel (19% and 27% Pu), three loop primary system, three intermediate sodium loops to steam generators**
- **Reactor core:** 198 fuel assemblies (108 inner/90 outer), 19 control assemblies (15 primary/4 secondary), 150 radial blanket assemblies
- **Coolant inlet 730°F (388°C), outlet 995°F (535°C)**
- **10psi steel containment**
- **Two independent reactor shutdown systems (both by moveable rods)**
- **Decay heat removal through three independent loops**
  - Pony motors on primary and secondary pumps
  - Auxiliary decay heat removal through water side of SG
- **Direct decay heat removal system independent of HTS loops**
- **Core physics and structural design for inherent negative power and temperature reactivity feedbacks**
Clinch River Breeder Reactor Plant (CRBRP)
CRBRP Regulatory Review -- 1

- CRBRP was licensed as a commercial power reactor by NRC
  - Project suspended in accord with the Presidential order in 1977
  - Licensing activities continued to obtain the equivalent to a construction permit in 1983
- Site selection in 1972, environmental report early 1975, PSAR April 1975
- Site issues: seismic (0.18g) and tornado (290 mph rotation)
  - Consistent with other TVA sites
- As for FFTF, HCDAs received much regulatory review attention
  - Early agreement (1976) between NRC and the project that HCDAs would not be a design basis for containment
  - However, the role of severe accidents and characterization of their consequences dominated the attention of the interveners, the regulators, and the project
  - Licensing, and treatment of severe accidents, set the critical path for construction
PSAR preparation was the responsibility of W-ARD
- General design criteria
- Preliminary design
- Design basis event (DBE) analyses for PSAR Ch. 15

The LMFBR base program, and particularly ANL provided significant resources to address design and licensing issues
- TREAT fuel testing; basic phenomenological test measurements and prototypic TOP and LOF transient tests
- LOF and TOP accident analyses (SAS3D computer code)
- Coolant and structural dynamics tests and analyses
- Post-accident heat removal analyses

ANL provided direct support to CRBRP licensing
- Preparation of technical reports of analyses, experiments, and tests for use as PSAR support documents
- Participation in meetings with NRC staff and ACRS
CRBRP Regulatory Review -- 3

- HCDA issues compared to FFTF
  - Bigger reactor (975 MWt vs 400 MWt), more fuel
  - Positive coolant void reactivity worth (~3$ vs ~0$)
  - LOF sequence bounded energetics as in FFTF, but because of the positive coolant void worth, cladding failures and fuel melting occurred at higher power than in FFTF (~10 \(P_o\) vs ~1\(P_o\))
  - Higher power LOF caused other phenomena in accident sequences that raised energy releases in analyses

- CRBRP structural limits (vessel head bolt strength) corresponded to an accident energy release of 661 MW-s: project structural margin beyond the design basis (SMBDB)
  - NRC (LANL) performed independent analyses: 1200 MW-s
Ultimately,
- The ASLB ruled against intervenor’s contention that HCDAs should be a design basis
- NRC staff stated: “It is our current position that the probability of core melt and disruptive accidents can and must be reduced to a sufficiently low level to justify their exclusion from the design basis accident spectrum.”
- CRBRP project, with ANL support, built a technical case to justify exclusion
- CRBRP met licensing requirements for construction without inclusion of HCDAs in the design basis
SAFR and PRISM Background and Chronology

- Beginning in the early/mid 1980’s, DOE funded conceptual design studies for modular advanced liquid metal reactor plants
  - At Rockwell International, the Sodium Advanced Fast Reactor (SAFR)
  - At General Electric, the Power Reactor - Inherently Safe Module (PRISM)
- Preliminary Safety Information Documents for SAFR and PRISM were submitted to NRC in November, 1986
- The initial SAFR and PRISM concepts focused on innovative design approaches for economics and safety
  - Design simplification based on passive safety performance
- DOE subsequently (ca 1988?) selected the PRISM concept for continued development in the Advanced Liquid Metal Reactor (ALMR) program
- NRC issued pre-application SERs for SAFR (1991) and PRISM (1994)
- DOE support for PRISM design studies ceased with the cancellation of the ALMR program in 1994
Rockwell International SAFR Design

- Multiple (4) power units per site co-located with a spent fuel processing facility
- 900 MWt reactor, U-Pu-Zr metallic fuel, pool type primary system, two intermediate loops
- 60 year plant life
- Reactor core: 96 driver fuel assemblies, 46 internal and 48 radial blanket assemblies, 6 control assemblies, 3 safety assemblies
- Coolant outlet 950°F (510°C), inlet 675°F (357°C)
- Inherent response for emergency reactor shutdown
  - Inherent reactivity feedbacks in temperature and flow transients
  - Self-Actuated Shutdown System (SASS); thermally-activated magnetic latch release
- Two natural circulation decay heat removal systems
  - Direct heat removal from hot sodium pool (DRACS)
  - Ambient air cooling of guard vessel (RACS)
- Compact containment building design
General Electric PRISM Design

- Multiple power modules (6) co-located with a spent fuel reprocessing facility (Module reactor size grew as the concept evolved, to 840 MWt in 1995. The SER looks at the 471 MWt design)
- 840 MWt, U-Pu-Zr metallic fuel, pool-type primary system, two intermediate loops
- Reactor core (burner): 192 fuel assemblies, 10 control assemblies, 3 safety assemblies
- Coolant outlet 930°F (500°C), inlet 680°F (360°C)
- One safety grade automatic reactor protection system, with a manually operated safety grade ultimate shutdown system (3 safety rods)
  - Inherent accommodation of ATWS transients without core melt, significant reactivity addition, or large radiological release
- Shutdown cooling by turbine bypass with emergency removal systems
  - Air cooling system (ACS) on the steam generator shell
  - Primary sodium auxiliary cooling system (PSACS); reqs. valve action
  - Reactor vessel air cooling system (RVACS)
- Compact containment shell design
SAFR/PRISM Safety Design/Licensing

By the time (mid 1980’s) SAFR and PRISM were being designed, technical understanding of severe accident progression had advanced to a point that reactor designs could be specified to greatly reduce the consequences of accident initiators that led to severe accident conditions (coolant boiling, fuel melting, cladding failure) for FFTF and CRBRP designs

- Beyond design basis, double fault accidents with failure of the automatic scram system; loss of coolant flow, reactivity addition, or loss of normal heat rejection

Full scope, integral testing in EBR-II and FFTF (ca 1986) provided confirmation of design features performance that limited accident consequences to elevated temperatures short of coolant boiling or fuel melting, with margins

- Inherent, passive reactor response provided by designs with negative reactivity feedbacks to reduce power and with natural circulation cooling to remove heat

For SAFR and PRISM, the FFTF and CRBRP severe accident initiators cause elevated coolant temperatures that trigger inherent protection
Safety Design and Licensing Implications

- The double fault accident sequences that led to core disruption in FFTF and CRBRP were considered as challenges to containment margins
  - The same initiators in current sodium fast reactor designs do not produce conditions that approach containment margins
  - Fuel cladding, reactor vessel, and containment building integrity are maintained
- These performance characteristics were reflected in the SAFR and PRISM designs, which included simplifications of shutdown, cooling, and containment features
- To achieve core melt conditions in modern liquid metal reactor designs, it is necessary to assume accident initiators with very low probability
  - For example, “triple” fault initiators or extremely large earthquakes
  - Proper characterization of such initiators requires probabilistic analysis and a risk-based assessment
- Passive safety mechanisms effectively increase containment margins and public safety
Lessons Learned -- 1

- FFTF and CRBRP experiences demonstrate that a liquid sodium-cooled reactor plant can be licensed
  - FFTF underwent NRC review, and CRBRP construction was approved
  - Requires interpretation of 10CFR50 App. A in view of low pressure, chemically active liquid metal coolant, compliance with intent, and possibly additional criteria to cover physical characteristics not considered in the LWR criteria
  - Compliance with defense-in-depth principles in design specifications

- Effective management of the licensing process can be achieved
  - Recognition by both applicant and regulator of each other’s responsibilities
  - Focus on top-level goals and actions necessary to fulfill both regulatory and project requirements
Lessons Learned -- 2

- Innovative SAFR and PRISM safety design features promise increased margins to beyond-design-basis safety limits, high reliability safety systems, and safety design simplification
  - Reliance on inherent mechanisms for power and reactivity control and natural circulation heat removal
- Early recognition of potential issues of significance and establishment of a framework for resolution
  - Example: Passive reactivity feedbacks and natural circulation heat removal. Proof of reliability by test?
  - Example: Severe accident prevention and consequence mitigation. Risk informed assessment?
  - Others
- Begin technical interactions at the earliest possible opportunity
Domestic and International Accidents and Lessons Learned

- EBR-I Power Excursion
- Fermi-1 Fuel Assembly Inlet Blockage
- BN-350 Steam Generator Leak
- Phenix Reactivity Anomaly
- SuperPhenix
- Monju Sodium Leak
Experimental Breeder Reactor-1 (EBR-I)

- Experimental Breeder Reactor-I (EBR-I) was built at the National Reactor Testing Station in Idaho to demonstrate fuel breeding.
- From 1951 to 1963, EBR-I was operated with four core designs to demonstrate breeding and to develop an understanding of liquid metal fast reactor performance:
  - Pu breeding demonstrated by February, 1952
  - Testing platform for reactor physics, fluid dynamics, and power generation
  - 1.4 MWt, generated 200 kW of electricity for NRTS
- NaK cooled, U-235 (94% enriched) metal fuel in stainless steel cladding:
  - 217 pin locations on a triangular 0.494 in. pitch; 0.384 in. fuel OD; 0.448 in. cladding OD; 8.5 in. core height
  - 227°C inlet, 316°C outlet, 20 psig
  - Mk I and II cores used top support, above-core shield plates, and a bottom tube plate to position fuel pins
  - Mk III (and IV) core used hexagonal tubes and wire wraps for fuel pin positioning
  - Mk IV core used Pu fuel (1962)
EBR-I Reactor

(Reactor Shield in Background)  

EBR-I Core Mk I and II
**EBR-I Mk I and II Above-Core Shield Plate**

Fuel element holes 0.460 in. OD (Cladding 0.448 in. OD)
**EBR-I Accident**

- The Mk II core loading was found to have a prompt positive power coefficient, especially for very low power/very low flow conditions.

- In November, 1955, during a test to investigate the prompt positive component of the power coefficient, an unanticipated power excursion resulted in fuel melting.
  - Repeat of a previous test, with additional instrumentation
  - 60 s initial period from 50 W, no coolant flow
  - Excursion during 500 s to < 1 s period, ~10 MW (est.)
  - Scram by control rods failed to terminate the excursion; manual scram by blanket (reflector) drop
  - After the test, radiation alarms upon flow restart

- Subsequent investigations identified fuel rod bowing as the source of the positive reactivity feedback.
  - Inward pin bending in the core due to radial power profile and insufficient radial support to prevent compaction

- Core was replaced and operation continued through 1963
  - Mk III and IV cores used wire wraps and hex tubes for radial support
EBR-I Mk III Core Loading

Plan View at Midplane

Elevation View
The EBR-I core melt accident demonstrated the sensitivity of compact fast reactors to small changes in core fuel density
- Sensitivity magnified in a very small, high enrichment core like EBR-I

Subsequent fuel assembly designs have used wire wraps and hexcans in the U.S. (grids in Europe) to provide control of fuel geometry
- Must be sufficiently ‘loose’ to accommodate thermal expansion and swelling, but ‘tight’ enough to provide support and positioning
- Trend to bottom support of fuel pins and free axial expansion upward within the hexcan

Hexcans are usually supported at the bottom in the core grid plate, and constrained radially in a manner to accommodate hexcan swelling and creep
- Early designs used ‘free bowing’ concept (EBR-II) to give room for withdrawal in refueling; accommodation of steel swelling
- Later designs (FFTF) used a ‘limited free bow’ design to manage hexcan bending to assure a negative power coefficient
Fermi-1

- 200 MWt power station located on the western shore of Lake Erie south of Detroit
- Designed by Atomic Power Development Associates (APDA) and constructed by Power Reactor Development Co. (PRDC) for Detroit Edison
  - Critical August 1963, first power August 1966
  - Sodium cooled, 288°C inlet, 427°C outlet, 120 psia
  - Metal fuel, Zr cladding 0.158 in. OD, 31 in. height, square pin pitch
- Subassembly flow blockage and fuel melting accident during power ascension on October 5, 1966
  - Metal fuel core removed and replaced with oxide core; full power 1969
- Operation ceased in 1972
Fermi-1 Primary System
Fermi-1 Core Inlet Plenum and Melt-Down Section
Fermi-1 Fuel Assembly
Fermi-1 Fuel Melting Accident

- Prior to October 5, high fuel assembly outlet thermocouple readings had been observed during low power operations
  - Assemblies with abnormal temperature readings were relocated to positions under different thermocouples
  - The location(s) of the high temperature readings changed on each start-up, but not in correlation with the assembly movements
- On Oct. 5 during a power ascension at 34 MWt, building radiation alarms sounded, indicating fuel damage
  - The reactor had previously operated at 100 MWt without problems
- Subsequent investigations revealed fuel melting in two adjacent assemblies
  - Another adjacent assembly was bent, with no internal damage
- A ‘foreign object’ was found in the inlet plenum, which later proved to be a crumpled Zr plate from the melt-down section liner
  - The loose Zr plate had apparently been swept by flowing coolant to cover (partially or completely) the inlet nozzle of various assemblies during the multiple start-ups
Fermi-1 Fuel Melting Accident – Lessons Learned

- Assembly inlet nozzle designs since Fermi-1 have included multiple coolant inlet passages so that complete external blockages are ‘impossible’ by design.
- Considerable research and testing of both external and internal blockages have been performed to understand and quantify the damage mechanisms and limits.
- In the U.S., the assembly blockage scenario (external and internal) has been addressed in the assembly design (inlet flow diversity), in the inlet plenum design (coolant flow distribution and assurance of assembly supply), in the instrumentation design (detection by multiple thermocouples, delayed neutron detectors, gas tags), and in fuel handling equipment design (casks).
- Internationally, in some countries the fuel assembly blockage scenario has become a design basis accident.
**BN-350**

- 750 MWt power station located on the eastern shore (~ 2 mi. inland) of the Caspian Sea near Aktau (Shevchenko), Kazakhstan
  - 130 MWe, 150 MWt desalination

- Power level during early operation (1973-1975) limited (350-550 MWt) by leaks in the steam generator evaporator tubes
  - Plant designed for 5 of 6 loop operation (One loop spare)
  - In each loop; two evaporators with bayonet tubes and two superheaters with U-tubes
  - 816 bayonet tubes in each evaporator, 33 mm OD x 3 mm thick (originally 32x2) with a welded end cap and a 16 mm x 1.4 mm internal downcomer tube
  - In start-up tests, found leaks at the tube-sheet welds and the end cap welds
  - In operation, eight evaporator leaks through 1975 involving all but one of the loops (Loop No. 4 never had a leak)
    - *Three major leaks with extensive damage*
BN-350 Plant Layout

Fig. A.3.3. Plan View of BN-350, Reactor Building Only. Symmetrical layout—3 loops to the right of the reactor; 3 to the left of the reactor. 1—Reactor; 2—Refueling cell; 3—Wash cell; 4—Three of 6 IHX compartments, 2 shells in each compartment; 5—Three of 6 steam generator compartments; 6—Secondary dump tanks. For B-B see Fig. A.3.17.
BN-350 Flow Diagram

FLOW DIAGRAM OF THE NUCLEAR DESALTING COMPLEX BN - 350
BN-350 Coolant Loop (1 of 6)

Fig. 1. Schematic diagram of a cooling loop of the energy-production system of BN350.
BN-350 Steam Generator and Evaporator Tube

Fig. A.3.23. BN-350 Steam Generator. 1—Saturated steam inlets, 2—Superheated steam, 3,7—Sodium inlet and outlet, 4—Feedwater inlet, 5—Water level, 6—Sodium level, 8—Burst diaphragm, 9—Evaporator, 10—Superheater.
BN-350 Steam Generator Evaporator Experience

- Through 1974, two major leaks and three smaller leaks
  - Initiated from end cap welds; micro-cracks in end cap weld seam zone, attributed to mechanical deformation at end cap manufacture

- In 1974, decision to re-tube all evaporators except Loop No. 4
  - By February 1975, three of five loops had been re-tubed
  - After 7 days of operation, one of the evaporators in the most recently re-tubed loop failed (No. 5), leading to the most significant leak (Balent report)
    - 120 tubes failed, 800 kg water leak
    - This steam generator was dismantled and replaced with a Czechoslovakian steam generator

- It is claimed that safety systems (rupture disk and blowdown) prevented destruction of the evaporator vessels for the three large leaks (i.e. no sodium leaks)
  - 1975 US delegation visit (Balent) report speculated that the reaction products stayed within the vessel shell, and this aggravated tube failure propagation

- After re-tubing, some leaks still occurred, but emphasis on sodium and feedwater quality control, early leak detection, and remediation (tube plugging) resulted in stable plant operation at design power levels
BN-350 Steam Generator Failures – Lessons Learned

- Soviet steam generator technology experience prompted needs for
  - Improved steam generator component manufacturing techniques; tube drawing/forging and welding
  - Design for failure prevention; sodium and feedwater quality control
  - Design provision for tube failure;
    - Detection; quick recognition and action to prevent propagation
    - Containment; blow-down relief to control intermediate sodium system pressure
    - Remediation; plugging of leaking tubes

- U.S. and Western technology contrast
  - EBR-II evaporator/superheater experience
**Phenix**

- 563 MWt, 250 MWe power station located near Marcoule, France
- Criticality 1973, full power 1974
- 85 cm core height, 430 cm subassembly height
- 217 wire-wrapped pins per subassembly
- Number of subassemblies: 55 inner core, 48 outer core, 90 radial blanket, 1317 reflector and shield
- Free-Standing (Free-Flowering) Core Restraint
  - Accommodation of thermal/mechanical/irradiation effects for refueling
- Four rapid, large, negative reactivity excursions triggered automatic scrams due to power reduction: 6 August ’89, 24 August ’89, 14 September ’89, and 9 September 1990
  - Intensive investigations failed to provide identification of the cause(s)
  - Operations twice interrupted and then resumed following testing, installation of additional instrumentation, and analyses of events
Phenix Site
Phenix Reactor Building
Phenix Pool-Type Primary System
Phenix Reactor Core
Phenix Negative Reactivity Transients, $P$ (Mwt) vs $t$ (ms)

Phenix Reactivity Excursions - 1

- After the first two events (August ’89), the cause was attributed to ‘interference’ in operation of the plant control system instrumentation, which had been modified just prior to 1989, but no specific fault was found.

- After the third event (September ’89), the cause was attributed to passage of a gas bubble through the core periphery (negative reactivity) due to plugging of inlet plenum vents designed to prevent gas build-up. After a shutdown, maintenance, and analyses, the reactor was restarted in December ’89.

- Two cycles later, the fourth event (September ’90) invalidated the gas bubble hypothesis:
  - Reactor operation ceased and an intensive investigation was begun.
  - A panel of experts was convened to consider all possible causes.
  - Reactor and plant tests and repairs were performed.

- By the end of ’91, the root cause had not been identified, but the ‘strong presumption’ was (rapid) radial expansion of the subassemblies.
Phenix Reactivity Excursions - 2

- Through 1992, analyses and testing with installation of special reactor surveillance equipment continued
  - Focus on causal phenomena: coolant voiding, control rods motions, and core movements
- Given the amplitude and speed of the events, only core movements could cause the observed behavior
- Modeling and simulations were performed, but no one specific scenario or cause was identified
  - Final explanation: Outward (radial) expansion of the subassembly lattice, followed by a return
- Ultimately, consensus that operations could be safety resumed
  - No further events to date
- Lesson learned? My opinion: Avoid free flowering core restraint design.
Super Phenix (Creys-Malville)

- 2990 MWt, 1242 MWe power station located 50 KM east of Lyon, France
- Criticality 1985, full power 1986
- 1 m core height, 5.4 m subassembly height
- 271 wire-wrapped pins per subassembly
- Number of subassemblies: 193 inner core, 171 outer core, 234 radial blanket, 1288 reflector and shield

On March 8, 1987, a leak in the fuel storage tank was detected
  - Leaking sodium was contained by the storage tank guard vessel
  - Stored fuel (new fuel, one partly irradiated subassembly, and dummy subassemblies used for pre-startup testing) were unloaded and the tank was drained.
    - Investigations began to determine the location of the leak (Sept. ’87)

It was ultimately concluded that the tank could not be repaired, and alternative fuel handling equipment and procedures were adopted

The expense associated with this and other events led to closure of the plant in 1998 (Down 2 yrs for technical, 4 ½ yrs for ‘administrative’)
Super Phenix Plant Site
Super Phenix Plant Coolant Flow Diagram

1 - élément combustible fissile
2 - élément combustible fertile
3 - barre de contrôle
4 - pompe de circulation du sodium
6 - cuve du réacteur (acier inoxydable)
7 - cuve de sécurité
8 - enceinte de confinement
9 - couvercle
9a - Atmosphère de gaz de protection (Argon)
10 - échangeur de chaleur intermédiaire (1 parmi 4)
11 - circuit de sodium secondaire
12 - pompe de circulation du sodium secondaire
13 - Générateur de vapeur (1 parmi 4)
14 - vapeur
15 - pré-réchauffeur
16 - pompe à eau d’alimentation
17 - condenseur
18 - eau de refroidissement (fleuve)
19 - pompe à eau froide
20 - turbine haute pression
21 - turbine basse pression
22 - génératrice
23 - bâtiment réacteur
Super Phenix Plant Plan View
Super Phenix Plant Elevation View
Fuel Storage Tank Detail
April 26th, 1980: Storage drum being placed in the reactor building.

This photo has been taken from the sloping handling ramp. The criss-cross girdering of the vessel roofs can be seen and the system for connecting this roof to the hoop anchored in the main structure: 42 slings fixed to the roof flanges and the hoop supports.
Inside the Fuel Storage Tank During Fabrication

Creys-Malville: Inside the lower part of the storage drum.
Penetrating the diagrid bottom plate are the lower parts of the thimbles into which the fuel elements will be placed. Note the holes for the circulation of sodium.

Creys-Malville: Storage drum during assembly viewed from the inside.
Left: the axis of the diagrid carousel. Its upper plate is covered with hardboard sheets to facilitate movement during assembly. Right: note the cooling system tubes (sodium-sodium exchange) behind their protective grids.
**Fuel Storage Tank Leak Location**

**FIG. 2.21.** Super-phenix fuel storage drum leak localization.
Super Phenix Fuel Storage Tank Leak

- Leak caused by a horizontal crack ~ 60 cm long on the lower welding bead of a cooling system support plate
  - Investigation showed cracks at other locations as well; other support plates and tank wall weld beads
- The “most probable scenario” causing the crack was identified as “the nature of the drum steel (ferritic 15 D3) and the simultaneous presence of three factors: the existence of start (sic) sites (micro-cracking) in zones of high hardness, residual stresses close to the elastic limit of the material, and lastly, the contributions of hydrogen which allowed the brittling phenomena to occur.”
  - It is customary to use austenitic stainless steel in contact with liquid sodium at high temperatures (reactor vessels and pipes)
- Lesson learned: Select the correct material for high temperature sodium service
**MONJU**

- 714 MWt, 280 MWe power station located 12 km NE of Tsuruga, Japan
- Criticality 1994, full power 1995
- 93 cm core height, 420 cm subassembly height
- 169 wire-wrapped pins per subassembly
- Number of subassemblies: 108 inner core, 90 outer core, 172 radial blanket, 324 reflector and shield
- On December 8, 1995; sodium leak detected in the room housing the “C” secondary loop (non-radioactive sodium) during operation at 43% power
  - High temperature at the IHX outlet and smoke alarm at 19:47
  - Reactor shutdown begun at 20:00, manual trip at 21:20
  - C loop draining at 22:55, complete at 00:15
  - Inspection confirmed sodium leak at thermocouple well; ~1 m\(^3\) mound of Na\(_2\)O on the steel floor, aerosol on the walls and floor
  - Leak caused by failure of a thermocouple well
- Clean-up and repairs were made, but restart was delayed by legal actions, court decisions, appeals, safety reviews, etc.
  - Restart scheduled for 2008
MONJU Site
MONJU Flow Diagram

Primary cooling system (Sodium)

Secondary cooling system (Sodium)

Water & steam system

Outer shielding wall

Reactor containment vessel

Control rod drive mechanism

IHX

Primary pump

Isolation valve

Superheater

Air cooler

Turbine

Generator

Condenser

Sea water

Cooling water pump

Leak location

Secondary pump

Evaporator

Isolation valve

Argonne National Laboratory
MONJU Reactor Vessel and Core
MONJU Coolant Systems

**Primary systems**
- R/V sodium temperature
  - Inlet: 397 °C
  - Outlet: 529 °C
- Sodium mass: 760 ton

**Secondary systems**
- IHX sodium temperature
  - Inlet: 325 °C
  - Outlet: 505 °C
- Sodium mass: 760 ton

Outer shield (1.6m)

Concrete Separation wall (1m)

Diagram of the coolant systems showing various loops and components such as IHX, PCV, PMCP, and CV.
MONJU Secondary Sodium Leak Location on C Loop

Fig. 2. Sketch of the affected area.
**Original Secondary Circuit Thermocouple**

![Diagram of the thermocouple well of the secondary circuit]

*Fig. 3. The thermocouple well of the secondary circuit.*
Damaged Secondary Circuit Thermocouple

Fig. 4. The sodium leak flow path.
MONJU Thermocouple Sodium Leak

  - Video (6 min., in Japanese) shows post-leak consequences, clean up activities, repairs and remediation measures
- Thermocouple well tip failed due to flow-induced cycle fatigue
- All thermocouples in the secondary loop were replaced
Thermocouples Replaced on Secondary Circuit

![Diagram of Thermocouples Replaced on Secondary Circuit](image)

Fig. 7 Modified design concepts for the thermocouple wells on the secondary cooling system.
Countermeasures against Sodium Leakage

- Replacement of Secondary Thermocouple wells
  - Leakage Control by Metallic Gasket Type Seal and Welded Seal
  - Leakage Detection
  - Prevention of Flow-induced Vibration

- Improvement of Facilities
  - Improvement of Drain System, Reduction of Drain Time
  - Integral Sodium Leakage Monitoring System
  - Subdivision of Building / Nitrogen Gas Extinguisher System
Improvement of Drain System, Reduction of Drain Time

Draining Time: \( \approx 50 \text{ minutes} \rightarrow \rightarrow \rightarrow \rightarrow \rightarrow \leq 25 \text{ minutes} \)
Integrated Sodium Leakage Monitoring System
Installation of Nitrogen Gas Injection System
MONJU Sodium Leak and Lessons Learned

- Event had no radiological consequences, no injuries, no environmental harm
  - Post-event information management, legal action, license review
- Technical lessons
  - Faulty thermocouple well design; inadequate prototype testing, lack of backup leak prevention
  - Some experts question need for pipe penetrations
Safety Testing Results

- EBR-II Shutdown Heat Removal Tests
EBR-II Shutdown Heat Removal Test (SHRT) Program

- Thermal-hydraulic testing at EBR-II began to support safe and reliable operation of EBR-II, but evolved to become a broader program to support design and performance assessment for advanced liquid metal reactors.

- Early testing (1974) focused on steady-state fission and decay heat removal by natural circulation, and pioneered the use of specially instrumented fuel subassembly (XX07) for flow and temperature measurements.

- Subsequent tests examined the transition from forced to natural circulation with updated instrumentation (XX08), from a variety of initial conditions:
  - Ex: Primary and secondary pump trips with scram from hot standby
  - Smooth, benign transitions to natural circulation.

- The SHRT program employed new instrumentation (XX09) and extended the test matrix to transients from full power, with and without scram:
**EBR-II Design Overview**

- 62.5 MWt, 20 MWe power station located at Argonne National Laboratory-West, Idaho
- Initial mission: Demonstrate fuel breeding and closed cycle operation with reprocessing of metallic fuel
  - Following mission fulfillment, shift to irradiation testing of advanced fuels
- Dry critical 1961, wet critical 1963, full power 1964, shutdown 1994
- Sodium cooled, 371°C inlet, 473°C outlet, 47 psig
- Fuel pins 0.17 in. OD, 13.5 in. core height; metal fuel in SS cladding
- First fuel processed in Fuel Cycle Facility in September 1964; recycled fuel irradiation in April 1965
- Mission oriented to irradiation testing in 1969; supporting FFTF and CRBRP oxide fuel testing and development
- Integral Fast Reactor (IFR) program began in mid 1980’s
  - Testing and demonstration of high burnup metallic fuels
  - Shutdown Heat Removal Test series 1984-86; natural circulation decay heat removal and passive shutdown in ATWS events (unprotected loss-of-flow and loss-of-heat-sink)
EBR-II Site at Argonne-West
EBR-II Site at Argonne-West
EBR-II Coolant and Power Conversion Systems
EBR-II Pool-Type Primary System
EBR-II Reactor Tank
EBR-II Reactor Vessel Assembly
EBR-II Reactor Core Layout

- A series of tests, originally intended to qualify EBR-II for continued operation, that evolved into an experimental program supporting the safety and design of advanced liquid metal reactors
- Testing ranged from demonstration of natural circulation decay heat removal to whole-plant simulation of unprotected (without scram) loss-of-flow (ULOF) and loss-of-heat-sink (ULOHS) accident from full power and flow
- Test results provided data for validation of computer codes used in design and safety analysis of advanced LMRs
- Instrumented subassembly XX09, equipped with calibrated thermocouples and flow meters, provided real-time measurements of coolant temperatures and flow rates during the tests
- The ULOF and ULOHS tests demonstrated the ability of a pool-type, metal-fueled LMR to provide self-protection in beyond-design-basis accidents
XX09 Instrumented Subassembly and Subchannel Model

XX09 Thermocouples

SASSYS XX09 Model
EBR-II Primary Heat Transport System SASSYS Model
EBR-II Intermediate Heat Transport System SASSYS Model
EBR-II SHRT-17 Protected Loss-of-Flow XX09 Results
SASSYS Calculation - 2006

XX09 Transient Coolant Flow

31 TTC Transient Temperatures

Measurement, 31TTC Calculation

Lower Flow Meter
Upper Flow Meter
Calculation
EBR-II SHRT-17 Protected Loss-of-Flow XX09 Temperatures
SASSYS Calculation - 2006

Steady State t = 0 s

Transient State t = 85 s
EBR-II SHRT 39 Unprotected Loss of Flow Sequence Results – 1
NATDEMO/HOTCHAN Calculation - 1986

Pump coastdown without scram... causes transient temperature rise...
EBR-II SHRT 39 Unprotected Loss of Flow Sequence Results – 2
NATDEMO/HOTCHAN Calculation - 1986

introducing negative reactivity... reducing reactor power to decay heat.
EBR-II Unprotected Loss of Heat Sink Sequence Results – 1
NATDEMO/HOTCHAN Calculation - 1986

Secondary pump trip without scram... causes transient inlet temperature rise...
introducing negative reactivity... reducing reactor power to decay heat.
Significance of EBR-II SHRT Tests

- The EBR-II SHRT tests demonstrate that passive safety mechanisms in sodium-cooled, metal-fueled, pool-type reactors can limit the consequences of double-fault accidents
  - No coolant boiling, cladding failure, or fuel melting
  - Reliance on inherent performance characteristics: negative reactivity feedback to reduce reactor fission power, and natural circulation decay heat removal

- Larger reactors can be designed to have these performance characteristics
  - High thermal conductivity of metallic fuel (low fuel operating temperature) reduces positive Doppler reactivity feedback upon power reduction
  - Pool-type design provides thermal inertia to buffer (slow down) impacts of accident initiators
  - Arrangement and vertical separation of heat source and sink elevations promotes natural circulation
  - Core restraint and support design to provide negative reactivity for increasing coolant temperature (as in FFTF and CRBRP)

- A series of test intended to simulate passive safety performance of an advanced LMR
- Unprotected (without scram) Loss-of-Flow from reduced power (range 10% to 50%) and full flow
- Nine Gas Expansion Modules (GEMs) were installed to provide negative reactivity during the coolant flow coastdown
  - Empty hexcans, sealed at the top, and installed at the core periphery. As the inlet pressure decreases, the coolant level falls, introducing void and increasing neutron leakage
- ULOF tests preceded by flow transients for reactivity feedbacks characterization
  - Pump trips with scram to natural circulation
  - Static measurements of GEM reactivity worth
- Reactor coolant flow and temperature measurements were obtained with the normal plant instrumentation (pump speed, coolant loop flowmeters, subassembly outlet thermocouples)
- Two fast thermocouples mounted on subassembly outlets in rows 2 and 6; (Post Irradiation Open Test Assembly – PIOTA)
**Why GEMs? Reactivity Swing for Power Reduction**

- Oxide Fuel
  - (Doppler Coeff. = - 0.005)
  - 892°C BP
  - ~1.5 $ (360°C Inlet)

- Metallic Fuel
  - (Doppler Coeff. = - 0.003)
  - 510°C Outlet
  - ~0.3 $ (360°C Inlet)
GEM Coolant Level for Power and Flow Conditions

CALCULATED GEM SODIUM LEVEL vs. PLANT CONDITIONS

- - - 210°C, 100% FLOW
- - - 316°C, 100% FLOW
- - - FULL POWER AND FLOW

- - - 210°C, 10% FLOW (REFUELING)

- - - 316°C, 10% FLOW

- - - 10% FLOW, FULL POWER TEMPERATURE

SODIUM LEVEL
FFT Core Loading for ULOF Testing
FFTF PIOTA Configuration

FIGURE 9
ABOVE CORE THERMOCOUPLE INSTALLATIONS
Lessons Learned from EBR-II and FFTF Testing

- The experience of planning and executing special tests beyond the original mission of the reactor and plant can provide new insights into the capabilities of the facility
  - Assurance of safety performance, confirmation of margins
- Results of testing can contribute new knowledge that guides performance expectations and selection of design features for future reactors
  - Passive safety performance based on natural circulation shutdown heat removal and inherent reactivity feedbacks
- To maximize test value, special equipment may be necessary
  - Instrumentation to measure performance directly
  - Flow coastdown (pump power supply)
  - Negative reactivity feedback enhancement
Inherent passive safety

- Inherent passive safety characteristics of sodium fast reactor systems (wrt loss of flow without scram, etc.).
  - Inherent reactivity shutdown
  - Natural circulation decay heat removal
- These performance characteristics can be achieved in reactors sized and configured for commercial deployment
- The following examples are provided for a large, low conversion ratio conceptual design to demonstrate passive safety performance for protected and unprotected loss-of-flow accident sequences
SFR Safety Analysis

- Safety analyses were carried out for a 1000 MWt pool-type concept to provide a quantitative safety assessment of reactor and plant performance.
  - Evaluations were made for both the metal and oxide core designs.

- Scope of analyses focuses on the ability of the SFR to provide inherent protection against damaging consequences following low-probability accident sequences involving multiple equipment failures.

- Two accident sequences were evaluated:
  - Protected Loss of Flow (PLOF)
  - Unprotected Loss of Flow (ULOF)

- Analyses were performed with the fast reactor safety analysis code SAS4A/SASSYS-1.
Safety Analysis Approach

- Protected Loss of Flow (PLOF)
  - Initial conditions assume full power operation at BOEC.
  - Total loss of normal power to the reactor cooling system at $t = 0$, with complete failure of the emergency power supply system.
  - Balance of plant is assumed to cease operation and provide no heat rejection capability.
  - Immediate reactor scram following power failure.

- Unprotected Loss of Flow (ULOF)
  - Initial and accident conditions are identical to the PLOF case.
  - Additionally, the reactor protection system fails to scram either the primary or secondary control rods.
  - Power control is exclusively through reactivity feedback mechanisms.

In both cases, the only heat removal path is through the emergency heat removal system (DRACS) by natural circulation.

- PLOF simulations were carried out to 40,000 seconds (~11 hours)
- ULOF simulations were carried out to 4,000 seconds (~1 hour)
Analysis Summary

- Both metal and oxide core designs demonstrate significant safety margins to coolant boiling and fuel damage in PLOF accident sequences.
- The metal core design also exhibits significant safety margins in the ULOF accident sequence. This is a direct consequence of
  - High thermal conductivity and low operating temperature of metal fuel.
  - Favorable negative reactivity feedback due to thermal expansion

However...

- ULOF analyses for the oxide core design indicate that margins to coolant boiling may not be adequate
  - Inadequate margins despite significantly longer flow halving time assumed for oxide (20 seconds) compared to metal (5 seconds).
  - Initially high fuel temperatures result in significant positive Doppler feedback when trying to reduce temperatures.
- Additional enhancements, such as a self-actuating shutdown system (SASS) device, may be required for the oxide core to increase ULOF safety margins.
End of Presentation

- Questions?
Outline of Presentation

- SFR Fuels Experience in the US
  - Fuel Types
  - Fuel Performance Issues
  - Experience/Testing
- Fuel Test Plan (1994) for PRISM Prototype
- Fuel Specification
- Experience with Fuels Containing Minor Actinides (AFCI/GNEP)
- Approach to Fuel Development and Qualification
SFR Fuels Experience in the US
SFR Fuels Experience in the US

- **Metallic Fuels**
  - EBR-I, Fermi-1, EBR-II, FFTF
  - U-Fs, U-Mo, U-Zr, U-Pu-Fs, U-Pu-Zr, others

- **Mixed Oxide Fuels (MOX)**
  - EBR-II, FFTF
  - \((U,Pu_{0.2-0.3})O_2\)

- **Mixed Carbide Fuels (MC)**
  - EBR-II, FFTF
  - \((U,Pu)C\) w/15% \((U,Pu)C_3\)
  - No current interest
Metallic Fuel Design (EBR-II)

Features of a Metallic Fuel Pin (from Pahl, et al, 1990)
Metallic Fuel Design (FFTF)

FFTF Series III.b Metallic Driver Fuel Design (from Pitner and Baker, 1993)
Important Metallic Fuel Performance Phenomena

- Irradiation growth
- Fuel swelling and fuel-cladding mechanical interaction (FCMI)
- Gas release
- Fuel constituent redistribution
- Fuel-cladding chemical interaction (FCCI)
Metallic Fuel Behavior—Axial Growth

Axial Fuel Growth, from Pahl et al, 1990
Metallic Fuel Behavior—Swelling & Restructuring

As fabricated U-20Pu-10Zr

- REDISTRIBUTION OF U AND Zr OCCURS EARLY
- INHOMOGENEITY DOESN’T AFFECT FUEL LIFE

Zr-rich phases

X423A at 0.9% BU

X419 at 3% BU

X420B at 17% BU

50 µm

November 27-28, 2007

DOE/NRC Seminar Series on Sodium Fast Reactors
Fuel Performance and Qualification
Metallic Fuel Behavior—Swelling & Gas Release

- **Swelling**
  - Low smear density fuels
  - Rapid swelling to 33 vol% at ~2 at.% burnup

- **Gas Release**
  - Inter-linkage of porosity at 33 vol% swelling results in large gas release fraction
  - Decreases driving force for continued swelling

U-19Pu-10Zr (γ-phase) at 2 at.% burnup
Metallic Fuel Behavior—Fuel Constituent Redistribution

U-Pu-Zr

U-Pu-Am-Np-Zr

Lower Melting Phase
Fuel-Cladding Inter-diffusion
- RE fission products (La, Ce, Pr, Nd) and some Pu reacts with SS cladding
- Interaction product brittle
- Considered as cladding wastage

U-19Pu-10Zr with D9; 12 at.% burnup (from Pahl, et al, 1990)
MOX Fuel Design (FFTF)

FFTF He-bonded MOX Fuel: a) Driver Fuel and b) Core Demonstration Experiment Fuel
(from Bridges et al, 1993)
Important MOX Fuel Performance Phenomena

- Fuel swelling and FCMI
- Fuel restructuring
- Gas release
- FCCI
- Fuel-coolant compatibility

November 27-28, 2007
DOE/NRC Seminar Series on Sodium Fast Reactors
Fuel Performance and Qualification
Diameter and cesium fission product accumulation in high-temperature MOX pins, HT9-clad (a) and D9-clad (b). Cs interacted with MOX fuel causing FCMI. (from Bridges, et al., 1993)
MOX fuel ceramography of FFTF driver fuel produced by Kerr-McGee and Babcock and Wilcox, showing restructuring as a function of burnup. (from Hales, et al, 1986)
MOX fuel operated at high temperature and undergoing restructuring exhibits high gas release.

(from Lambert, et al, 1994)
Hypostoichiometric MOX for SFRs

- As-fabricated O/M < 2.00 to suppress free oxygen at high burnup, mitigate FCCI
- O/M ratio affects fabrication
- O/M ratio affects properties

Sample 1 – MOX + MAs
Sample 2 – MOX+MAs+REs
Sample 3 – MOX+MAs+REs+NM
(from Morimoto, et al, 2005)

Melting T vs O/M
(from Morimoto, et al, 2005)
Run-beyond-cladding-breach (RBCB) of MOX accompanied by fuel/Na reaction and initial crack extension

Fuel loss can be related to degree of interaction.

Reactant layer becomes coherent and inhibits further reaction with coolant.

Stainless-Steel Cladding & Duct Performance

- **Performance Issues**
  - Cladding dilation
  - Duct dilation, bowing, or twisting

- **Irradiation Behavior**
  - Void swelling (AS)
  - Irradiation creep (AS & FMS)
  - Irradiation embrittlement (AS & FMS)

- **Alloys to Address Issues**
  - Advanced austenitic stainless steels
  - Ferritic & tempered-martensitic stainless steels
  - Oxide-dispersion strengthened steel alloys
Duct Performance—Bowing & Dilation

Effects on fuel handling forces and the Power Reactivity Decrement (PRD) to get to 62.5 MWt versus EBR-II run number. Anomalies were caused by assembly hex-duct bowing and dilation. (from Shields, 1981)
Duct Performance—Length Change

R.D. Legget, L.C. Walters / Status LMR fuel development in the USA

Fig. 4. FFTF subassembly length change with various duct materials.
Void swelling of commercial steels irradiated in EBR-II at ~420°C. (from Gelles, 2004)
Duct Performance—Void-induced Embrittlement

Severe void-induced embrittlement in X18H10T (304ss-like) hexagonal assemblies from core and reflector regions of BOR-60. (from Garner, 2000)
Ferritic/Martensitic Stainless Steel Development

Development of ferritic/martensitic alloys for high-temperature applications. (from Allen, 2004)
Oxide Dispersion Strengthened (ODS) Ferritic/Martensitic Alloys

- Ferritic/martensitic alloy use limited by high temperature stress rupture properties.
- Further strengthening achieved by addition of very small oxide particles (usually Y$_2$O$_3$).

European ODS ferritic/martensitic alloy fabrication.
(from Lindau et al, 2005)
Oxide Dispersion Strengthened Ferritic/Martensitic Alloys

Nanocomposited Ferritic (NCF) Steel

(MA Fe-12Cr-3W-0.4Ti + 0.25Y₂O₃ : 12YWT) Miller, Hoelzer, Zinkle ORNL

3-D Atom Probe Tomography

• very high number density of ~10²⁴/m of uniformly distributed nanoclusters
  - site occupancy on bcc Fe lattice
  - average size r₉ = 2.0 (+/- 0.8) nm
• elongated grains containing high dislocation number density (TEM)
Yield (a) and tensile (b) strength of ODS alloys compared to a non-ODS Ferritic/Martensitic steel. (from Klueh, et al 2005)
Stress rupture properties of ODS alloys compared to a non-ODS Ferritic/Martensitic steel. (from Klueh, et al 2005)
Limitations to Use of Ferritic/Martensitic Steels

- Ferritic/martensitic stainless steels (e.g., HT9, Mod 9Cr-1Mo, HCM12, etc.) are expected to be applicable to 200 dpa peak dose (~140 MWd/kg burnup), 650°C peak cladding temperature.

- ODS ferritic-martensitic stainless steels (FMS S/A duct) is expected applicable to 250 dpa peak dose (~ 180 MWd/kg burnup) and 700°C peak cladding temperature.

- Major issues are accumulation of high burnup demonstration data and establishment of mass-production technology.
<table>
<thead>
<tr>
<th>Driver Fuel Operation</th>
<th>Metallic</th>
<th>Mixed Oxide</th>
<th>Mixed Carbide</th>
</tr>
</thead>
<tbody>
<tr>
<td>≥ 120,000 U-Fs rods in 304LSS/316SS 1-8 at.% bu</td>
<td>&gt;48,000 MOX rods in 316SS (Series I&amp;II) 8 at.% bu;</td>
<td>None applicable</td>
<td></td>
</tr>
<tr>
<td>~13,000 U-Zr rods in 316SS 10 at.% bu</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Through Qualification</td>
<td>U-Zr in 316SS, D9, HT9 ≥ 10 at.% bu in EBR-II &amp; FFTF</td>
<td>MOX in HT9 to 15-20 at.% bu (CDE) MOX in 316SS to 10 at.% bu</td>
<td>None applicable</td>
</tr>
<tr>
<td>Burnup Capability &amp; Experiments</td>
<td>600 U-Pu-Zr rods; D9 &amp; HT9 to &gt; 10 - 19 at.% in EBR-II &amp; FFTF</td>
<td>4300 MOX rods in 316SS to 10 at.%; fab var’s; CL melt 3000 MOX rods in EBR-II; peak at 17.5 at.% bu 2377 MOX rods in D9 to 10-12 at.% bu; some at 19 at.% bu</td>
<td>18 EBR-II tests with 472 rods in 316SS cladding; 10 rods up to 20 at.% w/o breach 5 of which experienced 15% TOP at 12 at.% 219 rods in FFTF, incl 91 in D9, 91 with pellet &amp; sphere-pac fuel</td>
</tr>
<tr>
<td>Safety &amp; Operability</td>
<td>6 RBCB tests U-Fs &amp; U-Pu-Zr/U-Zr(5) 6 TREAT tests U-Fs in 316SS (9 rods) &amp; U-Zr/U-Pu-Zr in D9/HT9 (6 rods)</td>
<td>18 RBCB tests; 30 breached rods 4 slow ramp tests 9 TREAT tests MOX in 316SS (14 rods) &amp; HT9 (5 rods)</td>
<td>10 TREAT tests (10 rods; Na or He bond); ≤ 3-6 times TOP margins to breach Loss-of-Na bond test; RBCB for 100 EFPD; Centerline melting test</td>
</tr>
</tbody>
</table>
Transients Phenomena—Metallic Fuels

- Pre-failure Behavior
  - Axial expansion
  - Cladding strain due to gas pressure
  - Possible fuel-cladding liquefaction

- Failure Behavior
  - Failure generally near top of fuel column
  - Stress rupture due to gas pressure in cladding thinned by eutectic-like penetration and weakened at high temperature

- Post-failure Behavior
  - Fuel injection into coolant
  - Low stored energy, no reaction with coolant, local sodium voiding

- Overpower capability 4.0 to 4.4 times nominal power
Transient Phenomena—Metallic Fuels
Fuel/Cladding ‘Eutectic’ Formation

U-10Zr / HT9 at 800°C, 1 hr
(from H. Tsai, et al, 1990)

1 - Unaffected cladding
2 - Fuel/cladding solid-state interaction
3 - Fuel/cladding liquid phase
Transient Phenomena—MOX Fuels

- **Pre-failure Behavior**
  - Axial relocation (apparently, upward axial motion)
  - Cladding strain due to FCMI and gas pressure

- **Failure Behavior**
  - Failure generally in upper 1/3 of fuel column
  - Cladding melt-through with gas pressure and FCMI, cladding weakened at high temperature

- **Post-failure Behavior**
  - Fuel dispersal into coolant
  - Relatively high stored energy, reaction with coolant, local sodium voiding

- **Overpower capability 3.0 to 4.5 times nominal power**
Metallic and MOX Fuels Well Developed

- **Metallic Fuels (U-Zr or U-Pu-Zr)**
  - Acceptable performance and reliability demonstrated up to 10 at.% burnup, with capability demonstrated to 20 at.% burnup
  - Robust overpower capability demonstrated in TREAT tests: ~ 4 to 5x’s nominal power; failures near top of fuel column; pre-failure axial expansion
  - Performance issues typically creep rupture at high burnup, accelerated due to FCMI.
  - Performance phenomena with U-Fs, U-Zr & U-Pu-Zr are the same. Burnup, temperature and cladding performance are key variables

- **MOX Fuels**
  - Acceptable performance and reliability demonstrated up to 10 at.% burnup, with capability demonstrated to 20 at.% burnup
  - Robust overpower capability demonstrated in TREAT tests: ~ 3 to 4x’s nominal power; well above primary and secondary FFTF trips; failures near core mid-plane; pre-failure axial fuel motion
  - Performance issues typically creep rupture at high burnup, accelerated due to FCMI (and FCCI if O/M not controlled).
## Technical Readiness Levels for SFR Fuels

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### Transmutation Fuel
- **TRU-metal, TRU-oxide** (roughly same TRL)
- Metal experience: mostly U.S.
- Oxide experience: mostly International (French and Japan)

### Driver Fuel/Analog Transmutation Fuel
- **Metal (U-Pu-Zr)**
  - Not formally qualified
  - Not used in industrial-scale

### Driver Fuel Analog Transmutation Fuel
- **Metal (U-Zr)**
  - Oxide (U,Pu)
  - Qualified for reactor operations
  - Successful mission operations
  - Operational database wider for MOX, especially considering International experience
Metallic and MOX Fuels—Summary

- Metallic and MOX fuel performance in SFRs are both well known, with good experience in the US
- MOX fuel is particularly well developed in France and Japan
- GNEP application of these fuel forms to an actinide transmutation mission will bring significant new challenges
Fuel Test Plan for PRISM Prototype
Created by Argonne National Laboratory, GE and Westinghouse for metallic fuels:

- “In response to the Energy Policy Act of 1992... complete R&D activities by the end of 1996 for U-Pu-Zr driver fuel and U-Zr blanket fuel in order to initiate Title II design activities for the Prototype ALMR, and, by the end of 1998, to extend the database to include U-Pu-Zr fuel which contains minor actinides (Am, Np) in quantities representative of light water reactor spent-fuel recycle.”

The Fuel Test Plan (June 1994) outlined the remaining issues and schedules for achieving these goals. It also responded to all of the questions concerning fuel performance raised by the NRC in:

Fuel Test Plan (1994)—Issues Identified for U-Pu-Zr Fuels

### Steady-State Performance

- **Fuel Swelling**—well characterized; no expectation that MAs and higher concentrations of REs would have an effect

- **Constituent Redistribution, FCMI, and FCCI**—effects of MAs not well characterized; effect of higher RE concentrations needs to be assessed *(also true for MOX)*

- **Fuel Pin Length** (i.e., evaluation of experimental database to longer pin designs)—well characterized; no expectation that MAs and REs would have an effect

- **Fission Gas Release**—well characterized; no expectation that MAs and REs would have an effect
Fuel Test Plan (1994)—Issues Identified for U-Pu-Zr Fuels

- **Transient Performance**
  - **Fuel/Cladding Compatibility**—effect of MAs not well characterized; effects of REs needs to be assessed. (MOX—O/M modifications may be needed)
  - FCMI—well characterized; no expectation that MAs and REs would have an effect
  - Fuel Melting—unusual compositions may need characterization. (MOX—O/M modifications may be needed)

- **Post Failure Performance (Run-Beyond-Cladding-Breach)**
  - Fuel Loss, Fuel/Coolant Interaction—well characterized; no expectation that MAs and REs would have an effect. (MOX—confirmatory testing may be required depending on O/M modifications)
Fuel Test Plan (1994)—General Issues Identified

■ Fuel Reliability
  – Statistical evaluation of fuel lifetime—fuel element reliability for recent designs has been controlled by cladding and duct properties (e.g., creep rupture for cladding, swelling for ducts). Expected lack of influence from MAs and REs on fuel reliability needs to be demonstrated in LTAs.

■ Fuel Modeling
  – Semi-empirical models (LIFE4, SIEX, FRAPCON?) seem adequate to predict MOX lifetime.
  – Incorporation of ‘first principles’ or ‘atomistic’ models may be used to short-cut design to demonstration scope.
  – LIFE-METAL needs more mechanistic models.
Fuel Test Plan (1994)—General Issues Identified

- **Fabrication (i.e., do modifications in fabrication methods affect fuel performance?)**
  - **Metallic Fuel**: The simplicity of metallic fuel fabrication well suited for transfer to a remote setting. Waste from molds and crucibles, potential losses of Am (due to high vapor pressure) needs to be reduced/eliminated by modification of casting process; replace injection casting with bottom pour (no vacuum, reusable molds, etc.).
  - **MOX Fuel**: Proven fabrication methods in glovebox setting. Hot cell/remote setting may create challenges. O/M control with the inclusion of MAs and REs unproven.
Fuel Test Plan (1994)—General Issues Identified

- Cladding and Duct Performance
  - Ferritic/martensitic steels expected applicable to 200 dpa dose (~140 MWd/kg fuel burnup), 650°C peak temperature; should be applicable for ducts to higher exposures.
  - ODS ferritic stainless steel cladding expected applicable to 250 dpa dose (~180 MWd/kg fuel burnup) and 700°C peak temperature.
  - Major issues
    - Accumulation of high burnup/dose demonstration data
    - Establish industrial production base
Fuel Test Plan (1994)—Conclusions

- Most fuel performance issues are well understood and adequately tested
  - U-Pu-Zr metallic fuel
  - (U,Pu)O₂

- Variations in fuel compositions to include MAs and possible carry-over of RE fission products will require additional testing

- Fuel modeling and code development needs additional effort, especially for metallic fuels
Fuel Specification
EBR-II experience showed that early specifications contained too much ‘process control’ information

- Chemistry over-specified (i.e., concern for potential minor contaminants that would not affect fuel performance)
- Dimensional control of fuel slug over-specified (i.e., to ensure the fuel and Na bond height would be as expected, but these did not critically influence thermal hydraulics)
- The last fuel specification (Mark-V) written reflected much-relaxed tolerances and was largely based upon delivering a homogeneous fuel with a known/controlled reactivity.
EBR-II Mark-V Fuel Specification

- Recycled metallic fuel (U-Pu-Zr in HT9 cladding) containing carry-over RE fission products and actinide isotopes characteristic of recycled EBR-II fuel
- Written to exclude process control information.
- Chemistry
  - Slug top & bottom sampled for homogeneity.
  - C, O, N, and Si impurity analysis to ensure Zr in solution (Zr important to FCCI and these impurities tie-up Zr and render it ineffective); all other impurity elements analyzed for information only.
  - Actinide tolerances relaxed (e.g., Pu = 20 ± 1 wt.%); isotopics measured for use in determining acceptable slug weight
EBR-II Mark-V Fuel Specification

- Fuel slug dimension and weight
  - Dimensional guidelines relaxed (e.g., ±0.003 in. on diameter, ±0.1 in on length)
  - Fuel slug mass (tolerances set by physics analysis for EBR-II):
    \[ 1.23^{239}Pu + 235U + 0.51^{240}Pu + 0.10^{238}U = 45.4 \pm 2.3 \text{ grams per slug} \]
  - Na bond height (0.50 ± 0.25 in.)

- Radiography to measure Na bond height used for bond quality.
- Closure welds qualified and 100% visual inspection, 100% leak check (unless qualification showed this could be eliminated).
Fuel Specification

- Draft metallic fuel specification for PRISM (by GE with ANL input) based upon Mark-V approach (GE 23A3912, Rev.B, 2-28-92).

- MOX fuel specification should utilize same approach, but performance issues will require some dimensional tolerances to remain tight due to He bond. Control of starting O/M, etc. may require additional chemistry control.
Experience with Fuels Containing Minor Actinides
GNEP Fuels Campaign Objective

- The ultimate objective of the fuel development campaign is to develop and qualify the initial transmutation fuel/target for use in fast burner reactors over the entire range of compositions:
  - obtain closure of the fuel cycle,
  - maintain the commercial competitiveness for nuclear energy
- If a step-wise approach is adopted by DOE in closing the fuel cycle, develop and qualify fuels to support the intermediate steps.
- Maintain a long-term R&D program
  - risk mitigation
  - capabilities and long-term competitiveness

*Qualification means* “demonstration that the fuel will perform predictably and acceptably under normal and accident conditions”. This is achieved by
- Rigorous testing for a limited number of fuel compositions, fabrication processes and cladding materials up to the level of lead-test assemblies
- An extensive modeling and simulation approach to quickly extend the empirical database to the entire range of variables that are needed to meet the GNEP objectives.
SFR Transmutation Fuels with Minor Actinides (MAs) and Rare Earth (RE) Fission Products

Unique Features of SFR Transmutation Fuels
- Pu content, which depending on CR selected may be higher than historic database (with corresponding decrease in U content)
- Minor actinides (Am, Np, Cm) present in significant quantities
- Rare earth fission product (La, Ce, Pr, Nd) carry-over from recycle step may be non-trivial

Gives Rise to Challenges and Unknowns
- Need for remote fuel fabrication
- Likely need for new fabrication methods (e.g., due to Am volatility; waste minimization, etc.)
- Effects on fuel performance must be determined
AFC Test Series in the Advanced Test Reactor

<table>
<thead>
<tr>
<th>Test Strategy</th>
<th>AFC-1</th>
<th>AFC-2</th>
<th>AFC-3</th>
<th>AFC-4</th>
</tr>
</thead>
<tbody>
<tr>
<td>AFC Test Series in the Advanced Test Reactor</td>
<td>Scoping – Many compositions</td>
<td>Scoping – Focused compositions</td>
<td>Focused compositions</td>
<td>Focused compositions</td>
</tr>
<tr>
<td>Nominal conditions</td>
<td>Nominal conditions</td>
<td>Nominal &amp; limiting conditions</td>
<td>Nominal conditions</td>
<td></td>
</tr>
<tr>
<td>Capsule Type</td>
<td>Drop-in</td>
<td>Drop-in</td>
<td>Instrumented lead</td>
<td>Drop-in</td>
</tr>
<tr>
<td>Fuel Types</td>
<td>Metals Nitrides</td>
<td>Metals Oxides</td>
<td>Metals Oxides</td>
<td>Metals Oxides</td>
</tr>
<tr>
<td>Key Features</td>
<td>Baseline + MA</td>
<td>Baseline + MA + RE</td>
<td>Temperature control</td>
<td>Recycle feed Remote fabrication</td>
</tr>
</tbody>
</table>

Test series in progress.
Future test series.
AFC-2A,B Currently Under Irradiation in the ATR

- **AFC-2A,B Test Matrix**

<table>
<thead>
<tr>
<th>Rodlet</th>
<th>AFC-2A&amp;B</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>U-20Pu-3Am-2Np-15Zr</td>
</tr>
<tr>
<td>2</td>
<td>U-20Pu-3Am-2Np-1.0RE-15Zr</td>
</tr>
<tr>
<td>3</td>
<td>U-20Pu-3Am-2Np-1.5RE-15Zr</td>
</tr>
<tr>
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</tr>
<tr>
<td>5</td>
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</tr>
<tr>
<td>6</td>
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</tr>
</tbody>
</table>

RE=6% La, 16% Pr, 25% Ce, 53% Nd

- **AFC-2A,B Test Objectives**
  - LHGR = 350 W/cm; PICT = 550°C
  - Burnups of 10 at.% (2A) and 25 at.% (2B)
  - Group recovery of 30 year-cooled PWR TRU
  - Effect of RE fission product carry-over on FCCI
AFC-2C,D Currently in Fabrication

AFC-2C,D Test Matrix

<table>
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<tr>
<td>1</td>
<td>(U_{0.75},Pu_{0.20},Am_{0.03},Np_{0.02})O_{1.95}</td>
</tr>
<tr>
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- LHGR = 350 W/cm; PICT = 550°C
- Burnups of 10 at.% (2A) and 25 at.% (2B)
- Group recovery of 30 year-cooled PWR TRU
- Effect of O/M on FCCI
Metallic Fuel with MA—X501 Fabrication

- U-20.2Pu-9.1Zr-1.2Am-1.2Np
- Injection cast at 1450°C
- Inhomogeneous microstructure
- Am and Np segregate to phases with variable composition

Lower section, slower cooling

6 U
3 Pu
6 Np
86 Zr

21-47 U
14-49 Pu
9-19 Zr
0-25 Am
0-18 Np
Impurities
Metallic Fuel with MA—X501 Irradiation

- LHGR = 450 W/cm
- PICT = 540°C
- Burnup = 7.6%
- $^{241}$Am transmutation = 9.1%

Gas Release
- Fission gas = 79%
- Helium = 90%
Metallic Fuel with MA—FUTURIX-FTA

- **U-29Pu-4Am-2Np-30Zr** and **Pu-12Am-40Zr** Fuel Pins
  - Pins received at Phénix, Sept06
  - Safety Report Completed, Jan07
  - Approval granted by Phénix Safety Committee, Feb07
  - Assembly Fabrication Complete, Mar07
  - Approval by French National Safety Authority, Apr07
  - Insertion into Phénix Reactor, Apr07
  - Phénix startup for Cycle 55-1, May 5, 2007
  - Irradiation currently underway
    - Irradiation complete at 240 EFPDs, Jan09
Metallic Fuel with MA+RE—METAPHIX

- Joint research with ITU
- Fabrication MA-Metal slug by arc-melting
- Property measurements of MA-Metal slug
  - Melting temperature
  - Thermal conductivity
  - Mechanical property
  - Microstructure
  - Compatibility with cladding material
- Irradiation tests at Phenix (Jan. 2004~)
  (METAPHIX-1-2-3)
  - Reference Metal (U-Pu-Zr) fuel
  - 5%MA-Metal fuel
  - 2%MA/2%RE-Metal fuel
  - 5%MA/5%RE-Metal fuel
  (Burnup, 1at.%, 5at.%, 10at.%)

(From Arai and Pillon, 2004)
MOX Fuel with MA+RE—Fabrication

MA-bearing MOX Fuel
Fabrication and Characterization

- Simplified pelletizing method
  - Die wall lubrication pelletizing, Sintering, O/M adjustment
    - Good accuracy of Pu content adjustment by solution mixing
    - Pellet fabrication with die wall lubrication
    - Good Sintering of Am/Np/simulated FP pellet with low O/M ratio

- Property measurements
  - MA and simulated FP bearing MOX fuel
    - Melting point
    - Thermal conductivity
    - Oxygen potential, etc.

(From Minato, et al., 2006)
MA-MOX Fuel Development in Europe (2)

Conclusion on homogeneous recycling
– No significant impact on the fuel properties and fuel behaviour proved at least up to 6.4% of BU
  • Formation of a central hole, equivalent to what is observed on standard fuel irradiated under similar condition
  • No signal of inner corrosion, no accelerated corrosion
  • Normal FP and Pu redistribution
  • Behaviour of FG similar to that of standard pins
– But higher helium production (x4)
  – 100% released during irradiation
  – 58% released during cooling time

(From Arai and Pillon, 2004)
Modeling & Simulation Fully Integrated with Fuel Development and Qualification

Modeling & Simulation

Analysis Platforms and Codes

Fabrication Models

Performance Models

Integral Experiments

Fabrication

Characterization

Irradiation

PIE

R&D Infrastructure

Fabrication Capabilities (Small hot-cells, Gloveboxes)

State-of-the art Characterization Equipment & Support Infrastructure

Rodlet-scale Fast-Spectrum Irradiation Capability

State-of-the art PIE equipment & Support Infrastructure

November 27-28, 2007
DOE/NRC Seminar Series on Sodium Fast Reactors
Fuel Performance and Qualification
Modeling and Simulation Strategy

(LIFE4, SIEX, LIFE-METAL, FRAPCON)

**Legacy Codes:**
- Sensitivity studies
- Design support for fuels
- Empirical model support

**ASCI Thermo-Mechanical Codes:**
- Intermediate-term results
- Test bed and feasibility assessment
- Import ASCI technology to NE needs

**Lower length-scale Modeling:**
- Fundamental understanding
- Reduced reliance on empirical data
- Collaborations with Office of Science

**Basic Experiments**
- Fundamental properties
- Phenomenological tests for model development

**Next Generation Fuel Performance Code:**
- Long-term qualification code
- Includes all important phenomenology
- Specifically tailored to fuel physics
- State-of-the-art numerical models
Modeling and Simulation Forecast

- **Legacy Codes**
- **ASCI Thermo-Mechanical Codes**
- **Fuel Performance Code**

Time:
- 2012
- 2017

Technology:
- Yesterday's
- Today's
- Tomorrow's

- ~5 yr
- ~10 yr
Approach to Fuel Development and Qualification
Review of 4 phases of fuel development process
- 1. Fuel candidate selection
- 2. Concept definition and feasibility
- 3. Design improvement and evaluation
- 4. Fuel qualification and demonstration

Example of EBR-II & FFTF experience with fuel qualification/confirmation testing
### TRL Objective in Fuel Qualification

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I. Fuel Candidate Selection

II. Concept Definition & Feasibility
- Scoping Fuel Test Series 1 (inexpensive screening tests)
- Scoping Fuel Test Series 2 (prototypic conditions)
- Scoping Transient Tests

III. Design Improvement and Evaluation
- Design Parameters Test
- Fabrication Variables Test
- High Power Test (2-sigma LHGR or fuel temperature)
- Undercooling test (2-sigma cladding temperature)
- Transient Response Tests
  - Fuel Downselection
  - DBA Transient Tests
  - Final Fuel Selection

IV. Fuel Qualification and Demonstration
- LTAs, Nominal Conditions
- LTAs, Overpower and/or Undercooling Conditions
- Core Safety Testing
- DBA Confirmation Transient Tests

Years from Start

In-reactor irradiation
Postirradiation examination

Prototypic Spectrum Test Facility

Implementation
Fuel Development Phase 1: Fuel Candidate Selection

- **Objective:** Based on previous experience, identify candidate fuel forms/concepts that appear capable of meeting mission needs.
- **Selection criteria might include:**
  - Ability to accommodate desired fuel compositions
  - Experience with similar fuel types or analogues
  - Suitability of established fabrication techniques, or the potential for successful innovative techniques
  - Anticipated performance capabilities (e.g., temperature, burnup, or fluence)
  - Anticipated safety-related behavior (which may be quite speculative at an early stage)
  - Suitability of design, considering issues such as fuel-cladding compatibility, fuel-coolant compatibility, and fuel properties
  - Compatibility with envisioned back-end fuel cycle technology
  - Expected cost of fabrication
- **Achieves TRL 2**
Fuel Development Phase 2:
Concept Definition and Feasibility

- **Objective:** Establish a reference concept/design
- Determine how the fuel can be fabricated
  - Process scoping & feasibility
  - Fabricate characterization and test samples
- Determine and assess key properties
  - Feasibility issues for irradiation testing
- Use scoping irradiation tests to assess phenomena envisioned to impact feasibility and fuel lifetime
  - Simple experiments
  - Prototypic conditions as possible
- **Achieves TRL 4**
**Objectives:**
- Optimize the fuel design for economics, performance and safety
- Produce a *Fuel Specification* and a *Fuel Safety Case* for a core of fuel
- Establish predictive fuel performance code (or codes)

**Develop engineering-scale fabrication processes and equipment**

**Assess fuel properties vs. composition or processing variations**

**Irradiation testing to:**
- Determine sensitivity of performance to fuel design and fabrication variables and to operating conditions
- Establish burnup limits and safety margins for various operating conditions (normal and off-normal)

**Develop fuel behavior models and predictive codes**

**Achieves TRL 6**
Fuel Development Phase 4: Fuel Qualification and Demonstration

- **Objectives:**
  - Qualify production-line fuel as the driver fuel for a demonstration reactor
  - Demonstrate the safety and reliability of a core of fuel through successful operation of the demonstration reactor
  - Validate predictive fuel performance code (or codes)

- **Demonstrate production of fuel in conformance with Fuel Specification**

- **Demonstrate through LTA irradiation that fuel/fuel assembly behavior is within the bounds of the Fuel Safety Case**

- **Accumulate reactor performance data and operating experience with a core of fuel to support licensing of first-of-a-kind unit**

- **Achieves TRL 7 or 8**
Example for Metallic Fuels in EBR-II

- Confirmation Testing (Mark-III, IIIA, and IV)
  
  - Performance phenomena limiting useful lifetime of a fuel assembly (breach, hardware dimensional change, etc.) are known. Limits can be predicted and pre-set.
  
  - Use of modeling [Cumulative Damage Fraction (CDF) or other].
  
  - Run a set of assemblies representing the range of possible operating conditions to an exposure (burnup, neutron fluence, etc.) in excess of the limiting exposure to prove capability is as predicted.
  
  - The tests lead the remaining core in exposure.
  
  - Four assemblies used [1 inner core, 1 outer core (normal flow), 1 outer core (high flow), 1 orificed to nominally achieve 2-σ peak cladding temperatures]
## Example for Oxide and Metallic Fuels in FFTF

### Confirmation Tests in FFTF to Support Exposure Limits

*(from Baker, et al, 1990)*

<table>
<thead>
<tr>
<th>Type</th>
<th>Fuel</th>
<th>Duct &amp; Cladding Material</th>
<th>Number of FFTF Tests*</th>
<th>Transient Testing</th>
<th>Driver Cores Produced</th>
</tr>
</thead>
<tbody>
<tr>
<td>Series I</td>
<td>(U, Pu)O₂</td>
<td>316</td>
<td>17F</td>
<td>Yes</td>
<td>2 cores</td>
</tr>
<tr>
<td>Series II</td>
<td>(U, Pu)O₂</td>
<td>316</td>
<td>--</td>
<td>--</td>
<td>2 cores</td>
</tr>
<tr>
<td>Series III.a</td>
<td>(U, Pu)O₂</td>
<td>D9</td>
<td>1F, 4R</td>
<td>No</td>
<td>--</td>
</tr>
<tr>
<td>Series III.b</td>
<td>U-10Zr</td>
<td>HT9</td>
<td>2P, 6F</td>
<td>No</td>
<td>--</td>
</tr>
<tr>
<td>Series IV</td>
<td>UO₂</td>
<td>D9</td>
<td>1P</td>
<td>No</td>
<td>--</td>
</tr>
</tbody>
</table>

*F=full assembly, P=partial assembly, R=related test*
Summary

- SFR Fuels Experience in the US
  - Fuel Types
  - Fuel Performance Issues
  - Experience/Testing

- Fuel Test Plan (1994) for PRISM Prototype
- Experience with Fuels Containing Minor Actinides
- Fuel Specification
- Fuel Development and Qualification
Spent Nuclear Fuel Reprocessing

Dr. Terry Todd
Idaho National Laboratory

Nuclear Regulatory Commission Seminar
Rockville, MD
March 25, 2008
Outline

- Spent nuclear fuel
- Background and history of spent nuclear fuel reprocessing
- PUREX process description
- Current reprocessing activities in the world
- Criticality control in PUREX plants
- Accidents in PUREX plants
- Recent process modifications to PUREX
  - COEX/NUEX/UREX
- Questions
Spent Nuclear Fuel – what is it?

Most heat production is from Cs and Sr, which decay in ~300 yr
Most radiotoxicity is in long-lived fission products and the minor actinides, which can be transmuted and/or disposed in much smaller packages.
Spent Nuclear Fuel – current US situation

- Currently stored in pools or dry storage at the 60+ nuclear reactor sites in the U.S.
- Generated at approximate rate of 2100 MTHM/yr
- Slated for direct disposal into Yucca Mountain geologic repository
  - Yucca Mountain is not licensed or open at this time, spent fuel inventory will exceed legislated capacity before it is opened

March 25, 2008
Spent nuclear fuel accumulation – direct disposal

- Technical Capacity
- Current Nuclear Energy Generation
- Statutory Capacity of First Repository

Year:
- 1980
- 1990
- 2000
- 2010
- 2020
- 2030
- 2040
- 2050

Civilian Spent Nuclear Fuel (MTHM Cumulative):
- 0
- 30,000
- 60,000
- 90,000
- 120,000
- 150,000

Cumulative Spent Nuclear Fuel (MTHM Cumulative):
- 30,000
- 60,000
- 90,000
- 120,000
- 150,000

Chart indicates the accumulation of spent nuclear fuel from 1980 to 2050, with a focus on the technical capacity and current nuclear energy generation.
Spent Fuel Processing (recycling)

96% of the metals in Spent Fuel (excluding cladding and hardware) can be recovered, with only a small fraction sent to the geologic repository.
Spent fuel recycling

**Benefits**
- Natural resource conservation
- Reduction of waste heat load and radiotoxicity
- Reduced dependence on foreign oil, LNG, and coal

**Challenges**
- Cost
- Impact to the environment
- Proliferation and safety concerns
- Public acceptance
Spent fuel – radiotoxicity
Began during Manhattan Project to recover Pu-239
- Seaborg first separated microgram quantities of Pu in 1942 using bismuth-phosphate precipitation process
- Process scaled to kilogram quantity production at Hanford in 1944
  - A scale-up factor of $10^9$ !!!

Solvent extraction processes followed to allow concurrent separation and recovery of both U and Pu and

Reprocessing transitioned from defense to commercial use
- Focus on recycle of uranium and plutonium
- Waste management
Bismuth Phosphate Process

- Dissolution of irradiated fuel or targets in nitric acid
- Pu valance adjusted to Pu (IV) with sodium nitrite
- Add sodium phosphate and bismuth nitrate
  - Pu (IV) precipitates as Pu$_3$(PO$_4$)$_4$
- PPT re-dissolved in nitric acid, oxidized to Pu (VI), then re-ppt BiPO$_4$ to decontaminate Pu from fission products
- Recover Pu by reducing to Pu (IV) and re-ppt
- Repeat cycles w/ LaF to further decontaminate
Advantages of Bismuth Phosphate Process

- Recovery of >95% of Pu
- Decontamination factors from fission products of $10^7$

Disadvantages of Bismuth Phosphate Process

- Batch operations
- Inability to recovery uranium
- Required numerous cycles and chemicals
  - Produced large volumes of high-level waste
REDOX Process

- First solvent extraction process used in reprocessing
  - Continuous process
  - Recovers both U and Pu with high yield and high decontamination factors from fission products
- Developed at Argonne National Laboratory
- Tested in pilot plant at Oak Ridge Nat. Lab 1948-49
- REDOX plant built in Hanford in 1951
- Used at Idaho for U-235 recovery

Hanford REDOX -Plant (1951)
REDOX Process

- Hexone (methyl isobutyl ketone) used as the extractant
  - Immiscible with water
  - Used to purify uranium ore concentrates
  - Extracts both uranyl and plutonyl nitrates selectively from fission products

- Plutonium oxidized to Pu (VI) for highest recovery

- U (VI) and Pu (VI) co-extracted, then Pu is reduced to Pu (III) by ferrous sulfamate and scrubbed from the solvent

- Hexone is highly flammable and volatile

- Large amounts of nonvolatile salt reagents added to process increased waste volume
BUTEX Process

- Developed in late 1940’s by British scientists at Chalk River Laboratory
- Utilized dibutyl carbitol as solvent
  - Lower vapor pressure than hexone
- Nitric acid was used as salting agent
  - Replaced need to use aluminum nitrate as in REDOX process
    - Lower waste volumes
- Industrial operation at Windscale plant in UK until 1976
Tributyl phosphate used as the extractant in a hydrocarbon diluent (dodecane or kerosene)

- Suggested by Warf in 1949 for the recovery of Ce (IV) from rare earth nitrates
- Developed by Knolls Atomic Power Lab. and tested at Oak Ridge in 1950-52
- Used for Pu production plant at Savannah River in 1954 (H-canyon facility still operational in 2008)
- Replaced REDOX process at Hanford in 1956
- Modified PUREX used in Idaho beginning in 1953 (first cycle)
Advantages of PUREX over REDOX process

- Nitric acid is used as salting and scrubbing agent and can be evaporated – results in less HLW
- TBP is less volatile and flammable than hexone
- TBP is more chemically stable in a nitric acid environment
- Operating costs are lower
West Valley, NY
- First plant in US to reprocess commercial SNF
- Operated from 1966 until 1972
- Capacity of 250-300 MTHM/yr
- Shutdown due to high retrofit costs associated with changing safety and environmental regulations and construction of larger Barnwell facility

Morris, IL
- Construction halted in 1972, never operated
- Close-coupled unit operations with fluoride volatility polishing step

Barnwell, SC
- 1500 MTHM capacity
- Construction nearly completed- startup testing was in progress
- 1977 change in US policy on reprocessing stopped construction
- Plant never operated with spent nuclear fuel
Commercial reprocessing history Non-US (all PUREX)

**France**
- Magnox plant in Marcoule began operation in 1958 (~400 MT/yr)
- Magnox plant in La Hague began operation in 1967 (~400 MT/yr)
- LWR oxide plant (UP2) began in La Hague in 1976 (800 MT/yr)
- LWR oxide plant (UP3) began in La Hague in 1990 (800 MT/yr)

**United Kingdom**
- Windscale plant for Magnox fuel began in 1964 (1200-1500 MT/yr)
- THORP LWR oxide plant began in 1994 (~1200 MT/yr)

**Japan**
- Tokai-Mura plant began in 1975 (~200 MT/yr)
- Rokkasho plant currently undergoing hot commissioning (800 MT/yr)
Reprocessing history in Russia

- **Mayak**
  - Plant B operated from 1949 to 1960
    - Acetate precipitation followed by precipitation from fluoride solutions
    - High level wastes discharge to Techa river, then Lake Karachai
  - Plant BB operated from 1957 to 1987
    - Similar acetate precipitation process, but repeated twice
  - Plant RT-1 (PUREX Process)
    - Operation began in 1976
    - 400 MTHM/yr capacity
    - Multiple lines to process commercial, HEU and naval fuels
PUREX Process – history Russia

- **Krasnoyarsk -26**
  - Processing of Pu production reactor fuel began in 1964 using PUREX process
  - Construction of new RT-2 plant began in 1972 (1000-1500 MTHM/yr capacity)
    - *Plant construction never completed*

- **Tomsk -7**
  - Processing of Pu production reactor fuel began in sometime after 1955 using PUREX process
Tri-butyl phosphate forms soluble complexes with uranyl nitrate and plutonium nitrate (neutral species of U(VI) and Pu(IV)).

Spent fuel is dissolved in nitric acid and is then mixed with a solution of TBP in a hydrocarbon diluent (immiscible with aqueous phase).

At higher nitric acid concentrations (>0.5 M) the plutonium and uranium partition to the organic (solvent) phase while most of the metals and fission products stay in the aqueous phase.

Once separated from the fission products, the solvent can be mixed with another aqueous solution of low acidity (<0.01 M) and the uranium and plutonium will partition back to the aqueous phase.

To separate plutonium from uranium, a reductant is added to the aqueous stream, reducing Pu(IV) to Pu(III), which is not soluble in the organic solvent and partitions to the aqueous phase while U(VI) remains in the solvent.
PUREX Process- Basic principles

1) Mix Phases

2) Allow to Settle

\[
\begin{align*}
\text{UO}_2^{2+} + 2\text{NO}_3^- + 2\text{TBP} &\rightleftharpoons \text{UO}_2(\text{NO}_3)_2 \cdot 2\text{TBP} \\
\text{Pu}^{4+} + 4\text{NO}_3^- + 2\text{TBP} &\rightleftharpoons \text{Pu}(\text{NO}_3)_4 \cdot 2\text{TBP}
\end{align*}
\]
PUREX Process- Basic principles

- Extraction
  - Separates metal to be recovered
  - Recovers product in solution

- Scrubbing
  - Removes impurities from metal

- Stripping
  - Feeds solution

Organic Solvent

Feed Solution
TBP is an effective extractant, but is too dense and viscous to use pure
- Hydrocarbon diluent used to improve physical characteristics
- Typically 30 vol% TBP is used in the PUREX process
- Diluents typically dodecane or kerosene (straight or branch chain hydrocarbons ranging from C-10 to C-14)

Salting effect
- Uranium and plutonium extraction is a function of nitrate concentration (called salting effect)
PUREX Process – Nitric acid dependence

\[ \frac{[\text{HNO}_3]}{\text{M}} \]
PUREX Process – Nitric acid dependence

![Graph showing the dependence of \( D \) on \([\text{HNO}_3] / M\) for U(IV), Np(IV), and Pu(IV).](image-url)
Advantages of liquid-liquid extraction
- Continuous operation/ High throughput
- Countercurrent operation/ High purity and selectivity
- Recycle solvent, minimizing waste

Disadvantages of liquid-liquid extraction
- Solvent degradation due to hydrolysis and radiolysis
- Degradation products interfere with process chemistry
  - Dibutyl and monobutyl phosphates
    - Efficiently extract Pu, but cannot strip Pu from DBP or MBP
- Requires substantial tankage and reagents
PUREX Process – Process unit operations

- Fuel decladding
- Dissolution/ feed clarification
- Separations
- Product conversion
- Waste treatment
Fuel Decladding
Dissolution/ feed clarification

- Nitric acid dissolves UO₂ pellet from cladding hull, forming UO₂(NO₃)₂ in solution
- Dissolver product contains approx. 300 g/l uranium
- Releases radioactive off-gas (iodine, krypton, xenon, carbon-14, small amounts of tritium)
- Undissolved solids are removed by centrifugation before transfer to extraction process
Separations

- Continuous, countercurrent extraction operations are performed in mixer settlers, pulse columns or centrifugal contactors
- First cycle separates uranium and plutonium together from fission products
- U and Pu are then separated and sent to separate purification cycles
PUREX Process – Process unit operations

Separations
– Countercurrent PUREX flowsheet

Coextraction U and Pu
FP Scrubbing
U Scrubbing
Pu Stripping
U Stripping

Solvent

Raffinates (FP)
Feed (U, Pu, FP...) Scrub Pu Solution
Reducing Solution
Diluted Nitric Acid

March 25, 2008
Mixer Settlers
- Discrete stage units (with efficiencies < 1)
- Low capital cost
- Requires large amount of floor space (but low headroom)
- Large solvent inventory
- Long residence times
Pulse Extraction Column

- Several feet of column needed for one theoretical stage
- Low capital cost
- Requires large amount of head space (40-50’), but little floor space
- Moderate solvent inventory
- Long residence times
PUREX Process – Process unit operations

- Pulse column at La Hague
  UP3 plant
**Centrifugal Contactors**

- Each unit near one theoretical stage
- Higher capital cost
- Requires little headroom or floor space, but requires remote maintenance capability
- Small solvent inventory
- Short residence times
Product Conversion

- Uranyl nitrate is converted to UO$_3$ by denitration at elevated temperature
  - Produces NO$_x$ off-gas
- Plutonium nitrate is precipitated by oxalate or peroxide and calcined to PuO$_2$
PUREX Process – Process wastes

LIQUIDS

- HLW (RAFFINATE FROM FIRST CYCLE – TANK WASTE)
- LAW (SOLVENT SCRUB; EVAPORATORS)

GASES

- $^{85}$Kr (DISSOLVER OFF-GAS; UNTREATED IN THE PAST)
- $^{129}$I (DISSOLVER OFF-GAS; REMOVED FROM EARLIEST DAYS)
- $^{14}$C (AS CO$_2$) (DISSOLVER OFF-GAS; UNTREATED IN THE PAST)
- $^3$H (MOSTLY AS TRITIATED WATER VAPOR)

SOLIDS

- HLW (CONTAMINATED EQUIPMENT; CLADDING HULLS?)
- LAW (MISCELLANEOUS WASTES FROM OPERATIONS)
High Level Waste Treatment

- High level waste is the remaining liquid after U and Pu have been removed (contain fission products and transuranium actinides)
- Wastes from weapons production at Hanford and Savannah River were neutralized using NaOH and stored in carbon steel underground tanks
  - Hanford - 177 ~ one million gallon tanks
  - Savannah River – 51 ~ 750,000 gallon tanks
- Multi-billion dollar waste treatment plants are in operation (Savannah River) or under construction (Hanford) to treat these wastes by converting the highly radioactive liquids into glass
- France, Russia and the UK convert their high-level waste into glass
PUREX Process – Current Commercial Operating Facilities

THORP, UK

La Hague, France

Rokkasho, Japan
**Commercial PUREX operations**

- **La Hague, Rokkasho and THORP**
  - Utilize pulse columns for first cycle extraction
  - Mixer settlers and centrifugal contactors in purification cycles
  - All are located near the ocean, discharge iodine to the ocean

- La Hague UP2 and UP3 plants - 800 MTHM/yr each
- Rokkasho – based on UP3 design- 800 MTHM/yr
- THORP – 1200 MTHM/yr design capacity, actual capacity less than 1000 MTHM/yr
Commercial PUREX operations – La Hague UP2/UP3
Criticality Control in the PUREX Process

Factors that affect criticality safety

- Fissile nuclide ($^{235}$U, $^{233}$U or $^{239}$Pu)
- Fraction of fertile nuclide diluting fissile nuclide ($^{238}$U, $^{232}$Th or $^{240}$Pu)
- Mass of fissile nuclide
- Geometry
- Volume
- Concentration of fissile nuclide
- Neutron moderators
- Neutron reflectors
- Neutron absorbers
The preferred method of criticality control are engineered controls, such as limiting geometry to be criticality safe under any credible conditions

- This often leads to conservative assumptions for credible conditions and adds to cost and complexity of the process
- Limits equipment size and process throughput

Administrative controls have greater operational complexity (procedures, standards, etc), but offer greater design flexibility and throughput

- Typically, administrative controls require a double parameter failure for a criticality to occur (no one single control failure would cause a criticality)
### Criticality Control in the PUREX Process

#### Single-parameter subcritical limits for uniform aqueous solutions

<table>
<thead>
<tr>
<th>Parameter</th>
<th>(^{235}\text{U})</th>
<th>(^{233}\text{U})</th>
<th>(^{239}\text{Pu})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass of fissile material, g</td>
<td>760</td>
<td>550</td>
<td>510</td>
</tr>
<tr>
<td>Solution cylindrical diameter, cm</td>
<td>13.9</td>
<td>11.5</td>
<td>15.7</td>
</tr>
<tr>
<td>Solution slab thickness, cm</td>
<td>4.6</td>
<td>3.0</td>
<td>5.8</td>
</tr>
<tr>
<td>Solution volume, L</td>
<td>5.8</td>
<td>3.5</td>
<td>7.7</td>
</tr>
<tr>
<td>Concentration of fissile nuclide, g/L</td>
<td>11.5</td>
<td>10.8</td>
<td>7.0</td>
</tr>
<tr>
<td>Areal density of fissile nuclide, g/cm(^2)</td>
<td>0.4</td>
<td>0.35</td>
<td>0.25</td>
</tr>
<tr>
<td>Uranium enrichment wt% (^{235}\text{U})</td>
<td>1.0 %</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Criticality accidents in reprocessing plants

**Mayak 1953**
- Procedural errors led to an unrecognized accumulation of 842 g of plutonium (as Pu nitrate solutions) in one vessel, which became critical and brought the vessel contents to boiling. The operators transferred contents of another vessel to the first, ending the reaction.

**Mayak 1957**
- The accident occurred in a glovebox assembly within which uranium solution was precipitated into vessels. An unexpectedly large amount of uranium precipitate accumulated in a filter receiving vessel. The operator at the glovebox observed the filter vessel bulge prior to ejection of gas and some solution and precipitate from the vessel within the glovebox.

**Mayak 1958**
- Following the criticality accident at the same facility in 1957, an apparatus had been constructed to test criticality phenomena in fissile solutions. A 400-liter tank on a platform was used for measurements involving solutions; after each experiment, the tank was drained into individual 6-liter containers of favorable geometry. On this occasion, the tank contained uranyl nitrate solution (90% U-235) and was being drained for another experiment. After filling several 6-liter containers, operators decided to circumvent the standard procedure to save time. Three operators unbolted the tank and lifted it to pour directly into containers. The presence of the operators provided sufficient neutron reflection to cause a criticality excursion, producing a flash of light and ejecting solution as high as the ceiling, 5 meters above the tank.
Criticality accidents in reprocessing plants

- **Oak Ridge 1958**
  - A leak in a tank containing uranyl nitrate solution (93% U-235) was discovered on 15 June. The leak was not properly logged. The following day other tanks were being drained into a 55-gallon drum; uranium solution from the leaking tank also entered the drum. The operator nearest the drum noticed yellow-brown fumes rising from the drum's contents; he retreated before seeing a blue flash as the criticality excursion occurred. Excursion power output rose for at least 3 minutes, then ended after 20 minutes

- **Idaho 1959**
  - Air sparging cylinders containing highly enriched uranyl nitrate solution initiated a siphon that transferred 200 L of solution to a 5000 gallon tank containing about 600 liters of water. The resulting criticality lasted about 20 minutes
Criticality accidents in reprocessing plants

■ Idaho 1961
  - 40 L of 200 g/L uranyl nitrate solution was forced up from a 5 in diameter section of an evaporator into a 24 in diameter disengagement cylinder, well above normal solution level. Operators were attempting to clear a plugged line with air, which entered the evaporator, forcing the solution upward

■ Hanford 1962
  - Plutonium solution was spilled onto the floor of a solvent extraction hood. Improper operation of valves allowed a mixture of plutonium solutions in a tank that became supercritical. The excursion continued at low power levels for 37.5 hours, during which a remotely controlled robot was used to check conditions and operate valves. Criticality was probably terminated by precipitation of plutonium in the tank to a non-critical state
**Criticality accidents in reprocessing plants**

- **Mayak 1968**
  - Solutions of plutonium were being transferred from a large tank into a stainless steel vessel using a glass bottle. While a worker was pouring a second load from the glass bottle into the vessel, a criticality excursion occurred.

- **Idaho 1978**
  - A leaking valve allowed water to dilute the scrub solution used in the first cycle extraction process. This leak was undetected because of a failed alarm system. Because of the dilution, highly enriched uranium was stripped from the organic solvent (normally would remain in solvent). Over the course of a month, the concentration of uranium increased in the large diameter bottom of the scrub column, resulting in a criticality.
Tokai-mura 1999

Three operators were engaged in processes combining uranium oxide with nitric acid to produce a uranium-containing solution for shipment. The uranium involved was 18.8% U-235. The procedure used deviated from that licensed to the facility. In particular the uranium solution was being placed in a precipitation tank for dispensing into shipment containers, not the more narrow vessel (geometrically favorable to minimizing criticality risks) prescribed by license. While two workers were adding a seventh batch of uranium solution to the tank, a criticality excursion occurred.
Major industrial accidents in reprocessing plants

- **Red Oil**
  - Created by decomposition of TBP by nitric acid, under elevated temperature
    - *Influenced by presences of heavy metal (U or Pu), which causes higher organic solubility in aqueous solution and increases the density of the organic solution (possibly > aqueous phase)*
    - *Decomposition of TBP is a function of nitric acid concentration and temperature*
  - Primary concern is in evaporators that concentrate heavy metals in product
  - Red oil reactions can be very energetic, and have resulted in large explosions at reprocessing facilities
  - Typical safety measures include diluent washes or steam stripping of aqueous product streams to remove trace amounts of TBP before evaporation or denitration
  - Major accidents detailed in DNFSB report “Tech 33” Nov. 2003
Controls to avoid Red Oil accidents

How do we avoid red oil in reprocessing facilities?

- Temperature control
  - Maintain solutions at less than 130 °C at all times

- Pressure control
  - Adequate ventilation to avoid buildup of explosive gases

- Mass control
  - Minimize or eliminate organics (TBP) from aqueous streams
    - Decanters, diluent washes, etc.

- Concentration control
  - < 10 M HNO₃
  - With solutions of uranyl nitrate, boiling temperature and density must be monitored

- Multiple methods need to be employed so that no single parameter failure can lead to red oil formation
Other major accidents in reprocessing facilities

- **Mayak 1957**
  - Liquid high-level waste was stored in underground tanks. The high level waste, coming from the B plant, contained sodium nitrate and acetate salts, from the acetate precipitation process. Cooling system in one of the tanks failed, and the temperature in the tank rose to 350 °C. The tank contents evaporated to dryness, causing a massive explosion (estimated to be equivalent to 75 tons of TNT). Over 20 MCi of radioactivity were released to the environment.

- **Tokai-mura 1997**
  - A fire occurred in the bitumen waste facility of the demonstration reprocessing plant at Tokai-mura. Bitumen is used to solidify intermediate-level activity liquid radioactive waste. The fire apparently occurred after errors made in monitoring a chemical reaction. The fire was not completely extinguished and about ten hours later, after chemicals had accumulated, an explosion occurred which ruptured the confinement of the facility.
Other major accidents in reprocessing facilities

- **Hanford 1997**
  - Hydroxylamine nitrate and nitric acid were stored in a tank and allowed to evaporate to dryness. The resulting explosion destroyed the tank and blew a hole in the roof of the building. Hydroxylamine is a reagent used to reduce plutonium valance from (IV) to (III).

- **THORP 2005**
  - A pipe failure resulted in about 83,000 L of highly radioactive dissolver solution leaking into the stainless-steel lined feed clarification of the THORP facility. This solution contained about 20 MT of uranium and plutonium. The leak went undetected for months before being discovered. No injuries or exposure to radiation. The plant is still shutdown in 2008.
Recent modifications to the PUREX Process

- Industrial reprocessing firms have a high degree of confidence in the PUREX process, however, the PUREX process has been the subject of criticism for the past 30 years related to the separation of a pure plutonium stream.
- Recall that the PUREX process co-extracts both uranium and plutonium, then partitions them into separate streams.
- Modifications to the PUREX process have recently been proposed and developed that leave a small fraction of the uranium with the plutonium, producing a mixed product for production of mixed oxide (MOX) fuel.
- These modified processes have been called COEX™, NUEx or UREX+ 3 and are all based on modified PUREX chemistry.
- Calling these processes “co-extraction” to differentiate them from PUREX is misleading because the PUREX process also co-extracts uranium and plutonium.
Recent modifications to the PUREX Process

- Each specific process has its own proprietary methods of stripping plutonium from the solvent, with a fraction of uranium.
- In the PUREX process, the nitric acid concentration in the second scrub is kept higher than ~0.5 M to keep the uranium in the organic solvent, while the plutonium is reduced to the trivalent state and partitions to the aqueous phase.
- In the modified process, the acid concentration in the second scrub stream is maintained at a controlled value (typically lower than 0.5 M) to allow a small amount (~1%) of the uranium to partition to the aqueous stream along with the plutonium (III).
- After the plutonium and small fraction of uranium are removed in the second scrub stream, uranium is stripped from the solvent by using dilute (0.01 M) nitric acid.
Recent modifications to the PUREX Process

Simplified flowsheet for U and U/Pu products

Feed

Scrub

2nd Scrub

U Strip

Raffinate

Pu + U Product

U Product

Solvent
Summary

- Spent nuclear fuel reprocessing is a mature technology, having over 50 years of industrial experience with the PUREX process.
- Nuclear energy must solve the waste disposal issue soon for it to grow. This solution could include building more repositories, reprocessing fuel and/or a combination of both.
- History has shown that there must be a strong emphasis on safety, including criticality safety, safeguards and industrial safety.
- New “evolutionary” processes employ minor adjustments to PUREX process chemistry to keep from producing pure plutonium and facilitate more near-term implementation.
UREX+ Process Overview

Candido Pereira
Argonne National Laboratory

Topical Seminar Series on Nuclear Fuel Separation Processes
March 26, 2008
Outline

- Separations Overview
- Solvent Extraction
- Process Modeling and Simulation
- Processing Options
- Demonstration of UREX+ Processes
Separations are a central component of the GNEP scheme and implementation of an advanced fuel cycle in the U.S.
Separations Can Potentially Reduce the Volume, Activity and Toxicity of the Waste Bound for the Repository

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Oxide Fuel %</th>
<th>Significance</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>~95</td>
<td>Hazard comparable to ore.</td>
</tr>
<tr>
<td>Cs and Sr</td>
<td>0.3</td>
<td>~30 yr half-life. Significant heat source.</td>
</tr>
<tr>
<td>Pu and Np</td>
<td>~1</td>
<td>Isotopes have long half-lives. Recycle.</td>
</tr>
<tr>
<td>Am and Cm</td>
<td>0.1</td>
<td>Isotopes have long half-lives and high toxicity. Can be recycled to fast reactors.</td>
</tr>
<tr>
<td>I and Tc</td>
<td>0.1</td>
<td>&gt;10^5 yr half-life. Mobile in groundwater. Separate and stabilize for long-term disposal.</td>
</tr>
<tr>
<td>Other Fission Products</td>
<td>~3.5</td>
<td>Low heat and toxicity. Stabilize for disposal.</td>
</tr>
</tbody>
</table>
Examples of Some Potential Aqueous Processing Options Meeting GNEP Objectives: UREX+

<table>
<thead>
<tr>
<th>Process</th>
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<th>Product #3</th>
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<td>U/Pu/Np</td>
<td>Am</td>
<td>Cm</td>
<td>FP/Ln</td>
</tr>
</tbody>
</table>

Notes: (1) in all cases, iodine is removed as an off-gas from the dissolution process.
(2) processes are designed for the generation of no liquid high-level wastes

- **U**: uranium (contributor to dose rate, and the mass and volume of high-level waste)
- **Tc**: technetium (long-lived fission product, minor contributor to long-term dose)
- **Cs/Sr**: cesium and strontium (primary short-term heat generators, affect waste form loading and repository drift loading)
- **TRU**: transuranic elements: Pu - plutonium, Np - neptunium, Am - americium, Cm - curium (primary long-term dose rate contributors)
- **Ln**: lanthanide (rare earth) fission products
- **FP**: fission products other than cesium, strontium, technetium, iodine, and the lanthanides
Solvent Extraction
Aqueous solvent extraction processes are the reference methods under GNEP for treatment of LWR spent fuel

- High degree of technological maturity; industrial practice in France, the United Kingdom, Russia, and Japan
- Capable of achieving high decontamination factors for separated products
  - For thermal recycle, must eliminate high cross section fission products nearly completely (DF>10,000 required)
  - For fast reactor recycle, must reduce the rare earth content (DF>250 for separation of lanthanides from transuranics)
- Provides flexibility in degree of partitioning of radionuclides present in spent fuel
  - Suite of process variants under development
Example of Process Flowsheet Design

\[ D = \frac{[M]_d}{[M]_a} \]

- \( D \): Dilution Ratio
- \([M]_d\): Concentration of Acid in Dilute Solution
- \([M]_a\): Concentration of Acid in Acid Solution

Graph showing the relationship between Acid Concentration and the Dilution Ratio for different stages (M(1) and M(2)). The graph also indicates the concentration ranges for Dilute and Concentrated phases.
CCD-PEG Process from 2006 UREX+ Demonstration

- Feed: UREX Raffinate, All TRU and FP
- Scrub: HNO$_3$
- Strip: Guan, Carbonate, DTPA
- Wash: PEG-HNO$_3$

Extraction

Raffinate: All TRU and FPs

Product: Cs, Sr, Rb, Ba
Typical Profiles for Extraction Processes

AMUSE Simulation of Cs and La in CCD-PEG Process

Cs Stage Concentration Profile

La Stage Concentration Profile
The Argonne Model for Universal Solvent Extraction—AMUSE is used to:

- Design and optimize solvent extraction flowsheets
  - PUREX, UREX, NPEX, TRUEX, CCD-PEG, SREX
  - TALSPEAK module is under development
- Perform sensitivity analysis to determine key process variables and their control bound
  - Flow rates, number of stages, concentration of feed components, concentration of solvent, temperature
Structure of AMUSE code
Extraction Model Derived from Literature Data

Model of extraction behavior for Yttrium (III)
- Nitric acid (0.4-15 M)
- TBP in Kerosene

D value data from SCARGILL (1957)
Examples of AMUSE Outputs

Tabular reports are created for
- Influent and effluent compositions and flow rates
- Stage profiles that include
  - Distribution ratios
  - Component concentrations in organic and aqueous phases

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<thead>
<tr>
<th>Stream Identity</th>
<th>Flow Rate</th>
<th>Ag</th>
<th>Am</th>
<th>Ba</th>
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Aqueous Spent Fuel Processing Involves A Number Of Key Operations in Addition Solvent Extraction
Top-level Modeling of Aqueous Separations Operations: UREX
Demonstration of the UREX+ Processes
UREX+1a Process

- A baseline process for LWR treatment
- Demonstrated with dissolved spent fuel
  - Flowsheets developed using the AMUSE code
Fission Products and Actinides Are Separated Based on Chemical Differences: UREX+1a Example

Thermal Neutron Fission of U-235

| La  | Ce  | Pr  | Nd  | Pm  | Sm  | Eu  | Gd  | Tb  | Dy  | Ho  | Er  | Tm  | Yb  | Lu  | Ac  | Th  | Pa  | U   | Np  | Pu  | Am  | Cm  | Bk  | Cf  | Es  | Fm  | Md  | No  | Lr  |
Processing Options for Recovery of U, Pu, Np

- **UREX**
  - Uses same solvent as PUREX (TBP in n-dodecane)
  - Recovers high purity U while leaving Pu with fission products
  - Recovers Tc in high yield

- **NPEX**
  - Uses same solvent as PUREX
  - Recovers mixed Pu and Np
  - Can generate a U/Pu/Np product in a predictable ratio
  - Requires UREX be run first to remove U from feed

- **Co-Extraction**
  - Uses same solvent as PUREX
  - Recovers U and Np/Pu or U/Pu/Np mixture
  - Likely requires multiple cycles to control U:Pu/Np ratio
  - Tc not targeted; recovery complicates processing
UREX Process for U and Tc Recovery

Fresh Solvent
30% TBP in n-dodecane

Feed
Dissolved Fuel

Scrub
Complexant
Dil. HNO₃

Strip
Dil. HNO₃

Extraction
Scrub
U/TC Strip

Raffinate
FP and all TRU

U/TC Product

Spent Solvent

Feed
U, Tc

Column Feed Adjust

Tc Sorption Column

Tc-loaded Resin

U Product
NPEX Process from 2007 UREX+ Demonstration

Fresh Solvent
30% TBP in n-dodecane

Feed
FPEX Raffinate After Feed Adjustment

Scrub
Dil HNO₃

Strip
Dil. UNO₃ Complexant

Extraction

Scrub

Strip

Raffinate
Remaining FP and MA

Product
U, Pu, Np

Spent Solvent
Co-Extraction Process from 2004 UREX+ Demonstration

- **Solvent**: 30% TBP in n-dodecane
- **Feed**: Dissolved Fuel Nitric Acid
- **Scrub**: Conc. Nitric Acid
- **Zr-Scrub**: Dil. Nitric Acid

**Extraction**

- **Raffinate**: Minor An., RE., Zr, Nitric Acid
- **Solvent**: 30% TBP in n-dodecane
- **Extraction Feed**: Nitric Acid
- **Loaded Solvent**: Pu, Np, U, Tc
- **Strip**: Complexant Conc. Nitric Acid
- **Scrub**: Dil. Nitric Acid

**U/Tc Re-Extraction**

- **U/Tc-Loaded Solvent**
- **Np/Pu Strip**: Nitric Acid
- **Np/Pu Product**: Nitric Acid

**U/Tc Strip**

- **U/Tc Product**: Dil. Nitric Acid
- **Clean Solvent**: (for Recycle)

**Uranium/Technetium Strip**

- **Strip**: Dil. Nitric Acid

**Final Products**

- **Dil. Nitric Acid**
Processing Options for Cs and Sr Recovery

**CCD-PEG**
- Solvent is chlorinated dicarbollide and polyethylene glycol in FS-13
- Recovers Cs and Sr in high yield
- Solvent requires a dense sulfone diluent
- PEG is partially soluble in aqueous phase
- Acidity of feed is low
  - Requires feed adjustment when run after NPEX, TRUEX, COEX
  - Degradation of complexants affects extraction
- Complexant required is used to strip solvent

**FPEX**
- Solvent contains a calixrene, crown ether, and modifier in Isopar-L®
- Isopar-L is a refined kerocene and similar to that of other UREX+ processes
- Derived from CSSX process being implemented at SRS
- Less-developed than CCD-PEG process: composition not yet finalized
- Cs and Sr D-values slightly lower than for CCD-PEG
- Acidity of feed is low
  - Requires feed adjustment when run after NPEX, TRUEX, COEX
  - Degradation of complexants affects extraction
CCD-PEG Process from 2006 UREX+ Demonstration

Recycle Solvent
CCDPEG
FS-13

Feed
UREX Raffinate
All TRU and FP

Scrub
HNO₃

Strip
Guan. Carbonate
DTPA

Wash
PEG-HNO₃

Extraction

Scrub

Strip

Wash

Raffinate
All TRU and FP/s

Product
Cs, Sr, Rb, Ba
FPEX Process from 2007 UREX+ Demonstration

Recycle Solvent
FPEX Solvent

Feed
UREX Raffinate
FP and all TRU

Scrub
Nitric Acid

Strip
Dilute Nitric Acid

Wash
Nitric Acid

Extraction

Raffinate
All TRU and FP
except gp I and II

Product
Cs, Sr, Ba, Rb

Wash

Topical Seminar Series on Nuclear Fuel Separation Processes
UREX+ Processing Options for An and Ln Recovery

- **TRUEX**
  - Solvent is CMPO and TBP in n-dodecane
  - Recovers rare earths and transuranics
  - Extensive database and development
  - Readily compatible with NPEX, COEX, UREX
  - Expensive extractant but n-dodecane diluent
  - May require multiple scrubs to achieve purity requirements

![CMPO](image)

**CMPO**

octyl(phenyl)-N,N-diisobutylcarboxymethyl phosphine oxide
TRUEX Process from 2005 UREX+1 Demonstration

**Feed**
UREX Raffinate
After feed adjust

**Fresh Solvent**
CMPO, TBP
n-dodecane

**Scrub 1**
Oxalic Acid
HNO₃

**Scrub 2**
Conc. HNO₃

**Scrub 3**
Dil. HNO₃

**Strip**
Lactic Acid
DTPA

**Extraction**

**Scrub 1**

**Scrub 2**

**Scrub 3**

**Strip**

**Raffinate**
Non-RE FPs

**Product**
Pu, Np, Am, RE
Lactic Acid
DTPA

**Spent Solvent**
Processing Options for An and Ln Separation

- **TALSPEAK**
  - HDEHP in n-dodecane
  - High separation factors and An recoveries have been demonstrated
  - Can be run for Am/Cm or Group TRU recovery (Pu/Np/Am/Cm)
  - Feed requires good pH control
  - Slow Extraction Kinetics
  - TRU product in complex

- **Cyanex 301® Extractant**
  - Cyanex 301 in n-dodecane
  - High An recoveries
  - Feed requires good pH control
  - Only partial An-Ln separation was observed
  - Poor solvent stability > limited recycle
    - *No longer under consideration*
TALSPEAK Flowsheet from 2006 UREX+ Demonstration

- **Fresh Solvent**: HDEHP in n-dodecane
- **Feed**: Am, Cm, and RE in Lactate Buffer w/ DTPA
- **Scrub**: Lactic Buffer w/ DTPA, Conc. HNO₃
- **Extract**: Extraction
- **Strip**: Strip
- **Raffinate**: Am, Cm
- **Product**: All RE
- **Spent Solvent**
Cyanex-301 Extractant Flowsheet from 2003 UREX+ Demonstration

- **Solvent**: Cyanex 301, TBP, n-dodecane
- **Feed**: TRUEX Product, pH = ~3
- **Scrub**: pH = ~3
- **Strip**: Complexant

**Extraction**
- **Raffinate**: REs

**Scrub**
- **Product**: Am, Cm

**Strip**
- **Spent Solvent**
## Potential Aqueous Processing Options That Have Been Demonstrated At Argonne

<table>
<thead>
<tr>
<th>Process</th>
<th>Product #1</th>
<th>Product #2</th>
<th>Product #3</th>
<th>Product #4</th>
<th>Product #5</th>
<th>Product #6</th>
<th>Product #7</th>
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<tbody>
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<td>UREX+1</td>
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<td>Cs/Sr</td>
<td>TRU/Ln</td>
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<td>Cs/Sr</td>
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<td>Cs/Sr</td>
<td>U/Pu/Np</td>
<td>Am</td>
<td>Cm</td>
<td>FP/Ln</td>
</tr>
</tbody>
</table>

Notes: (1) in all cases, iodine is removed as an off-gas from the dissolution process.

(2) processes are designed for the generation of no liquid high-level wastes

U: uranium (contributor to dose rate, and the mass and volume of high-level waste)

Tc: technetium (long-lived fission product, minor contributor to long-term dose)

Cs/Sr: cesium and strontium (primary short-term heat generators, affect waste form loading and repository drift loading)

TRU: transuranic elements: Pu - plutonium, Np - neptunium, Am - americium, Cm - curium (primary long-term dose rate contributors)

Ln: lanthanide (rare earth) fission products

FP: fission products other than cesium, strontium, technetium, iodine, and the lanthanides
Five Demonstrations of UREX+ Processes With Spent Fuel Have Been Completed Successfully

- **2003**: Complete UREX+3
  - 3 Legs of Co-extraction (UREX+2)
  - Dissolved Fuel
  - UREX
  - CCD-PEG
  - NPEX
  - TRU
  - Phosphinic Acid Separation
  - Ln

- **2004**: Complete UREX+3a
  - Dissolved Fuel
  - UREX
  - FPEX
  - NPEX
  - TRU
  - TALSPEAK
  - Am/Cm
  - Ln

- **2005**: 3 Legs of UREX+1a
  - Dissolved Fuel
  - UREX
  - CCD-PEG
  - TrueX
  - TALSPEAK
  - TRU
  - Ln

- **2006**: Complete UREX+1a
  - Dissolved Fuel
  - UREX
  - CCD-PEG
  - TrueX
  - TALSPEAK
  - TRU
  - Ln

- **2007**: Dissolved Fuel
  - UREX
  - Ion Exchange
  - Ln

March 25-26, 2008
The UREX+ Process Demonstrations

- **R&D for UREX+ process development includes:**
  - Process design and optimization using AMUSE
  - Equipment design
  - Process demonstration and post-test analysis

- **Objective of demonstration:**
  - Demonstrate that all desired spent-fuel constituents can be separated by aqueous processing
  - Show that product specifications for recycle or disposal are achievable

- **Flowsheets developed using AMUSE**
  - Test run with spent fuel in 2-cm contactor
  - All processes were run until steady state was achieved
ANL Centrifugal Contactor Bank in Shielded Cell
Spent Fuel Was Dissolved in Nitric Acid at Ambient Pressure and Under Moderately Higher Pressures

- Tests were run with both clad and declad fuel
- 0.5-1 kg fuel batches
  - Sealed and ambient pressure vessels have been used
  - Undissolved solids were generally minimal
- Solutions were stable
UREX+2 and UREX+3 Were First Two UREX+ Process Demonstrations

- **UREX+3**, as initially designed, differed significantly from more recent process flowsheets
  - U-Tc separation was accomplished by selectively stripping
  - The Cyanex process was used for the Am/Cm separation

- **Co-extraction of U, Pu, Np, and Tc was used in first demonstration of UREX+2 process**
  - Pu/Np separated from U/Tc by selective stripping of solvent
  - U and Tc separated by ion exchange

---

Dissolved Fuel

UREX

- U
- Tc

CCD - PEG

- Cs/Sr

NPEX

- Np/Pu

TRUEX

- FPs

Phosphinic Acid Separation

- Am/Cm

Ln

---

Dissolved Fuel

Co-Extraction

- Np/Pu

CCD - PEG

- Cs/Sr

Ion Exchange

- U
- Tc

Am/Cm/Other FPs
2003 UREX+3: Product Distributions

- All primary components report to desired product streams
  - Np and Tc fractions lower than targets
  - U and Tc separated by selective stripping rather than ion exchange
  - High rare earth content in Am/Cm product

**Distribution in process effluent streams (%)**

<table>
<thead>
<tr>
<th>Component</th>
<th>Product</th>
<th>Raffinate</th>
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<tbody>
<tr>
<td><strong>2003 UREX</strong></td>
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<tr>
<td>Tc</td>
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<td>4.60</td>
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<tr>
<td>U</td>
<td>99.96</td>
<td>0.02</td>
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<td><strong>2003 NPEX</strong></td>
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<tr>
<td>Pu</td>
<td>99.6</td>
<td>0.4</td>
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<tr>
<td>Np</td>
<td>70.3</td>
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<tr>
<td><strong>Cyanex 301</strong></td>
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<tr>
<td>RE</td>
<td>27.2</td>
<td>82.8</td>
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<tr>
<td>Am</td>
<td>99.98</td>
<td>0.02</td>
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</table>
UREX+2 Co-Extraction Process: Product Distributions

- All primary components report to desired product streams
  - Tc, Np fractions lower than targets
  - Zr in Np/Pu product rather than raffinate due to other-phase carryover because of mis-match between contactor design and loaded-solvent density

<table>
<thead>
<tr>
<th>Component</th>
<th>Product</th>
<th>Raffinate</th>
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<td></td>
<td><strong>2004 CCD-PEG</strong></td>
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<tr>
<td>Cs</td>
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<tr>
<td>Sr</td>
<td>&gt;98.6</td>
<td>&lt;1.4</td>
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</table>
Urex+1a Spent Fuel Demonstration Consisted of Four Extraction Processes and an Ion Exchange Process

- Produces a TRU product to serve as the feedstock for a fast reactor fuel
- Process was run in 2005 and 2006
  - CCD-PEG was omitted in 2005
- TALSPEAK process flowsheet was reworked in 2006 to account for slow process kinetics
UREX+1a: UREX Process Results

- Excellent hydraulic performance
- High recovery of all products
- Excellent TRU/Ln recovery obtained with TRUEX
- Refined TALSPEAK process for 2006 test met targets

### Distribution in process effluent streams (%)

<table>
<thead>
<tr>
<th>Component</th>
<th>UREX</th>
<th>CCD-PEG</th>
<th>TALSPEAK</th>
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<td><strong>Year</strong></td>
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<td>2006</td>
<td>2006</td>
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<tr>
<td><strong>Component</strong></td>
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<td><strong>Raffinate</strong></td>
<td><strong>Product</strong></td>
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<td>99.997</td>
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<td>Tc</td>
<td>97.1</td>
<td>2.9</td>
<td>95.5</td>
</tr>
<tr>
<td>Cs</td>
<td>NA*</td>
<td>NA</td>
<td>&gt;99.85%</td>
</tr>
<tr>
<td>Sr</td>
<td>NA</td>
<td>NA</td>
<td>99.9%</td>
</tr>
<tr>
<td>Pu</td>
<td>99.8</td>
<td>0.2</td>
<td>&lt;0.005</td>
</tr>
<tr>
<td>Am</td>
<td>0.001</td>
<td>99.999</td>
<td>&lt;0.03</td>
</tr>
<tr>
<td>Ln</td>
<td>88</td>
<td>12</td>
<td>&gt;99.97</td>
</tr>
</tbody>
</table>

NA: CCD-PEG not run as part of 2005 demonstration
Determine breakthrough capacity of IX resin

Experimental conditions
- Three column test
- Room temperature

![Graph showing Tc in effluent over column volumes](image)
ANL UREX+ Spent Fuel Demonstrations

- UREX achieved process targets
- Excellent TRU/RE recovery obtained with TRUEX
- Refined TALSPEAK process for 2006 test met targets
  - Fuel requirements – DF > 2000
  - Repository requirement – TRU loss < 0.01%
- Excellent hydraulic performance in all processes
In FY-2007 UREX+3a Was Demonstrated

- UREX+3a is being developed to produce a U/Pu/Np product
  - Am/Cm product for targets
- FPEX was run in place of CCD-PEG for Cs/Sr recovery
- In NPEX, U was added to yield a U/Np/Pu product
- Chemical analysis is to be completed in FY-2008

*CCD-PEG may be run as an alternate for FPEX
UREX+3a: Preliminary Results

- Elements report to desired product streams in all processes
- FPEX demonstrated high Cs and Sr recoveries
- Refined TRUEX process showed excellent recovery of An and Ln
- Analysis is incomplete, additional results are pending

**Distribution in process effluent streams (%)**

<table>
<thead>
<tr>
<th>Element</th>
<th>Raffinate</th>
<th>Product</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>2007 UREX</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>U</td>
<td>0.001%</td>
<td>99.999%</td>
</tr>
<tr>
<td><strong>FPEX</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cs</td>
<td>BD*</td>
<td>(100%)</td>
</tr>
<tr>
<td>Sr</td>
<td>&lt;0.1%</td>
<td>&gt;99.9%</td>
</tr>
<tr>
<td><strong>2007 NPEX</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu</td>
<td>0.07%</td>
<td>99.93%</td>
</tr>
<tr>
<td>Np</td>
<td>Bkgd*</td>
<td>(100%)</td>
</tr>
<tr>
<td><strong>2007 TRUEX</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Am</td>
<td>BD</td>
<td>(100%)</td>
</tr>
</tbody>
</table>

*BD = below detection limits; bkgd = at background levels
Effects of pH on Ln Distribution

- **D-value Dependence on pH**
  - Light lanthanide (La) has lowest power dependency
  - Heavier lanthanide (Gd) has higher dependency

- **D value dependency on pH for HDEHP-extraction alone is +3 order**
  \[ \text{Me}^{3+} + 3(\text{HL})_2 \rightleftharpoons \text{Me} (\text{L-HL})_3 + 3\text{H}^+ \]

- **D value dependency based on DTPA-complexation can be up to -5 order**
  - Depending on H$_5$DTPA acid equilibria at each pH
Performed a series of one-stage tests in a 2-cm centrifugal contactor.

As flow rate increases, the minimum is shifted from Nd to Sm-Eu.

Based on contactor tests, it appears that mixing is less intense in the contactor than in our batch vessel.

@ equilibrium

Flow rate increases
Distribution of lanthanides and TRU easily met process goals
- < 1% mass of actinide product was lanthanides
- ≤ 0.01% of TRU in Ln product

Effects of higher throughputs (shorter contacting time) were seen
- Effect of flow rate not as strong as suspected
- Actinide behavior was more consistent than that of lanthanides

### Distribution in process effluent streams (%)

<table>
<thead>
<tr>
<th>Feed Flow Rate</th>
<th>Rare Earth Product</th>
<th>TRU Product</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>RE, %</td>
<td>TRU, %</td>
</tr>
<tr>
<td>Low</td>
<td>99.9998</td>
<td>BD</td>
</tr>
<tr>
<td>Mid</td>
<td>99.7</td>
<td>BD</td>
</tr>
<tr>
<td>High</td>
<td>99.997</td>
<td>0.002</td>
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</tbody>
</table>

BD = Below Detection Limits
Lanthanide Behavior—Aqueous-Phase Stage Profiles

- Light lanthanides (La and Pr) do not show a kinetic effect
- The heavier lanthanide (Gd) does
  - As did Eu
Am vs. Eu Behavior--Stage Profile

- Effects at higher flow rates (less contact time) are seen for Eu aqueous-phase stage profile
  - Better extraction at lower flow rates
- However, even at highest flow rate, separation factors for Eu over Am are ~50 in extraction section.
  - ~100 in scrub section
Summary of UREX+ Process Demonstrations with Spent Nuclear Fuel

- All demonstrations have proven the feasibility of the UREX+ processes to achieve program goals
  - UREX has been repeatedly demonstrated to be viable for the selective extraction of U and Tc
  - Co-extraction of U, Pu, Np, and Tc with subsequent selective stripping of Pu/Np and U/Tc proved viable
  - Cs/Sr recovery was achieved using both CCD-PEG and FPEX
  - NPEX process recovers Pu and Np; produces U/Pu/Np product
  - TRUEX has been demonstrated in four tests for An/Ln recovery
  - TALSPEAK successfully separated actinides from lanthanides at high purities as required for fuel fabrication
    - Tests at different flow rates proved the robustness of the process for scale-up
Electrochemical Processing of Spent Nuclear Fuel

Dr. Michael Goff
Idaho National Laboratory

Nuclear Regulatory Commission Seminar
Rockville, MD
March 25, 2008
Also Know As

- Dry processing
- Pyroprocessing
- Pyrochemical processing
- Pyrometallurgical processing
- Electrometallurgical treatment
- Etc.
“Pyrometallurgical processing techniques involve several stages: volatilisation, liquid-liquid extraction using immiscible metal-metal phases or metal-salt phases, electrorefining in molten salt, fractional crystallisation, etc. They are generally based on the use of either fused (low-melting point) salts such as chlorides or fluorides (eg LiCl+KCl or LiF+CaF2) or fused metals such as cadmium, bismuth or aluminium.”

From Wikipedia
“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

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

- Construct Assembly and Test
- Clad Fuel, Leak Test & Bond Test
- Re-cast pins, separate VYCOR and inspect
- VYCOR scrap
- Fuel Cycle Facility
- Melt Refining Furnace (1400 °C)
- Scraps Fuel Alloy
- 235U to enrich
- Skull Oxidation
- U & Pu from oxide
- Crucible & Fission Products to Waste
- Reject Elements
- Disassemble & Remove Elements
- Remove Cladding & Chop Fuel
- Clean Assembly (Remove Sodium)
Chemical Bases for Electrochemical Processing

(Kcal per equivalent)

Metal Waste

Uranium Product

Ceramic Waste

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

Hg Mo W Ni H
Zn, Cr V Mn, Be

Fe Cd

Zr

Pu

Ca, Na Sr Li K Ba, Cs, Rb
Electrorefining
Cathode Processing

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

Driver and Blanket Fuel are Treated in Parallel

- Driver Element Chopper
- Mk-IV Electrorefiner
- Cathode Processor
- Casting Furnace
- Mk-V Electrorefiner
- Blanket Element Chopper
- Interim Storage

Driver Fuel → Element Segments → Cathode Products → Uranium Product → Uranium
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 GNEP.
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 electrefining 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.
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:
  - Hg
  - Mo
  - W
  - Ni
  - H
  - Zn, Cr
  - V
  - Mn, Be

- Uranium Product:
  - Np
  - Sc
  - Mg
  - Cm, Am
  - Nd
  - Ce
  - Pr, La

- Ceramic Waste:
  - 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**

3. **ZEOLITE MILL**
   - Milled Zeolite
   - Dried Zeolite

4. **MILL/CLASSIFIER**
   - Ground Salt

5. **HEATED V-MIXER**
   - Glass added after producing salt-loaded zeolite

6. **FURNACE**
   - Glass and Salt-Loaded Zeolite

7. **CERAMIC WASTE**
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 → ELECTROREFINER

SALT → CERAMIC WASTE

INDUCTION FURNACE

ZIRCONIUM → INGOTS
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 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.
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
INL hosted first International Pyroprocessing Research Conference in August.

There were approximately 75 attendees, 34 from outside US.

Papers were presented from 7 countries:
- US
- ROK
- Japan
- France
- United Kingdom
- India
- Russia

70 presentations were made.

Next conference will be in Korea.
Electrochemical fuel cycle can be used for recycle of fast reactor spent fuel and treatment of thermal reactor spent fuel to product a feed for fast reactors.

Electrochemical processing of spent metallic nuclear fuel has been successfully demonstrated at engineering scale.

Critical infrastructure exits domestically to support development and demonstration of technology.

Work activities are focused on research, development, and demonstration in support of GNEP.

Development and qualification of high-level wastes are integrated with overall process development.
GNEP Waste Form Technology Development

John D. Vienna, PhD
Pacific Northwest National Laboratory, Richland, WA

DOE/NRC Seminar No. 1 in Topical Series on Advanced Waste Management and Waste Forms

April, 2008
Outline

- Aqueous Waste Streams and Waste Forms
- Electrochemical Waste Streams and Waste Forms
- Technology Readiness Assessment
- Testing Approaches/Waste Form Qualification
Aqueous Process & Waste Streams

Chopping
  → Cladding: Zircaloy
  → Hardware: SS

Volox
  → Gases: I, HTO, Kr, Xe, CO₂

Dissolution
  → UDS: Pd, Ru, Rh, Mo, Tc, Zr, O

UREX

Ion Exchange
  → Tc
  → U

FPEX
  → Cs/Sr: Cs, Sr, Ba, Rb

TRUEX
  → TMFP: Fe, S, Ru, Pd, Rh, Mo, Zr

TALSPEAK
  → LNFP: Ce, Ln, Pr, Nd, Y
  → TRU: Pu, Am, Cm, Np

LLW: Solvents
PPE
RBT
lab returns
off-gas…
Compacted Metals

**Hulls**
- <1% fuel after voloxidation and filter
- leaching removes all measurable residual fuel meat
- likely to be >100nCi TRU/g due to activation of U impurities in Zircaloy → GTCC

**Hull/hardware compaction**
- proven technology employed worldwide
- potential for encapsulation in metal matrix

Zircaloy and SS Scrap from Cogema Plant, LaHauge from: Management of Cladding Hulls and Fuel Hardware, Tech report 258, IAEA, Vienna, 1985
Aqueous Primary Off-gas Streams

Chopper → Voloxidizer → Dissolver

HEPA → Ru Trap → NOx Absorber

HTO → I Bed → CO₂ Scrub → Drier

In-can grouting → Dispose - SLB

Catalytic oxidizer
MS (3A) capture
Release from MS Condense
Mix in cement
Ag-Z capture
Remove bed
Encapsulate
Package
Dispose - Rep

NaOH scrub
Remove
Mix in cement
Dispose - SLB

Cryogenic Ag-Z
Release from Ag-Z
Combine w/ Kr
or Release

Cryogenic HZ
Release from HZ
Combine w/ Xe
Pressurize
Bottle
Decay storage

Remove
Leach
Dispose - SLB

VOG
Iodine on Ag-Z

- Mordenite selected as baseline getter/waste form
  - demonstrated at pilot scale in US
  - adopted for use in Hanford WTP
  - high DF and loading as getter
  - nitrate tolerant
  - base of a good waste form

- Likely to be encapsulated
  - to form monolith
  - improve leach resistance
  - geo-polymer at low temperature (<200°C)
  - low temperature glass (<500°C)
Aqueous Tc and UDS

Filter UDS solids, blend with other metal streams

Dissolver
Clarifier

UREX

TRU/FP

Tc IX

Reduce to metal

$UO_2(NO_3)_2 \rightarrow U_3O_8$ or $UO_3$

steam reform precipitated $[NBu_4]TcO_4$ w/ C $\sim 800^\circ C$

Blend

Potential additives: Zr (hulls), Fe (SS), UDS, reduced TMFP; mix in crucible (e.g. coated C)

Melt

furnace under Ar, $N_2$ or vacuum $\leq 1600^\circ C$

Package

remove crucible from furnace, load into canister, decon canister, seal

Dispose

repository
Metal Waste Form

- Based on Zr-Fe alloys
  - solubility of Tc, PGM, Mo, and other impurities in durable phases
  - corrosion rate similar to alloy 22 and SS316

Testing to Evaluate the Suitability of Waste Forms Developed for Electrometallurgically-treated Spent Sodium-bonded Nuclear Fuel for Disposal in the Yucca Mountain Repository, ANL-05/43, Argonne National Laboratory, 2005
Metal Waste Form Corrosion
Aqueous Cs/Sr

- **FPEX**
- **Strip**
- **Evaporate**
- **Blend w/ GFCs**
- **Feed Melter**
  - **Cast**
  - **Package**
  - **Dispose**

**Process Details**:
- Silicates or frit
- CCIM, 1350°C, recycle scrub
- Disposal container, 6 in
- Seal, decon
- Decay store, repository

April 23, 2008

NRC - GNEP Waste Management Discussion
Cs/Sr Glass

- Based on well developed and demonstrated technology
- Alumino-silicate glasses and glass ceramics
  - Fe$_2$O$_3$ or SnO$_2$ used for charge compensation, e.g.:
    \[
    \text{SrO} + \text{Fe}_2\text{O}_3 \xrightarrow{\beta\beta} \text{ZrO}_2 + 2\text{FeO}
    \]
    \[
    \text{Cs}_2\text{O} + \text{Fe}_2\text{O}_3 \xrightarrow{\beta\gamma} 2\text{BaO} + 2\text{FeO}
    \]
  - 30 to 64 wt% waste loading
  - density between 3 and 4 g/mL

![Graph showing density vs. waste loading with predicted and measured values.](graph.png)

Samples Contain Bubbles

Waste Loading (wt%) vs. Density (g/mL)
Cs/Sr Glass/Glass Ceramic

- PCT responses well below EA glass
- Between 0 and 52% crystals* (primarily pollucite, celsian, and other alumino-silicates)
- TCLP responses as a function of waste loading pending

* a roughly 80% crystal melt was made, but, wouldn’t pour
Aqueous Transition Metal Fission Products

TRUEX Raffinate

- Reduce
  - Electroreduction
    - $Tc, Fe, Zr, UDS$; in crucible
  - Blend
    - Inert or reducing cover, $\leq 1600^\circ C$
  - Melt
    - Remove crucible, load canister, decon canister, seal
  - Package
  - Dispose
    - Repository
Transition Metal Fission Products Metal

- Contain significant concentrations of platinum group metals (PGM) not soluble in glass
- Alloy properties the same as earlier described metal
- Adds little to volume of metal waste form volume, but, would increase glass volume by ~3×
- Requires reduction from nitrate solution → electrochemical reduction is being investigated
Aqueous Lanthanide Fission Products Glass

TALSPEAK Product

Blend

Feed Melter

Cast

Package

Dispose

Glass forming chemicals, (potentially Cs/Sr and/or TMFP)

HWIM, 1350°C (or CCIM if w/ Cs/Sr and/or TMFP)

Disposal container

Seal, decon

Repository
Lanthanide Glass

- Based on well developed and demonstrated technology
  - developed for SRS Am/Cm treatment
  - chosen and demonstrated for PIP
- Loadings at 60 wt% (oxide basis)
- Outstanding durability

Property Data for Simulated Americium/Curium Glasses, PNNL-13009, Pacific Northwest National Laboratory, 1999
Electrochemical Process & Waste Streams

- **Chopping**
  - Gases: Kr/Xe, HTO

- **Electro-refining**
  - U
  - UDS: SS, Zr, Pd, Ru, Rh, Mo, Tc

- **U/TRU Recovery**
  - U/TRU

- **LN Recovery**
  - Cs/Sr: Cs, Sr, Ba, Rb, I
  - LNFP: Ce, Ln, Pr, Nd, Y

**LLW:** Salts, PPE, RBT lab returns off-gas...
Electro-Chemical Metallic Wastes

Electro-Refiner

- Drain
  - Remove anode basket from electrorefiner and drain salt
- Distill
  - Transfer to crucible furnace and heat to distill residual salt
- Additives
  - Add trim chemicals needed for desired alloy composition
- Melt
  - Heat to ~1600 °C under reducing conditions
- Package
  - Load ingot into disposal canister, seal, decontaminate
- Dispose
  - Repository
EChem Metal Waste Form

- Developed for EBR-II fuel and blankets
- Demonstrated at full scale
- Same SS-15 Zr discussed for aqueous wastes
  - high durability
  - qualification testing well underway
Electro-Chemical Fission Products

Remove salt from electrorefiner
Recover lanthanides from salt as oxides

Occlude
Mix salt with zeolite ~500°C to occlude salt

Blend salt-loaded zeolite with binder glass

Load mixture into container, w/ weight

Heat in container to liquify binder glass and react salt and zeolite

Load container into disposal canister, seal, decon, repository

Remove Salt
Blend
Melt
Cast
Dispose

Glass forming chemicals

HWIM, 1350°C

Disposal container

Seal, decon, repository
Glass Bonded Sodalite

- Developed for EBR-II fuel/blankets

\[ \text{Na}_{12}(\text{AlSiO}_4)_{12} + 4\text{NaCl} \rightarrow 2\text{Na}_8(\text{AlSiO}_4)_6\text{Cl}_{12} \]

- Demonstrated at full scale
- Durability determined by glass corrosion rate → HLW rate-law used
Waste Form Testing

- Waste form testing performed to meet three objectives
  - model/predict long-term degradation
  - demonstrate qualification
  - define acceptable operating range

- The tests best suited to meet these objectives are in various states of development, acceptance, and use
  - appropriate test for more mature waste forms (e.g., HLW glass) are readily available
  - testing protocol for less common nuclear waste forms (e.g., encapsulated zeolite with AgI) have not been as well established

- Panel evaluated the test methods appropriate to meet each of the objectives
Testing Protocols for GNEP Waste Forms

- Test method selection and testing protocols based on consensus standards (ASTM, ANS, …)
- Panel recommended initial testing for each baseline waste form for each disposal condition
- More complete list for glass and ceramics
- Acceptable test responses depends on a host of assumptions
- General repository release models being developed to aid in determining acceptable test responses for various environments

Technology Readiness Assessment (TRA)

- Evaluating a technology (waste process/form) for readiness to implement/produce
- Concept from NASA, adopted by DOD, now being adopted by DOE
- Generates technology readiness level (TRL, 1-9) and technology maturation plan (TMP)

<table>
<thead>
<tr>
<th>Stage</th>
<th>Technology Development</th>
<th>Demo</th>
<th>Comm</th>
<th>Operations</th>
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<td>Bench → Pilot → Full</td>
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<td>Prototypes → Plant</td>
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<tr>
<td>TRL</td>
<td>1 → 2 → 3 → 4 → 5 → 6 → 7 → 8 → 9</td>
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</table>
Technology Readiness Assessment

- Screening technology readiness assessment performed for each of the key waste streams
- Certain technology elements for each waste form rank low on the readiness scale
  - Generally resulting in low TRL for all waste forms
  - Misleading to take TRL out of context
  - Qualitative judgments were made based on EM TRA process
- Screening TRA being reviewed by an independent expert panel and is being updated before issue
- Complete TRA to be performed after resolution of some waste form baseline issues
## Technology Readiness Assessment

<table>
<thead>
<tr>
<th>Stream</th>
<th>Description</th>
<th>Process/Form</th>
<th>Maturity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hulls (LWR)</td>
<td>Zirc-alloy</td>
<td>Leach, compact</td>
<td>H</td>
</tr>
<tr>
<td>$^3$H (both)</td>
<td>HTO</td>
<td>Cement</td>
<td>H</td>
</tr>
<tr>
<td>I (Aq)</td>
<td>AgZ</td>
<td>Encapsulated</td>
<td>L</td>
</tr>
<tr>
<td>Kr (both)</td>
<td>AgZ</td>
<td>Compressed gas</td>
<td>M</td>
</tr>
<tr>
<td>$^{14}$C (Aq)</td>
<td>CO$_3$ in NaOH soln</td>
<td>Cement</td>
<td>H</td>
</tr>
<tr>
<td>UDS (both)</td>
<td>PGM, Mo, Tc, Zr, (O in Aq, SS in Echem)</td>
<td>Metal alloy</td>
<td>L</td>
</tr>
<tr>
<td>$^{99}$Tc (Aq)</td>
<td>IX eluate</td>
<td>Metal alloy</td>
<td>M</td>
</tr>
<tr>
<td>Cs/Sr/Ba/Rb (Aq)</td>
<td>HNO$_3$ soln</td>
<td>Aluminosilicate ceramic or glass</td>
<td>M</td>
</tr>
<tr>
<td>LNFP (both)</td>
<td>LN in HNO$_3$ soln (Aq) LN$_2$O$_3$ (Echem)</td>
<td>Glass</td>
<td>H</td>
</tr>
<tr>
<td>TMFP (Aq)</td>
<td>Fe, S, PGM, Zr, Mo in HNO$_3$ soln</td>
<td>Metal alloy</td>
<td>L</td>
</tr>
<tr>
<td>FP (Echem)</td>
<td>Cs, Sr, Ba, Cl, K, Li</td>
<td>Glass bonded sodalite</td>
<td>M</td>
</tr>
</tbody>
</table>
Waste Form Strategy Evolution

- Critical evaluation of waste management baseline currently underway

- New baseline will strive to
  - minimize complexity and number of waste processes
  - minimize amounts of various waste types
  - use nature to guide \( \rightarrow \) match waste form to waste and disposal chemistries

- Key unknowns/uncertainties
  - repository type (Yucca Mtn currently limited to 70,000 MTHM)
  - waste type classification (NWPA based on PUREX)
  - requirements for capture of fission product gases
  - GTCC disposal
# Treatment of Aqueous Wastes

<table>
<thead>
<tr>
<th>Substance</th>
<th>Treatment</th>
<th>Disposal Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hulls</td>
<td>Compact</td>
<td>Metal for GTCC or Repository Disposal</td>
</tr>
<tr>
<td>Hardware</td>
<td></td>
<td></td>
</tr>
<tr>
<td>HTO</td>
<td>Capture Cement</td>
<td>Cement for LLW Disposal</td>
</tr>
<tr>
<td>$^{14}\text{CO}_2$</td>
<td>Capture Encapsulate</td>
<td>Zeolite in Geo-polymer for Repository Disposal</td>
</tr>
<tr>
<td>Iodine</td>
<td></td>
<td></td>
</tr>
<tr>
<td>UDS</td>
<td>Reduce Melt</td>
<td>UDS, Tc, TMFP Alloy for Repository Disposal</td>
</tr>
<tr>
<td>Tc</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TMFP</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cs/Sr</td>
<td>Vitrify Decay store</td>
<td>Cs/Sr, LN Glass for Repository Disposal</td>
</tr>
<tr>
<td>LN</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Treatment of Electrochemical Wastes

- **HTO**: Capture Cement → Cement for LLW Disposal
- **UDS**: Remove salt Melt → Alloy for Repository Disposal
- **LNFP**: Separate Remove salt Vitrify → High Ln Glass for Repository Disposal
- **Cs/Sr OFP**: Remove Cl Immobilize → Glass or Ceramic for Repository Disposal
Conclusions

- **GNEP waste treatment is on sound foundation**
  - waste forms selected to match waste chemistry and disposal environments → nature guides selection
  - many waste processes are based on mature technologies
  - no technical *show stoppers* identified

- **Waste form formulation and testing is well underway**
  - preliminary testing on all key waste forms
  - test methods being developed to meet three testing criteria
  - program in place to fill technology and qualification gaps

- **Waste form campaign is evolving**
  - reduce complexity and amount
  - take advantage of unique waste characteristics
  - focus efforts on increasing technology readiness

Dirk Gombert, Consulting Engineer
Idaho National Laboratory

DOE/NRC Seminar No. 1 in Topical Series on Advanced Waste Management and Waste Forms

April, 2008
Presentation Outline

- **Waste Treatment Overview**
  - Waste Streams
  - Evaluation Criteria
  - Recommended Waste Form Baseline
  - R&D and Trade Studies

- **Integrating Waste Management**
  - Integrating the fuel cycle
  - New concepts for risk based regulations & collaboration
  - Details of a strategy for managing all radioactive wastes

- **Concluding Points**
GNEP Overview

- Reduce America’s dependence on fossil fuels
- Provide abundant energy without generating carbon emissions or greenhouse gases
- Recycle used nuclear fuel to minimize waste and curtail proliferation concerns
- Safely and securely allow developing nations to deploy nuclear power to meet energy needs
- Assure maximum energy recovery from still-valuable used nuclear fuel
- Reduce the number of required U.S. geologic waste repositories needed for the remainder of this century
Closing the Fuel Cycle

[Diagram showing the fuel cycle process, including steps such as fabrication, light water reactor, enrichment, transmutation fuel fabrication, advanced burner reactor, storage, and waste disposal or recycling.]
Significantly reduce the long-lived nuclear waste to be disposed of in a geologic repository making disposal less complex and minimizing the need for additional repositories.

- Repository needed in all cases
- One repository may meet U.S. needs this century with GNEP
Treatment Baseline Report Scope

Technical Recommendations to Stabilize:

- UDS
- Tc
- Metals – hulls/hardware
- Fission products
- Cs/Sr
- Lanthanides
- Gaseous FP – I, H, C, Kr, Xe

Metal Waste Form
Decay Storage Waste Form
Oxide Waste Form (possibly combining Cs/Sr with Ln)
Specialized Waste Forms
Evaluation Criteria

- **Commercial Practicability (weighted 40%)**
  - Technical Practicality, Flexibility, Scalability, Waste Loading, Processing Cost, Secondary Waste Generation

- **Technical Maturity (weighted 10%)**
  - Development Cost/Schedule to Implement,

- **Waste Form Performance (weighted 40%)**
  - Thermodynamic/Rad/Mech/Thermal Stability, Chemical Durability

- **Stream Specific Criteria:**
  - Heat transfer, RedOx Sensitivity, Resistance to degradation by decay, Resistance to radiolysis, Ability to treat more than one waste, Experience with disposal of similar materials

- **Stakeholder Acceptance (weighted 10%)**
  - All processes designed to meet standards
### Treatment Baseline Report Recommendations

<table>
<thead>
<tr>
<th>Waste Stream</th>
<th>UREX+</th>
<th>Echem</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tc</td>
<td>Metal Alloy, possibly containing UDS and transition metal FP.</td>
<td>Metal Alloy containing UDS and transition metal FP.</td>
</tr>
<tr>
<td>Cs/Sr</td>
<td>Glass or Ceramic</td>
<td>Glass-bonded sodalite. May contain $^{129}\text{I}$ carried over from electro-reduction.</td>
</tr>
<tr>
<td>Ln</td>
<td>Glass, may be combined with Cs/Sr</td>
<td>Borosilicate glass or glass-bonded sodalite if combined with Cs/Sr.</td>
</tr>
<tr>
<td>FP</td>
<td>Included in metal waste form with Tc described above or Ln glass.</td>
<td>Included in metal waste form with Tc described above.</td>
</tr>
<tr>
<td>UDS</td>
<td>Included in metal waste form with Tc described above.</td>
<td>Included in metal waste form with Tc described above.</td>
</tr>
<tr>
<td>Metals—Cladding &amp; Hardware</td>
<td>Compacted metal. Metal ingot if cost effective or required for disposal.</td>
<td>Compacted metal. Metal ingot if cost effective or required for disposal.</td>
</tr>
<tr>
<td>Waste Stream</td>
<td>Waste Form(s)</td>
<td>UREX+</td>
</tr>
<tr>
<td>--------------</td>
<td>---------------</td>
<td>-------</td>
</tr>
<tr>
<td>Tritium</td>
<td>Grouted tritiated water (HTO).</td>
<td>Grouted tritiated water (HTO).</td>
</tr>
<tr>
<td>Iodine</td>
<td>Encapsulated silver zeolite.</td>
<td>Glass-bonded sodalite w/Cs/Sr and/or lanthanide FP.</td>
</tr>
<tr>
<td>Carbon-14</td>
<td>Grouted Na₂/CaCO₃ potentially including the tritiated waster.</td>
<td>Grouted Na₂/CaCO₃ with residual in glass-bonded sodalite w/Cs/Sr and/or lanthanide FP.</td>
</tr>
<tr>
<td>Kr</td>
<td>Decayed in fuel then released. Pressurized gas cylinder w/wo Xe if fuel storage is insufficient.</td>
<td>Decayed in fuel then released. Pressurized gas cylinder w/wo Xe if fuel storage is insufficient.</td>
</tr>
</tbody>
</table>
Chemically reduce and effectively capture the soluble Tc and transition metal elements from the expected aqueous solutions

Optimize alloy formulations for waste loading to immobilize Zr, Mo, Ru, Rh, Pd, and Tc

Evaluate alternative reductants to ferrous sulfamate in separations

Quantify capacity of alloys to accommodate non-metallic feed materials including Zr(MoO4)2 and TcO2

Develop a mechanistically-based model for alloy degradation
R&D Tasks - Cs/Sr (Decay Storage)

- Determine effects of transmutation and fate of the stable decay daughters.
- Optimize glass and ceramic formulations for waste loading, considering the effects of radiolysis, transmutation, thermal output, and durability during decay storage.
- Develop process for removal of Cs and Sr from Echem salts
- Develop and demonstrate flowsheets for Cs/Sr immobilization
- Evaluate benefits of heat management concepts including incorporating waste form in a metal matrix, internal or external fins, wet vs. dry storage, etc.
- Evaluate disposal options following decay storage
- Consider combination with other streams into common form
R&D Tasks - Lanthanides/FP (Oxide form)

- Optimize glass/ceramic formulations to immobilize Fe, Ln, Zr, Mo, Ru, Rh, and Pd for combined Ln/FP streams
- Optimize Ln-glass formulations
- Define operating envelope for processing glasses/ceramics in induction melter (CCIM for Ln/FP and HWIM for Ln)
- Evaluate alternative reductants to ferrous sulfamate in separations
- Develop glass/ceramic formulations for waste loading of Cs/Sr/Ln elements for processing in the CCIM
- Develop process for removal of Ln/FP from Echem salts and optimize waste form
R&D Tasks - Volatile radionuclides

- Characterize selectivity of radionuclides in gas treatment steps
- Evaluate disposal of $^{14}$C and HTO as LLW
- Select baseline I capture and immobilization materials and processes
- Evaluate long-term retention of iodine on waste form
- Evaluate need for Kr capture
- Develop non-cryogenic Xe/Kr separation methods if required
- Determine tritium DF on 3A molecular sieve from very low dew point gas streams
- Develop $^{14}$C waste forms for GTCC disposal
R&D Tasks - Activated Metals

- Characterize activation of hardware components to evaluate co-disposal versus segregation of hardware and TRU contaminated cladding.
- Define conditions required to melt the collected hardware, including the needed additives, melting temperature
R&D Tasks - General Studies

- Evaluate all waste/byproducts for beneficial reuse.
- Characterize recommended waste forms to support eventual acceptance to planned or conceptual disposal facilities.
- Evaluate potential for cross-contamination in waste streams and potential ramifications to disposition strategy.
- Determine the optimal waste stream combinations for co-immobilization and disposal.
- Perform trade studies for LLW treatment to reduce volume, recover radionuclides, and dispose.
- Develop detailed mass balances to show radionuclide inventories in various waste streams and amounts and categories of LLW.
- Collect data to qualify waste forms for disposal.
Waste Treatment Baseline Concluding Points

- GNEP Separations enable consideration of more efficient waste management strategy
- May make “HLW” classification obsolete
- Waste chemistry matched to waste form and environment to maximize durability and minimize mobility
- Many waste forms still conceptual
- Significant work needed to define characterization needs
- More work need to make waste forms and perform characterization
**GNEP Integrated Waste Management Strategy**

- **IWMS proposes a simpler system to dispose of all radioactive wastes**
  - Builds on a fuel cycle using advanced separations
  - Assumes 70,000 MTHM limit is lifted, or strategy applies to second repository
  - Suggests need for NWPA modification to allow risk-based waste management similar to provisions of 10 CFR 61 with added radionuclides
  - Could be applied to DOE, NRC, space, defense, medical, and diagnostic wastes

- **Emphasizes recycle and reuse, but based on economic recovery evaluation factoring in value of material and cost avoidance of disposal**

- **Considers need for industry to have a reliable system to routinely transport nuclear materials and dispose wastes**

- **Integrates current EM GTCC EIS effort**

- **Based on a collaborative approach among DOE-NE/EM/RW and NRC/EPA**
Nuclear Fuel Cycle Integrating Waste Management

Diagram:
- Fuel Fabrication
  - Isotopes
  - Safeguards
  - Schedule
  - Shipping

- Reactor Design/Operation

- Irradiated fuel storage
  - Schedule
  - Burnup
  - Conversion Ratio
  - Shipping

- Fuel Processing
  - Inventory Control
  - Product Purity
  - Safeguards
  - Separation Efficiency
  - Disposition Path
  - Shipping

- Material Recycle/Disposition
  - WAC
  - Waste Form Technology
  - Shipping
Separations and Stabilization by Chemical Family

Thermal Neutron Fission of U-235

La Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
Ac Th Pa U Np Pu Am Cm Bk Cf Es Fm Md No Lr

April, 2008
DOE/NRC Seminar No. 1 in Topical Series on
Advanced Waste Management and Waste Forms
Consider useful materials and radionuclides for recycle/reuse
- Fissile isotopes
- Commercial use of radionuclides

Expand technical bases for additional waste forms

Designation of a routine disposal pathway for GTCC not requiring a case-by-case performance assessment.
- >100 nCi TRU/g
- Activated metals
- Short and long half-life radionuclides

Consideration of the concept of “decay storage” in secure storage facilities to reduce heat impacts of radionuclides such as $^{137}$Cs, $^{90}$Sr, $^3$H, and $^{85}$Kr to decay to LLW/release limits.
Recycle/Reuse of Byproducts/Wastes

- **Similar to Rocky Flats Economic Discard Limit for Pu**
  - Considered value of uranium
  - Considered reactor time
  - Did NOT consider waste management

- **Economic Recovery Limit**
  - Value as fuel or industrial application
  - Cost of recovery/purification
  - Reduction to repository source term (inventory/duration) and heat
  - Fuel/target fabrication/irradiation/recycle costs
  - Remote vs. glovebox operations
  - Secondary waste generation
  - Potential for diversion and accidents
GTCC > 100 nCi TRU/g

- Source of materials
  - Contaminated cladding
  - Separations losses
  - Fuel fabrication losses
  - HVAC & process filters, maintenance, job-control, and balance-of-plant

- Value of reconsidering contamination limit

- WIPP precedent for defense wastes

- Japanese definition of TRU is long-lived, low heat
  - Actinides
  - Technetium
  - Iodine

- Pending EIS IWMS assumes WIPP precedent for defense-TRU
Activated Metals

- A Primary Category in the DOE-EM GTCC EIS
- GNEP sources of materials
  - Fuel cladding
  - Non-fuel-bearing hardware
  - Pressure vessels (other D&D metals)
- EIS options
  - Enhanced Surface Disposal
  - Borehole
  - Geologic Repository
- Pending EIS IWMS
Engineered Storage for Decay

- **Potential Radionuclies**
  - $^{137}\text{Cs}$
  - $^{90}\text{Sr}$
  - $^{85}\text{Kr}$
  - $^{3}\text{H}$
  - xxxCm

- **Heat Management Strategy Trade-Studies of Various Concepts**
  - Extended SNF aging
  - Interim waste form storage for up to 10 half-lives
  - Storage at reactor, reprocessing plant, repository, interim facility
  - Transportation, handling, processing, security, permitting issues
  - Facility M&O costs
  - Ultimate disposition of materials (cross-contamination, specific isotopes)
Integrated Waste Management Strategy Logic Diagram

- Reprocessing or Direct Disposal
  - Managed Storage
    - Fuel or Targets
      - Reactor
        - Heat
  - Managed Storage
    - Fuel or Targets
      - Reactor
        - Heat

- Systems Analyses

- Reuse Recycle
  - Waste

- Fissile or Fertile?
  - Yes
  - Economic Recovery?
    - Yes
      - Stage for Use
      - GTCC?
        - Yes
          - Repository
          - 100nCi TRU/g
            - >
              - Disposal per EIS
            - ≤
              - Activated Metal?
                - Yes
                  - Disposal per EIS
                - No
                  - Managed Storage?
                    - Yes
                      - Disposal Pending Decay
                      - Decayed GTCC
                      - Not Cost-Effective
                        - Disposal per EIS
                    - No
                      - Transmute?
                        - Yes
                          - Not Cost-Effective
                        - No
                          - Disposal per EIS

- Industrial Use?
  - Yes
  - Economic Recovery?
    - Yes
      - GTCC?
        - Yes
          - Repository
        - No
          - Class A/B/C Disposal Facility
          - Decayed ≤ Class C
          - Heat
          - Not Cost-Effective
            - Disposal per EIS
Transmutation of Long-lived Radionuclides

- Potential Radionuclides
  - $^{99}$Tc
  - $^{129}$I
  - $^{xxx}$Am
  - $^{xxx}$Cm

- Destruction in fuel or targets
  - Remote fabrication
  - Irradiation costs
  - Multiple recycles
  - Waste generation
Volume Effects in Waste Management

- **Repository capacity limited by legislation, dose, heat, or volume**
  - NWPA limits first repository/MRS until second repository is in operation
  - Potential dose is ultimate functional design criteria
  - Operational design criteria has thermal limits to meet dose requirement
  - Volume is generally not a limiting factor for repository capacity
  - Waste volume affects transportation and placement operations

- **Canistered (TAD) SNF ~ 1.24-1.51 m³/MTHM**

- **Areva reports ~ 0.31 m³ HLW+ILW/MTHM for 40 GWd/MTHM**
  - ~0.13 m³ glass/MTHM (Vitrified HLW MA/FP/UDS)
  - ~0.18 m³ metal/MTHM (Supercompacted ILW hulls and hardware)

- **Rokassho is similar to La Hague**
Comparison of WM volumes w/wo Reprocessing

- **IWMS suggests classification by risk using Class A/B/C, GTCC**
  - 0.013-0.060 m³ glass/MTHM (Cs/Sr/Ln w/wo TMFP from 50 GWhd/MTHM)
  - 0.007-0.004 m³ metal/MTHM (Tc/UDS w/wo TMFP varies inversely with glass)
  - 0.020-0.064 m³ total repository waste not including hulls/hardware or offgas wastes (iodine, ¹⁴C)
  - 0.047-0.072 m³ Hulls & Hardware
  - 0.002-0.009 m³ Iodine concept (order-of-magnitude)
  - 0.019-0.038 m³ ¹⁴C concept (order-of-magnitude)
  - 0.088-0.183 m³ ΣGTCC

- **Volume reduction ~2-6.5 glass + metal forms vs COEX HLW glass**
- **Volume reduction ~8-14 ΣGTCC vs. canistered SNF**

- Primary benefit is not volume reduction, rather more durable waste forms and a strategy to increase repository capacity
Secondary Wastes

- Job Control (maintenance, decontamination, PPE)
- Balance of Plant (laboratories, waste water, utilities, filters, etc.)
- GTCC estimated at ~ 5-20xHLW
- LLW Class A/B/C estimated at ~3-5xGTCC
- Economics of volume reduction
  - Limited waste disposal options
  - Industry has essentially eliminated MLLW generation
  - Waste minimization programs
  - Compaction, steam-reforming, and possibly incineration if cost-effective

DOE has taken responsibility for GTCC similar to SNF
  - Limited commercial cost incentive for GTCC waste disposal
IWMS Conclusion

- IWMS describes simplified process for managing all radioactive wastes based on risk
- Repository wastes grouped chemically with waste form matched to wastes
  - Maximizes durability
  - Minimizes volume
- Identifies trade studies to evaluate cost effectiveness of options
- Provides basis for collaboration among DOE-NE/EM/RW and NRC/EPA
- Independent of repository environment, thermal vs fast reactor recycling, fuel or target type, allows phased approach to separations including PUREX, COEX, UREX+, and Echem
Thank you for your kind attention.

Questions?

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208-526-4624
The Global Nuclear Energy Partnership (GNEP)

Overview of Systems Analysis and Waste Form Campaigns

Kathryn McCarthy
GNEP Systems Analysis Campaign Director
Mark Peters
GNEP Waste Form Campaign Director

April 23, 2008

Presentation to Nuclear Regulatory Commission
Rockville, MD
Outline

- GNEP/AFCI Mission and Requirements
- Overview of Systems Analysis Campaign
- Overview of Waste Form Campaign
- Questions and Discussion
GNEP/AFCI Mission Statement

Conduct a nuclear energy research, development, and demonstration program focused on nuclear fuel recycling and waste management to provide the infrastructure, technologies, and qualified personnel needed to enable the United States to close the nuclear fuel cycle and to support the safe, secure, economic, and sustainable global expansion of nuclear energy in a way that reinforces United States technological leadership.
GNEP/AFCI Program Requirements

Develop and demonstrate technologies to:

1. **Reduce the environmental and financial burden and uncertainty associated with long-term nuclear waste management.**
2. **Optimize nuclear waste management by:**
   - Minimizing the risk of waste that needs to be handled or stored.
   - Producing only solid wastes in robust waste forms.
   - Recycling and reusing materials to the maximum extent possible.
3. Recover the unused energy value of materials separated from spent nuclear fuel for future use.
4. Enhance the ability to safeguard the advanced fuel cycle facilities by developing technologies to more accurately account for, track, and manage nuclear materials.
5. **Support the near-term deployment of fuel cycle technologies (20 years) as well as define longer term deployments of next-generation technologies (50 years).**
6. **Make the closed fuel cycle as economical as possible.**
7. Accelerate deployment of nuclear reactors that are appropriate for regions with limited capacity electricity grids.
8. Permit deployed facilities to be licensable by the appropriate civil regulatory authorities.
9. Develop the research and development infrastructure needed to demonstrate closure of the fuel cycle at an engineering-scale in the United States.

10. Support the development of programs and processes that will encourage the education and training of the personnel that will be needed to staff industry, regulators, government, universities and national laboratories to allow closure of the fuel cycle in the United States and reinforce United States technological leadership in the world.
Mission: Carry out integrating analyses for GNEP development and infrastructure deployment, and enable a requirements-driven process for all technical activities

Objectives:
- Integrates information from the diverse technology development and R&D efforts
- Enables examination of a diverse set of scenarios
  - Evaluate off-ramps
  - Evaluate technology alternatives
  - Examine deployment options
  - Understand dynamics

Assumptions
- Focus on 1-tier and 2-tier relative to once-through
- All transuranics are recycled
Systems Analysis Increases Understanding of the Fuel Cycle

- Analyzes the entire fuel cycle to understand trends and relative relationships
- Systematically considers multiple objectives and metrics of performance – waste management, TRU reduction, uranium, economics
- Clarifies tradeoffs, e.g., 1-tier vs. 2-tier, fast reactor conversion ratio, timing of deployments, FR fuel cooling time, oxide vs. metal fuel
- Performs sensitivity analyses to better understand how the system performs
- Informs DOE’s decision on a path forward, and helps DOE to communicate to stakeholders
VISION Analyses: Assumptions for Nominal Cases are Based on Technical Feasibility

<table>
<thead>
<tr>
<th>1-tier</th>
<th>2-tier</th>
</tr>
</thead>
<tbody>
<tr>
<td>Centralized UOX separation starting in 2020 at 0.1% loss/recycle</td>
<td>Recover U-Pu, MA, Cs/Sr, etc.</td>
</tr>
<tr>
<td>Recover TRU, Cs/Sr, etc.</td>
<td>Recover TRU, Cs/Sr, etc.</td>
</tr>
<tr>
<td>1st fast reactor (CR=0.50) in 2022</td>
<td>MOX-U-Pu</td>
</tr>
<tr>
<td>MOX separation starting in 2035 at 0.1% loss/recycle; recover TRU, Cs/Sr, etc.</td>
<td></td>
</tr>
</tbody>
</table>

**Nuclear electricity generation**

- 1st UOX separation plant 800 tonnes/yr
- Later UOX separation plants 1600 tonnes/yr, deployed to eliminate used fuel backlog by 2100
- MOX separation deployment lags UOX by 15 years, at ~9x lower capacity
- FR used fuel separation capacity built as FRs are built (co-location)
It is feasible to eliminate excess used fuel storage before 2100 while recycling all separated TRU.

Total TRU reduction via recycle must consider both TRU consumed and TRU avoided.

Adding a MOX cycle significantly delays and reduces the number of fast reactors.

If fast reactor used fuel processing is centralized, more TRU is tied up in cooling used fuel and fewer fast reactors can be supported.

Closed fuel cycles appear to cost 5-6 mils more than once-through, but the uncertainty bounds are large.

Examination of cost drivers help show which improvements contribute most to closing the cost gap.
Material Transportation, Storage, & Disposal – Recent Accomplishments

- Continued repository benefit analyses (Alternative disposal pathways, response to questions)
- Material transportation requirements (Assess packaging availability for material shipment, storage)
- Technical evaluation of “Institutional Infrastructure” issues (policy, law, regulation, standards, practice, …)
- Support for GNEP facilities: (M-TSD strategy consistency, waste stream analyses, disposal alternatives)
Integrated Waste Management Strategy (IWMS) - Status

- IWMS identifies all waste streams
  - Builds on a fuel-cycle using advanced separations
  - Describes potential benefits of a NWPA modification to allow risk-based waste management similar to provisions of 10 CFR 61
  - Could be applied to DOE, NRC, space, defense, medical, and imaging wastes

- Emphasizes recycle and reuse, but based on economic recovery evaluation factoring in value of material and cost avoidance of disposal

- Considers need for industry to have a reliable system to disposition all byproducts/wastes during a phased transition PUREX, UREX+, Echem...

- Independent of 1 or 2 tier recycling concepts

- Integrates current EM GTCC EIS effort

- Anticipates combined oxide and metal waste forms, potential for alternative repository concepts

- Suggests a collaborative approach among DOE-NE/EM/RW, NRC/EPA
The mission is to develop and demonstrate durable waste and storage forms for management and disposal.

Fundamental understanding of long-term waste and storage form behavior can only come about through closely coupled theory, modeling, and experimentation.

Waste treatment and disposition must have acceptable cost impacts. The waste form campaign develops and demonstrates cost effective waste processes and durable forms.
Requirements for Waste Forms

- Performance (durability, heat transfer, radiation stability)
- Cost effective (low disposal volumes, low cost processes)
- Flexibility (waste composition, loading, process ranges, etc.)
- Predictable performance (natural analogues, release models)
- Match the waste stream to waste form and disposal environment (e.g., keep noble elements in metal waste form)
Designing Nuclear Waste Forms

- Waste chemistry, radionuclide(s) to immobilize, and disposal environment guide the selection of waste form

- Let nature guide, particularly when long-term performance is important

- Use *intelligent materials design*
  - Systematic approach rather than “cook and look” approach

- Design material in combination with process
  - Process influences form and vice-versa
Accomplishments to Date
Waste Management Integration/Technology Development

- Support development of IWMS and waste management systems analyses
- Preliminary technology readiness level assessment and evaluate waste and storage form durability testing approaches
- Analyses of waste form performance and development of performance requirements in a range of geologic settings
- Continued work on production and performance of candidate baseline waste and storage forms
  - Cs/Sr, I, Tc, TM, and LN for aqueous and electrochemical processes
Accomplishments to Date (cont.)
Modeling and Simulation and Long-Term S&T

- Conducted workshop and developed Roadmap for M&S and Long-Term S&T
- Future Directions
  - Development and qualification of advanced, more durable, tailored waste and storage forms
  - Development of advanced geologic disposal concepts in a range of geologic settings and geochemical environments
  - Enhanced understanding of geologic repository performance
  - Systems-level optimization of closed fuel cycle and geologic repository designs
Waste Form S&T of Specific Materials

- Structure and chemistry
- Phase transitions as a function of temperature and radiation field
- Chemical durability over a range of conditions (thermodynamic and kinetic studies)
- Corrosion mechanisms and rates
- Mechanical properties
- Response to radiation
“Integration by Simulation”
Waste Form Properties & Performance

Computational simulation of:
- Multi-scale simulations of waste form properties (molecular-scale)
- Modeling/simulation of waste form behavior in the near-field environment (meter-scale)
- Reference environments (fractured/monolithic, oxidized/reduced, salt/crystalline/clay)
- Reference waste forms (glass/metal/ceramic phases containing radionuclides)

Goal: The modeling and simulation effort will identify the controlling mechanisms and processes for different waste form materials in a range of geochemical environments at different spatial and temporal scales
- Applied in a general manner to provide a scientifically-defensible basis for waste form development, qualification, and future repository system analysis
Full Performance Simulation

University Involvement and International Collaborations

- **Universities**
  - Key to the success of GNEP waste form campaign
  - Goal is to establish integrated program

- **International Collaborations**
  - France
    - Potential collaborative tasks on waste management and advanced waste form S&T being formulated
  - Japan
    - Action Plan in place
    - Potential collaborative tasks on waste management and advanced waste form S&T being formulated as Part of Phase 1
    - Draft Phase 1 report complete
  - Australia
    - Action Plan in Place
  - Russia
    - Action Plan in place
  - China
    - Action Plan in place
Yucca Mountain Repository Program

- Removal of uranium and transuranics and disposition of fission product storage and waste forms as part of a closed fuel cycle would impact the potential technical capacity of Yucca Mountain (assuming statutory limit is adjusted)

- Many factors potentially control the technical capacity of a repository including:
  - Postclosure repository performance as measured by radiation dose to the public over 10,000+ years
  - Physical area available with suitable geologic and hydrologic characteristics
  - Waste form volume
  - Waste thermal output

- All of these factors require further analysis before the potential benefits can be clearly understood

- The technical capacity of Yucca Mountain will be capable of accommodating several generations of waste should the technologies being pursued under the GNEP prove successful
DOE Environmental Management E&T program is evolving and planning additional investment in processing and waste management technology development.

Interface with EM vital to success of GNEP in S&T and development of integrated waste management strategy.
Questions and Discussion
Backup
Advanced Waste Forms

- Basic Science Challenges, Opportunities, and Research Needs
  - Novel materials for advanced waste forms
  - Long-term predictions of advanced-waste-form performance in the natural environment
  - Thermodynamics for complex systems
  - Radiation and radiolysis effects
  - Interfaces and nano-scale dynamic behavior
Repository Modeling
- Waste Form and Material Degradation
- High Fidelity Assessment of the Geochemical Environment
- Software Infrastructure for Coupled System Performance Assessment
- Uncertainty Propagation
- Visualization
- Modernizing and Parallelizing Existing Legacy Codes
Priority Research Directions

- Chemistry of mineral surfaces, nanophases, and solutions
- Subsurface monitoring, imaging, and characterization
- Physics and chemistry of multiscale systems
- Biogeochemistry
Advanced Fuel Cycle Safeguards

Michael Miller
GNEP Safeguards Campaign Director
Los Alamos National Laboratory

U.S. Nuclear Regulatory Commission
Rockville, MD
June 10, 2008
Outline

- Introduction and Current Technical Basis
- Reprocessing
- Fuel Fabrication
- Fast Reactor
Introduction and Current Technical Basis

- Comparison of NRC, DOE, IAEA Definitions and Requirements
- Fundamentals of NDA
- Gamma-ray Based Instruments
- Neutron Based Instruments
- Calorimetry
Comparison of NRC, DOE, IAEA Definitions and Requirements
Safeguard Regimes

Three Safeguard Regimes
- NRC: Regulates commercial nuclear facilities
- DOE: Regulates defense nuclear facilities
- IAEA: Applies safeguards to Nonproliferation Treaty signatories

Purpose of Safeguard Regimes
- Domestic (NRC and DOE)
  - Protect against sabotage, theft and other threats of direct concern to the government and operators of the facility
- International (IAEA)
  - Goal is to detect diversion of significant amounts of fissile nuclear materials by the State/facility operator through verification of the State’s declaration
Types of Safeguards Measures

- **Nuclear Material Accounting**
  - Establishing the quantities of nuclear material present within defined areas and the changes in those quantities within defined periods.

- **Containment and Surveillance**
  - Ensures previously measured material in containers remains in place and unaltered or moves along declared and authorized paths
  - Examples: video and seals

- **Process Monitoring**
  - The use of process data (flow rate, temperature, pressure, density, etc.) to draw frequent conclusions on plant configurations, operations and material flows/inventories
  - Necessary for In-process, interim inventories
Safeguards Goal of Material Balance Areas (MBAs)

- Fully measured material balance
  - All materials accounted for
  - All materials have measured values
DOE Definition of MBA

- Subsidiary account of facility
- Geographical area with defined boundaries
- Identify location and quantity of nuclear materials
NRC Definition of MBA

- Identifiable physical area
- Quantify nuclear material with measured values
IAEA Definition of MBA

- Quantity of nuclear material in transfers can be determined
- Physical inventory can be determined
- Establish a material balance
Views of the MBA - Accountant

- Start with a “known” listing of material
- Record receipts and shipments
- The result is the ending, or “book” inventory:
  \[ BI + TI – TO = \text{Book Inventory} \]
- Verify that book inventory = physical inventory
  \[ MB = \text{Book inventory} – \text{Physical Inventory} \]
Views of the MBA - Scientist

- MBA is the “closed system” for which mass is conserved
- Determine the inventory through measurement at end of period
- Measure all the terms and determine the difference

Mass Balance = BI – EI + TI – TO
Views of the MBA – Physical Protection

- What is the target?
- Where is the target located?

An area containing nuclear material that:
- Must have controls
- Must be protected
Summary of MBA definitions

- Accounting unit of facility
- Physical location
- Measure materials
- Establish a material balance; loss detection
Material Accountancy

**Inventory Types**

- Long-term, Shut down Inventory
  - *Every 6-12 months the facility must cease processing and determine the amount of material contained within the facility*
- Interim Inventory, Shut down not required
  - *Every 1-2 months the facility must determine the amount of material within the facility, but shut-down is not required*
  - *In-process inventory is normally performed*
    - More difficult than shut down inventory
    - Larger errors due to estimates of material quantities
Detection requirements

- Inventory Difference (Material Balance, Material Unaccounted For) must be determined within certain error limits
- Those error limits determine how well the material must be measured
- Each regime has different error limits
Inventory Difference

**Inventory Difference (ID)** =

- Beginning physical inventory
- + Sum of increases to inventory (receipts)
- - Sum of decreases from inventory (shipments)
- - Ending physical inventory

**Sigma ID**
- The uncertainty in the ID
- A statistical combination of the uncertainty of each measurement that makes up the ID
- Determines what losses you can detect
### Sigma ID Requirements

Based on the Current, Most Stringent, Category I Requirements

<table>
<thead>
<tr>
<th>Agency</th>
<th>Goal/Requirement Terms</th>
<th>Sigma ID</th>
<th>Frequency of Long-Term, Shutdown Inventory</th>
<th>Frequency of Interim Inventory, Shutdown Not Required</th>
</tr>
</thead>
<tbody>
<tr>
<td>IAEA</td>
<td>Material Unaccounted For (MUF): 8kgs Pu abrupt in one month and 8kgs protracted in one year</td>
<td>Sigma ID &lt;= 2.42 kg of Pu</td>
<td>Annual</td>
<td>Monthly</td>
</tr>
<tr>
<td>NRC</td>
<td>Standard Error of Inventory Difference (SEID)</td>
<td>Sigma ID &lt;= 0.1% of active inventory</td>
<td>Semi-Annual</td>
<td>Monthly</td>
</tr>
<tr>
<td>DOE</td>
<td>Limit of Error (LOE)</td>
<td>Sigma ID &lt;= 1% of active inventory or Category II of material</td>
<td>At least Annually</td>
<td>Bi-monthly</td>
</tr>
</tbody>
</table>
Rough Estimate of Sigma ID for a Commercial Reprocessing Facility

- **IAEA goal for Sigma ID**
  - Absolute number
  - Independent of the facility’s throughput
  - More challenging for high throughput facilities.

- **NRC and DOE requirements for Sigma ID**
  - Percentages of the active inventory
  - Change with the throughput
  - Current NRC requirement is the most challenging of the three, even for this high throughput facility.

<table>
<thead>
<tr>
<th>LWR Spent Fuel Processed per year</th>
<th>800 MTHM</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu processed/yr</td>
<td>8,000 kg</td>
</tr>
<tr>
<td>(1% of spent fuel)</td>
<td></td>
</tr>
<tr>
<td>Pu processed per month</td>
<td>667 kg</td>
</tr>
<tr>
<td>(assumed to be active inventory at time of shut down inventory)</td>
<td></td>
</tr>
<tr>
<td>NRC’s Sigma ID is 0.1% of active inventory</td>
<td>667 g</td>
</tr>
<tr>
<td>DOE’s Sigma ID is 1% of active inventory</td>
<td>6.67 kg</td>
</tr>
<tr>
<td>IAEA’s goal for Sigma ID</td>
<td>2.42 kg Pu</td>
</tr>
</tbody>
</table>

NRC Seminar June 10, 2008
Improving Safeguards Effectiveness

- **Reduce sigma ID**
  - Better measurements
  - Less material
    - *Smaller material balance areas*
    - *More frequent inventories*

- **Additional Measures**
  - C/S
  - Processing Monitoring
  - Access
  - Inspector Presence
  - But, quantification needed
Fundamentals of NDA
The **Fully Measured Material Balance**

- Strive for highest possible accuracy
- Minimize uncertainty in Material Unaccounted For (MUF)
- Preferred measurement techniques
  - weighing, combined with
  - destructive analysis of samples
Some SNM items cannot, in practice, be measured by DA

waste, scrap, sealed product, unknown items, etc.
SNM comes not only in a variety of physical and chemical forms, but also in a variety of containers. Each container has an effect on the absorption of the emitted radiation and the measurement result.
Nondestructive Assay (NDA)

The quantitative or qualitative determination of the kind and/or amount of nuclear material in a sample without alteration or invasion of the sample.
Basic Measurement Principle

Since each gram of a given isotope decays at a specified rate, the amount of radiation per second is proportional to the number of grams of the SNM in question.

The measured radiation is proportional to the amount of SNM in the sample IF…

- all of the radiation produced in the sample is emitted, and
- all of the emitted radiation is detected

These assumptions are hardly ever met, so data analysis must correct for the “missing” radiation. This is the main challenge in developing SNM measurement techniques and instruments.
Role of NDA

- Rapid verification
- Measurements of poorly-characterized items
- Assay or verification of sealed items
  - items in storage
  - temporarily sealed items (from cleanout, scrap, ...)
  - fuel rods and assemblies
- Measurement of holdup (ducts, pipes, gloveboxes)
- Support of audits & inspections
  - by outside agencies (IAEA, Euratom, ABBACC, GAN,...)
  - by in-plant inspectors for internal assessments
Uses for NDA

- Ore location
- Process Control
- Quality control
- Health & safety
- Criticality control
- Material control
- Materials Accounting
- Waste characterization/verification/screening
- Nuclear Inspections (nonproliferation, arms control, regulation)
SNM Attributes

- Emits alpha & beta radiation
- Emits gamma ($\gamma$) radiation
- Emits infra-red (heat) radiation
  - [from $\alpha$ emissions into the matrix]
- Emits singles neutrons
  - [from ($\alpha,n$) matrix interactions, delayed neutrons]
- Emits coincidence neutron radiation
  - [from spontaneous and induced fissions in the sample]
- Is fissionable - can be induced to fission with neutrons.
Passive vs. Active NDA

**Passive NDA:** Measure the radiation emitted spontaneously by the sample.

**Active NDA:** Irradiate the sample and measure its response to the radiation.
Examples of Confirmatory Measurements

A Geiger counter is used to survey ductwork in a reprocessing plant to locate radioactive deposits. Quantitative holdup measurements then focus on these locations to determine the in-plant holdup.
Examples of Verification Measurements

Quantitative measurements — not used to replace accountability values.

- Sampling of SNM inventory during inspection
- Mass measurements during some emergency inventories
Gamma-ray Based Instruments
Basic Gamma-Ray Assay

Fundamental Measurement Principle

- Specific gamma rays (identified by energy) indicate the isotopic composition of the sample.
- Intensity of the gamma rays indicate the amount of the isotope.
Generic $\gamma$-Ray Assay Equation

\[ M_{SNM} = \frac{R_{Rad} \cdot CF}{K} \]

- $M_{SNM}$ = Mass of Special Nuclear Material (SNM)
- $R_{Rad}$ = measured radiation rate (counts per second) from SNM sample
- CF = correction for losses due to:
  - Sample self-absorption
  - Container absorption
  - Measurement system electronics
- K = Calibration constant (corrected response/gram SNM)
Gamma-Ray Signature

\[ \gamma \text{ rays from Alpha Decay} \]

\[ ^{235} \text{U} \rightarrow ^{231} \text{Th}^* \rightarrow ^{231} \text{Th} + \gamma \]

[Half-life = 710,000,000 y]

Gamma rays:
- 185.7 keV 54%
- 143.8 keV 11%
- 163.4 keV 5%
- 205.3 keV 3%

\[ \alpha (^4 \text{He}) \]

alpha emission

\[ ^{231} \text{Th}^* \]
Major $\gamma$-Ray Signatures

$^{239}\text{Pu}$

$^{239}\text{Pu} \rightarrow ^{235}\text{U}^* + \alpha$

Major gamma rays:
- $129.28\text{ keV}$  Intensity  $140,000$ gammas/(g-sec)
- $413.69\text{ keV}$  Intensity  $34,000$ gammas/(g-sec)

$^{235}\text{U}$

$^{235}\text{U} \rightarrow ^{231}\text{Th}^* + \alpha$

Major gamma ray:
- $185.72\text{ keV}$  Intensity  $43,000$ gammas/(g-sec)
Interaction of Gamma Rays with Matter

**PHOTOELECTRIC Effect**

**BEFORE**

Gamma ray (or x ray) “collides” with atomic electron, and

**AFTER**

imparts ALL of its energy to that electron

**COMPTON Effect**

**BEFORE**

Gamma ray (or x ray) “scatters” off an atomic electron, and

**AFTER**

imparts PART of its energy to that electron
Absorption of $\gamma$-Rays

$I = I_0 e^{-\mu \rho x}$

Transmission, $T = \frac{I}{I_0} = e^{-\mu \rho x}$

- $\rho$ = density ($\text{g/cm}^3$)
- $\mu$ = mass absorption coefficient ($\text{cm}^2/\text{g}$)
- $x$ = thickness (cm)
Mass Absorption Coefficient

![Graph showing Mass Attenuation Coefficient vs. Energy (keV)](image)

- 185.7 keV 235U ASSAY
- 400 keV 239Pu ASSAY
- 765 keV 239Pu ASSAY
- 1001 keV 238U ASSAY
Nal Scintillation Detector

Gamma ray transfers all of its energy to an electron.

Photoelectron ionizes atoms which relax by emitting light whose intensity is proportional to the energy lost by the gamma ray.

This light travels to the photocathode of a photomultiplier tube, which emits electrons that multiply as they interact with the dynode string and are finally captured by the anode.
Photoelectron ionizes Ge atoms liberating charge that is proportional to the energy lost by the $\gamma$ ray.

Electrons move toward the positive electrode and “holes” toward the negative one causing a charge pulse.

The preamplifier converts the charge pulse to a voltage pulse that is proportional to the energy of the $\gamma$ ray.
Uranium Spectral Features

High-Enriched uranium spectrum at high-(Ge) and low- (NaI) energy resolution

- 60 keV ($^{241}$Am)
- 185.7 keV ($^{235}$U)

Counts vs. channel graph showing peaks at 60 keV and 185.7 keV.
Plutonium Spectra

Counts (Log Scale)

Energy (keV)

93% $^{239}$Pu

Nal

CdZnTe

CdTe

HPGe

NRC Seminar June 10, 2008
The Isotopic “Fingerprint”

High-Enriched Uranium:
- 93% $^{235}$U, very little $^{238}$U
- 186-keV gamma-ray peak is strong compared to other radiation, such as x-rays
- little background above 186 keV from $^{238}$U

Low-Enriched Uranium
- 3% $^{235}$U, mostly $^{238}$U
- 186-keV gamma-ray peak is weak compared to other radiation
- significant background above 186 keV from $^{238}$U

Plutonium:
- 93% $^{239}$Pu, some $^{241}$Pu
- 414-keV gamma-ray peak from $^{239}$Pu evident, as is 208-keV from $^{241}$Pu
Energy Calibration

\[ E_{\gamma} (\text{keV}) = A \cdot \text{Channel}\# + B \]

Energy accurate to about 0.1 keV
### Energy Resolution

<table>
<thead>
<tr>
<th>Detector</th>
<th>FWHM</th>
<th>E</th>
<th>FWHM/E</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaI(Tl)</td>
<td>29 keV</td>
<td>661 keV</td>
<td>7.5%</td>
</tr>
<tr>
<td>NaI(Tl)</td>
<td>150 keV</td>
<td>1332 keV</td>
<td>10.6%</td>
</tr>
<tr>
<td>Ge (Planar)</td>
<td>540 eV</td>
<td>122 keV</td>
<td>0.44%</td>
</tr>
<tr>
<td>Ge (Coax)</td>
<td>1.75 keV</td>
<td>1332 keV</td>
<td>0.13%</td>
</tr>
</tbody>
</table>
$\gamma$-Ray Photopeak Area Analysis

$\gamma$ ray background is evaluated from the Compton continuum on either side of the peak

Counts/Channel

$\gamma$ Energy

Peak Centroid

Net Peak Area

Gamma Background

Gamma Energy

$E_\gamma$
Basic Gamma-Ray NDA Setup

Transmission source placed on far side of sample from detector.
Sample mounted on rotating platform, at a distance from the detector.
Detector views entire sample at one time.
Gamma Spectroscopy Instrumentation

- Battery-powered
- Computer-based
  - data acquisition
  - data analysis
- Commercially available
Sample Characteristics for Successful Gamma-Ray Assay

- Medium to low density
- Medium to small size
- Homogeneous (uniform) material distribution

Dense, heterogeneous materials are inappropriate for gamma-ray assay, because they have too much absorption. Such samples are better suited for neutron assay techniques.
Basic Calibration Procedure

Purpose: to establish the relationship between the response of the detector system and the amount of SNM in the sample.

- Set up measurement apparatus for assay campaign.
- Establish data acquisition & analysis procedures.
- Measure known samples (calibration standards) using these procedures and this setup.
Example Gamma Assay Calibration Curve

Note that absorption by the sample is increasingly important at higher SNM masses.

Application of the self-absorption correction (using the external source) removes the effect of this absorption.
Normal (Gaussian) Distribution

- 68% “Confidence Level”
- 95% “Confidence Level”

68.27% and 95.45% coverage intervals for a normal distribution.

N - 2σ  N - σ  N  N + σ  N + 2σ
Uncertainty in a Nuclear Count

For a measured number of counts, \( N \), from a nuclear counting process, the standard deviation in \( N \) is:

\[
\sigma (N) = \sqrt{N}
\]

The RELATIVE STANDARD DEVIATION (RSD or \( \sigma_R \)) of the measurement is:

\[
\sigma _R (N) = \frac{\sigma (N)}{N} = \frac{1}{\sqrt{N}}
\]

To improve the RSD, increase \( N \) (i.e., “take more data”):
- Count longer or repeat measurement (equivalent)
- Use “hotter” source
- Move sample and detector closer to one another
Measurement Control

Goals:
- To control the quality of the measurement process
- To verify the continued stability of the measurement system since the last calibration

Procedures
- Assay measurement control standards periodically
- Measure room radiation background periodically
- Measure data quality (appearance of spectrum, etc.)
  - electronics gain
  - detector resolution
- Note visual appearance of instrument
Measurement Precision and Bias

**Measurement Precision:**
The degree to which repeated measurements of the same sample give the same results.

**Measurement Bias:**
The amount by which the average of many results on a standard differs from the true value.
Methods to Improve Measurement Precision

- Count for longer time
- Operator Errors
  - Provide explicit assay procedures
  - Provide careful training for assay procedures
  - use checklists to verify execution of procedures
- Background Variations
  - Shield detector from extraneous signals
  - administratively control source traffic in counting area
  - perform regular background measurements, diagnostics
Dealing with Measurement Bias

- Sources of Measurement Bias
  - Statistical errors in calibration
  - Shifts in shielding, absorbers, collimators
  - Changes in geometry of setup (detector, sample stand, …)
  - Differences between sample container and container for calibration standards (diameter, height, wall thickness, …)
  - Lumps in sample (error in absorption correction)

- Strategies to Reduce Measurement Bias
  - Calibrate often and make calibration measurement precise
  - Anchor shielding and collimators to measurement system
  - Anchor detector and sample stations
  - Calibrate for several container sizes; develop geometry corrections to calibration; control sample containers to match standard containers.
  - Assure uniform samples; apply lump corrections, if possible.
Features

- Assayed standard Pu foil, built into instrument.
- Generally normal, quite stable performance
- Some excursions, in May, August, 1980 [Temperature problems when air conditioning fails]
- Slight downward drift in winter, 1980
Detector views the sample through a collimator.

Visible volume determined by collimator and the absorption coefficient of U.

The 186-keV $\gamma$-ray intensity is proportional to the $^{235}$U in the visible volume.
# Infinite Thickness for 186-keV γ Rays in Uranium Materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Density ((\rho)) (g/cm(^3))</th>
<th>Mean Free Path (cm)</th>
<th>“Infinite” Thickness (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metal</td>
<td>18.7</td>
<td>0.04</td>
<td>0.26</td>
</tr>
<tr>
<td>UF(_6) solid</td>
<td>4.7</td>
<td>0.20</td>
<td>1.43</td>
</tr>
<tr>
<td>UO(_2) (sintered)</td>
<td>10.9</td>
<td>0.07</td>
<td>0.49</td>
</tr>
<tr>
<td>UO(_2) (powder)</td>
<td>2.0</td>
<td>0.39</td>
<td>2.75</td>
</tr>
<tr>
<td>U(_3)O(_8) (powder)</td>
<td>7.3</td>
<td>0.11</td>
<td>0.74</td>
</tr>
<tr>
<td>Uranyl nitrate</td>
<td>2.8</td>
<td>0.43</td>
<td>3.01</td>
</tr>
</tbody>
</table>
SLAB CORRECTION FACTOR (Far-Field) $CF_{\text{atten}}$

Assumes that detector is far enough away from source that parallel $\gamma$-ray “beams” reach the detector.

FOR $D >> x$:

$$CF_{\text{atten.}} = \frac{\mu \rho x}{1 - e^{-\mu \rho x}} = \frac{-\ln(T)}{1 - T}$$
Solution Assays by Gamma-Ray Spectroscopy

- Absorption-corrected passive gamma-ray assay
- High-resolution $\gamma$ detector measures spectrum through floor of glovebox.
- Gives grams isotope in a 0.001 to 400 gram SNM/liter sample, with 0.2% to 1% accuracy in 1000 sec.
Absorption edge densitometry, x-ray fluorescence

K-Edge densitometer

Hybrid K-Edge XRF
Other Transmission-Corrected Gamma Assay Systems
Segmented Gamma Scanner (SGS)

- Passive, absorption-corrected gamma-ray assay of SNM
  - samples of final product
  - low-density U waste
- Sample is assayed one segment at a time

- Appropriate samples: small (<2 liters) or low density waste
- Accuracy possible in 1000 sec: 2% to 10% on 1 to 200 grams U

SNM Sample
Collimator
External Radiation Source
Movable Source Shield
Sample Movement
Sample Rotator
Detector Movement
Detector
Transmission Source
Segment Definition
Rotating Scan Table
Detector
Sample
Collimator
Gamma-Ray Tomography

- Segmentation is replaced by collimation that divides the sample into “voxels.”
- Measuring the transmission, in each voxel, gives a 3-D “density map” of the sample.
- The measured density and $\gamma$-ray rate from each voxel provide a 3-D image of the SNM content.
Passive Gamma-Ray NDA Applications:
Plutonium Isotopic Composition

A high-resolution spectrum provides gamma-ray intensities from the isotopes in the sample. Isotope ratios are computed from peak ratios. Since a relative measurement is performed, standards are not needed, and measurement geometry does not need to be controlled. Relative detector efficiency, as a function of gamma-ray energy, is obtained from the spectrum.
Portable In-Plant Holdup Measurements by Gamma-Ray Spectroscopy

- Portable multichannel analyzer with gamma detector and data analysis electronics.
- Operator can position a shielded detector in a variety of locations to meet variety of measurement requirements.
- Holdup measured at 5% to 50% accuracy on 0.1 to 10 grams SNM; accuracy very dependent on measurement geometry.
Gamma NDA Summary

- **Gamma NDA Technique:**
  - Count gamma rays from sample
  - Gamma-ray absorption correction very important
  - Appropriate samples: small to medium density/size, uniform

- **Accuracies Possible:**
  - 0.2% to 50%, depending on sample uniformity and technique
  - Best performance on solutions
  - Improved performance for high-resolution over low-resolution detectors.
  - Worst cases: Large, dense, heterogeneous samples [often assayable by neutron techniques]
Advanced Fuel Cycle Safeguards

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- Calorimetry
Neutron Based Instruments
The Neutron’s Role in Fission

- Initiator ("Trigger")
- Prompt Coincidence Emission
- Delayed Singles Emission
Introduction: Why Neutron Counting?

- Neutrons have great penetrability. Thus neutron signatures are sometimes the only way to rapidly assay large, dense samples.
- Neutron Count Data can be obtained rapidly. (But isotopic information is needed to interpret the data correctly and this adds more time to the assay.)
- Spontaneous Neutron Emission is a primary signature of the even isotopes of plutonium.
- Induced Fission Neutrons are a signature for both fissile plutonium and uranium.
History of Neutron Counting

- The first neutron assay instruments attempted to use the total neutron rate to deduce assay information -- however, accurate assays can be obtained only for a very few types of SNM.

- The next development was neutron coincidence counting. This technique has had wide application for international safeguards. It has had more limited application to US domestic safeguards because large errors can occur if the technique is not applied correctly to impure materials.

- Neutron Multiplicity Counting is an extension of neutron coincidence counting. It improves neutron assay accuracy dramatically by adding more measured information.
In standard neutron coincidence counting, a priori information must be used to obtain an assay. Does not work well for materials that contain variable, low-Z impurities.

In neutron multiplicity counting, the distribution of coincident multiplets in the neutron pulse stream is used to obtain a third measured quantity: the rate of coincident “triples”.

With careful detector design to reduce the variables to three, neutron multiplicity assays are possible without sample dependent calibrations.
Neutron Detection - “Well Counter”

Detector electronics registers the coincidence and total count rates

\[ ^3\text{He} + n \rightarrow p + t \ (Q=765 \text{ keV}) \]
Moderation

- **Moderation** is the process by which a neutron collides with matter and **loses energy**.
  - i.e. 2 MeV to 0.025 eV.
- **The probability of neutron detection in $^3$He is largest** when the neutrons have energies near **thermal**.
- Most energy lost (best moderation) when neutron collides with nuclei of similar mass. {i.e. hydrogen (protons)}.
  - Water
  - Polyethylene
- **Moderation usually takes many collisions (~27 for a 2MeV neutron in polyethylene)**.
Neutron Detector Die-Away

- After moderation, neutrons are lost in the detector by several processes:
  - Diffusing out of detector.
  - Diffusing to a $^3$He detector tube and being absorbed.
  - Absorption by hydrogen or cadmium.
- Hydrogen both moderates and absorbs the neutrons.

In most thermal detectors the neutron population decreases nearly exponentially in time. The time constant is called the die-away time.
The Shift Register - An Example

Four Pulses “Approach” the Escalator

1st neutron "triggers" accumulator

On Escalator: 0
In Accumulator: 0

Add 1 to Accumulator

Second neutron triggers accumulator

On Escalator: 1
In Accumulator: 1
Third neutron triggers accumulator

On Escalator

Add 2 to Accumulator

In Accumulator

2

Fourth neutron triggers accumulator

On Escalator

Add 3 to Accumulator

In Accumulator

3

6

Four Pulses Yield 6 Coincidences
What Do We Really Count?

• The Shift Register really counts coincidences.
• The Rossi-Alpha distribution is based on counting all possible coincidences.
• The formula for all possible coincidences is:

\[
\frac{n(n - 1)}{2}
\]

• Where \( n \) is the number of pulses.
The Shift Register Circuit

Input

Predelay

Shift Register

Up - Down Counter

R + A Scaler

A Scaler

Long Delay

Strobe
Multiplicity Shift Register

- **Input**
  - Predelay
  - Shift Register
    - Up-Down Counter
      - Sort by Number in Counter
        - Zeros Scaler
        - Ones Scaler
        - Twos Scaler
          - Long Delay
            - Accidentals Scalers

Multiplicity Distribution – 3.8 kg Pu metal

Multiplicity

Counts

P, Counts in R+A Gate
Q, Counts in A Gate
Calibration Curve Method

Need representative standards for each sample type and careful calibration measurements.
Known Alpha

Use singles and doubles to deduce a “multiplication” correction that linearizes the calibration. Works well for pure oxides, metals, and fluorides. Still need standards.
The Problem with Standard Coincidence Counting

There are 3 principal unknowns in neutron counting:

\[ ^{240}\text{Pu-effective mass, } a, \text{ and } M. \]

**Standard Coincidence Counting** provides only 2 pieces of measured information, singles and doubles (or totals and coincidences). To obtain an accurate assay, one must know a lot about the sample.

If the assumed information is incorrect, large errors can occur.

In **Neutron Multiplicity Counting**, 3 pieces of measured information are used with a mathematical model to deduce an assay that is far superior for most impure materials.
The Point Model Equations

\[ S = F \varepsilon M \nu_{s1}(1+\alpha) \]

\[ D = F \left( \frac{f_d}{2} \right) (\varepsilon M) \left\{ \nu_{s2} + \left[ \frac{(M-1)}{(\nu_{i1} - 1)} \right] \nu_{s1}(1+\alpha) \nu_{i2} \right\} \]

\[ T = F \left( \frac{f_t}{6} \right) (\varepsilon M) \left\{ \nu_{s3} + \left[ \frac{(M-1)}{(\nu_{i1} - 1)} \right] \left[ 3\nu_{s2} \nu_{i2} + \nu_{s1}(1+\alpha) \nu_{i3} + 3\left[ \frac{(M-1)}{(\nu_{i1} - 1)} \right]^2 \nu_{s1}(1+\alpha) \nu_{i2} \right\} \]

where:

- \( \varepsilon \) = detection efficiency
- \( f_d \) = fraction of doubles in the coincidence gate
- \( f_t \) = fraction of triples in the coincidence gate
- \( F \) = spontaneous fission rate = 473.5 n/s/g * effective PU-240 mass
- \( \nu_{n1} \) = average number of neutrons produced per fission event (n=s -- spontaneous fission, n= i -- induced fission)
- \( \nu_{n2} \) = average number of neutron pairs produced per fission event
- \( \nu_{n3} \) = average number of neutron "triplets" produced per fission event

- \( \alpha \) = ratio of (alpha, n ) neutron rate to the spontaneous fission rate
- \( M \) = fission multiplication
Neutron NDA Techniques

- Passive coincidence counting
- Active interrogation with coincidence counting
- Active interrogation with counting of delayed neutrons
The High-Level Neutron Coincidence Counter (HLNC)

The cylindrical counter (HLNC) surrounds the sample with $^3$He detectors. The data acquisition and analysis electronics are on the bench. This counter is commercially available.
Active Neutron Coincidence Counting

- Random (AmLi) neutron source induces fissions in fissile nuclei.
- High background, (reduced sensitivity) compared to passive counters (due to AmLi source).
- Fissioning source surrounded by neutron detectors.
- Prompt multiple neutron emission from induced fission detected as coincidence neutron events.
- Designed primarily for $^{235}\text{U}$ assay.

Coincidence electronics registers the coincidence count rate which is proportional to the mass of fissile isotopes.
Active Neutron Coincidence Counter (AWCC)
Plutonium Scrap NMC

Sample Cavity is 20 cm x 41 cm and is Cadmium Lined
The Shuffler Assay Principle

(a) The interrogating source is stored in heavy shielding.

(b) The interrogating source quickly moves to a position near the SNM. The source remains near the sample for a few seconds, inducing fissions.

(c) The interrogating source is quickly moved back to its shielded position; the detectors then count delayed neutrons.

The “irradiation-count” cycle is repeated many times to acquire adequate statistical precision. The periodic motion of the interrogating source, shuffling in and out of its shielded storage, gives the “Shuffler” its name.
$^{252}\text{Cf Shuffler}$
Sample Characteristics for Neutron Assays

- Uniformity still important, but not always critical
- High-density
- Large size
- Heterogeneous

*In addition, most samples assayable with gamma rays are also assayable with neutrons*

Special Cases for Neutrons

- Low or high-density waste (small amount of SNM)
- High-gamma-yield material (e.g., spent fuel)
Problems for Neutron Assays

- **Hydrogenous materials (Protons!)** [increased system response]
  - plastics
  - moisture
  - any other moderator

- **Absorption** by high-density SNM (fissile)
  - *n capture to induce fissions* (absorption)
  - *induced fissions* (multiplication)

- **Absorption** by neutron poisons [good neutron absorbers]
Appropriate Samples for **Active** Neutron Coincidence Counting and **Delayed** Neutron Counting

- All U samples [assays mass of $^{235}\text{U}$]
- Pu samples [assays mass of $^{239,241}\text{Pu}$, passive signal may complicate]
- Mixed U/Pu samples (MOX) [need passive and active measurement to sort out components]
- Low-mass or high-gamma samples (waste, spent fuel) - [delayed neutron counting (Shufflers)]
Neutron NDA Calibration

- Philosophy the same as for gamma NDA
  - Measure standards
  - Apply all relevant corrections

- Best to calibrate over a range of expected sample characteristics
Neutron Calibration Standards

- Standards’ characteristics:
  - Range of SNM masses
  - Range of matrices
  - Range of material types

- Establishes response variations from:
  - Moderation
  - Multiplication
  - Absorption
Measurement Precision in Neutron NDA

- **Origins**
  - Random decay processes (counting statistics)
  - Fluctuating neutron backgrounds

- **Strategies to improve precision**
  - Longer count times (improve statistics)
  - Background/source control
  - [Active] Longer count times for interrogation source
  - [Shuffler] Careful mechanical design and many count cycles minimize source placement problems.
Measurement Bias in Neutron NDA

- **Origins**
  - Calibration uncertainty
  - Chronic sample mis-placement in measurement well
  - Altered neutron backgrounds (singles and coincidence)

- **Strategies to minimize bias**
  - Calibrate often and with high precision.
  - Background/source control
Measurement Control for Neutron NDA

- Philosophy — still the same as for $\gamma$-ray measurements
  - Periodically check quality of data and instrument.
  - Periodically measure measurement control standard.
  - Check precision by repeated measurements of standard.
  - Check bias by verification of average standard value against declared value.

- Measurement Control standards for neutron NDA:
  - Stable SNM (encapsulated metal, sealed oxide, …)
  - $^{252}$Cf source (fission neutrons, decay rate known)
  - [AWCC] AmLi source built in + SNM sample
  - [Shuffler] Calibrated standard
The active Neutron Coincidence Collar is shown measuring a LWR fuel assembly. The collar surrounds the assembly on 3 sides with $^{3}$He tubes and functions like a conventional coincidence counter. On the 4th side of the assembly, an AmLi random neutron source is positioned to interrogate the sample. The collar operates like an AWCC, with the assay result giving fissile content per unit length along the fuel assembly. For MOX fuels, the collar can be operated in both active and passive mode. Also, the collar can also be used in a passive mode for $^{238}$U.
Neutron NDA Summary

- Neutron NDA Techniques:
  - Passive coincidence, active coincidence, delayed n
  - Absorption, multiplication, moderation corrections can be important
  - \((\alpha, n)\) interactions can affect results strongly (background, bias)
  - Appropriate samples: minimum hydrogen (moderator); passive - Pu, active - U; high-sensitivity (waste) - passive or delayed n.
  - Calibration curves of families of sample types recommended

- Accuracies Possible:
  - 1% to 50%, depending on sample composition and technique
  - Best performance on dry, dilute samples (no moderation, multiplication)
  - Worst cases: samples with varying moisture, moderation, matrix.
Calorimetry
Measurement Principle:

**Determination of Heat output** of the sample, as an indication of the amount of heat-producing isotopes of SNM in the sample.
Why Calorimetry?

- The most accurate NDA measurement (<1% uncertainty) of nuclear material
- You can’t hide the heat of the material
  - No absorption, moderation, multiplication,...
- Inherently matrix independent
  - Assay result independent of material, matrix or container type
  - matrix can only affect duration of analysis
  - No physical SNM standards needed!
- NIST-traceable technique
- Most DOE facilities are using calorimetry
  - Hanford, PNL, RF, LANL, SRS, LLNL, ANL-W...
Uses of Calorimetry

- Accountability Measurements
- Verification measurements
- Shipper receiver measurements
- Calibration of NDA working standards
- Measurement of biases and precisions of other NDA techniques
- Resolution of assay “outliers” and problem samples
- Benchmarking NDA measurement campaigns
- Process control measurements
- Product acceptance measurements

Considered the “gold standard” of NDA measurements throughout the complex
Relative Thermal Powers

$^{238}\text{Pu} \ (0.568 \text{ W/g})$

tritium $\ (0.324 \text{ W/g})$

plutonium $\ (0.002 - 0.014 \text{ W/g})$

human $\ (0.001 - 0.002 \text{ W/g})$

$^{233}\text{U} \ (0.00028 \text{ W/g})$

$^{235}\text{U} \ (93\% \ ^{235}\text{U}-235, \ 1\% \ ^{234}\text{U}, \ 2 \times 10^{-6} \text{ W/g})$

TRU limit $\ (100 \text{ nCi/gram}, \ 3 \times 10^{-9} \text{ W/g})$

Current calorimeter capability @ 1% accuracy
Fundamental Measurement Characteristics

- Precise NDA measurements based on heat output of sample
  - measurement accuracy of 0.1% to 0.5% for most cases
  - best suited for plutonium samples
- Sealed containers
- Assay results independent of matrix material
- Requires isotopics to convert power to grams
  - isotopics from: Gamma ray (NDA) or Chemistry (DA)
- Integrates over total sample volume
- Faster than DA, slower than NDA (γ or n)
# Half-lives and Specific Powers

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life (years)</th>
<th>Specific Power (Watts/gram)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-238</td>
<td>87.74 \text{04}</td>
<td>0.56757 \text{26}</td>
</tr>
<tr>
<td>Pu-239</td>
<td>24119 \text{26}</td>
<td>0.0019288 \text{03}</td>
</tr>
<tr>
<td>Pu-240</td>
<td>6564 \text{11}</td>
<td>0.0070824 \text{20}</td>
</tr>
<tr>
<td>Pu-241</td>
<td>14.348 \text{22}</td>
<td>0.003412 \text{02}</td>
</tr>
<tr>
<td>Pu-242</td>
<td>376300 \text{900}</td>
<td>0.0001159 \text{03}</td>
</tr>
<tr>
<td>Am-241</td>
<td>433.6 \text{14}</td>
<td>0.1142 \text{05}</td>
</tr>
<tr>
<td>H-3</td>
<td>12.3232 \text{43}</td>
<td>0.3240 \text{09}</td>
</tr>
</tbody>
</table>
Calorimeters measure the flux through heat sensors of thermal energy generated by radioactive decay

\[ \frac{dQ}{dt} = \frac{(T_{\text{cal}} - T_{\text{env}})}{R_{\text{Th}}} \]

- Heat Flux Sensors
- Heat-Generating Sample Material
- Insulation
- Sample Chamber
- \( T_{\text{cal}} \)
- \( T_{\text{env}} \)
- \( R_{\text{Th}} \) = Thermal Resistance
Twin Bridge Schematic

Identical thermal resistance

Temperature sensors

Thermal Resistance of ends increased relative to sides

$T_{\text{cal}}$

$T_{\text{env}}$

Reference

Sample
Why Twin Design?

![Graph showing signal level over time for Side One, Side Two, and their difference. The x-axis represents time in minutes, ranging from 0 to 60, and the y-axis represents signal level in microvolts, ranging from -150 to 200. The graph includes data points for each side and their difference over time.]
Resistance change measured by Wheatstone Bridge
Wheatstone Bridge Calorimeter

Final equilibrated value for 0.45 watt standard
Calorimeter sensor output independent of matrix
Calorimetric Assay of SNM (mass)

- Equation for total SNM mass

\[
\text{Mass (grams)} = \frac{\text{Thermal Power (Watts)}}{P_{\text{eff}} \text{ (Watts/g)}}
\]

- Measure isotopic composition of item (high resolution gamma spectroscopy or mass spectroscopy) and compute Effective Specific Power, \( P_{\text{eff}} \)

\[
P_{\text{eff}} \text{ (Watts/g)} = \sum P_i \times f_i
\]

- \( P_i \) = Isotope specific power (Watts/g)
- \( f_i \) = Isotopic fraction in sample, relative to plutonium
- \( i \) = all isotopes present (i.e. \(^{238}\text{Pu},^{239}\text{Pu},...,^{242}\text{Pu},^{241}\text{Am}\))
Effective Specific Power:

The rate of energy emission per unit mass of plutonium at the time of measurement.

\[ P_{\text{eff}} = \sum_{i=1}^{n} R_i P_i \]

Where:
- there are \( n \) isotopes in the sample
- \( R_i \) is the mass ratio of each isotope present
- \( P_i \) is the specific power of each isotope

• Isotopic information not required for monoisotopic items (e.g. \(^3\)H or \(^{241}\)Am)

\(^{238}\)Pu and \(^{241}\)Am can be important because of their very high specific heat values. \(^{239}\)Pu can be important because of it’s high abundance in most samples.
Calibration and Standards

- No SNM standards representative of material type are needed
  - Electrical standards
  - Heat standards
- HEAT output is measured, and heat is not absorbed or otherwise lost, unlike neutron or gamma NDA methods.
Situations that degrade Cal/Iso measurement performance

- Distortions to gamma-ray isotopic assay
  - Separated Am-241 and Pu, each in different matrices
  - Gamma-ray interferences
  - Pu-241/ U-237 equilibrium
  - Inhomogeneous isotopic distribution

- Distortions to calorimetry
  - Chemical reactions
  - Power emitting isotopes with no gamma-ray [e.g., $^{90}\text{Sr}$, $^3\text{H}$]
Calorimeter Performance

- **Plutonium:**
  - 0.2% to 0.5% on power measurements
  - 0.5% to 1.0% on specific power

- **Bulk HEU (multi-Kg):**
  - 1 to 3% on power measurements
  - 0.5 to 1.0% on specific power

- **Measurement time:**
  - 20 min. preconditioned high conductivity sample
  - 2-4 hours typical samples
  - 8-12 hours insulated samples (e.g., salts)
Appropriate Samples For Calorimetry

- Measurable heat outputs
  - Pu samples
  - High-mass U samples
  - Tritium

- Good heat-transfer characteristics
  - Time to thermal equilibrium
  - Throughput considerations
Calorimetry equipment

Calorimeter used for standards verification

Antech Inc. calorimeter

LANL-customized calorimeter
Calorimetry NDA Summary

- **Calorimetry NDA Technique**
  - Measure heat output from the sample
  - Measure specific power of the sample
  - Appropriate samples: heat-emitting (Pu, large U, T)

- **Accuracies Possible** [rivals chemistry]
  - Power: 0.1% to 0.5% (1-8 hr assay times)
  - Specific power (0.5% to 1%)
  - Best performance on isotopically homogeneous samples
  - Most useful on heterogeneous NM samples
Advanced Fuel Cycle Safeguards

Michael Miller
GNEP Safeguards Campaign Director
Los Alamos National Laboratory

U.S. Nuclear Regulatory Commission
Rockville, MD
June 10, 2008
Outline

- Introduction and Current Technical Basis
- Reprocessing
- Fuel Fabrication
- Fast Reactor
Safeguards for Advanced Fuel Cycle Facilities
Safeguards Applications in Advanced Facilities

UREX+ ⇔ Rokkasho Reprocessing Plant (RRP)

AFCF ⇔ MOX Fuel Fab Plant (PFPF & JMOX)

ABR/ABTR ⇔ Fast Breeder Reactors (Joyo & Monju)
Reprocessing
Rokkasho Reprocessing Plant (RRP)

Features common to UREX
- Spent fuel storage pool
- Head-end shear and dissolver
- Input accountability tank (IAT)
- Mixture with Pu in product
- High level liquid waste (vitrification)
- Solid wastes

Safeguards Features
- Remote, unattended monitoring
- Near real-time accounting (NRTA)
- Process monitoring
- Data authentication
- Containment and surveillance
- On-site inspector presence
- Approx. 20 different SG systems
NDA Systems Installed Throughout the Process

- ISVS – Integrated Spent fuel Verification System
- IHVS – Integrated Head end Verification System
- RHMS – Rokkasho Hulls Measurement System
- VCAS – Vitrified waste Canister Assay System
- HKED – Hybrid K-Edge Densitometer
- TCVS - Temporary Canister Verification System
- iPCAS - improved Plutonium Canister Assay System
- WCAS A/B - Waste Crate Assay System
Head-End Fuel Assembly Verification Camera & Radiation Detector (CRD)

- Verification of LWR fuel assemblies inside shearing cell
- Penetrates the 2-meter thick concrete shielding wall
- Shielded detector head includes cameras, neutron and gamma sensors
- Provides ID, type fuel, and shearing history
- Continuous operation by IAEA and Operator
Verify Quantity of Pu in Waste Streams

Vitrified Waste canister

Fission Chamber
Neutron Monitors

Los Alamos
NATIONAL LABORATORY
EST.1943

NRC Seminar June 10, 2008
Example of impact of new safeguards measurement technologies: improved Plutonium Canister Assay System (iPCAS)

Installed in RRP
March 2004:
decreased the uncertainty of each measurement (36kg containers of MOX) by ~200g Pu
Fuel Fabrication
Plutonium Fuel Production Facility (PFPF)

Safeguards Features:
- Measurement and monitoring of MOX feed, process, and product
- Glove-box holdup measurements
- NRTA and remote/unattended systems
- Containment/surveillance systems
- ~ 11 different SG systems at PFPF
NDA Systems Installed Throughout the Process

Feed storage  Process  Product storage

Receipt

Powder  Powder  Pellet  Pin  Assembly  Shipment

PCAS

INVS  HRGS  PSMC

Analysis

Waste

FPAS

WDAS  WCAS

FAAS

NRC Seminar June 10, 2008
Input MOX verification (PCAS), Process Holdup (GBAS)

Measure PuO\textsubscript{2} & MOX into facility

Process Line Holdup Detectors

Features
* Installed as part of facility robotics transfer system
* Continuous and unattended mode operation
Fuel Pin Measurement (FPAS), Fuel Assembly (FAAS)

- Measure MOX fuel pins during fabrication
- Measure MOX product assemblies leaving facility
Fast Reactor
Fast breeder reactors in Japan under full-scope IAEA safeguards

Safeguards Features:
- Fresh fuel assembly input gate monitors
- Reactor head seals and surveillance
- Reactor reload monitors and surveillance
- Fresh and spent fuel storage monitors
- Spent fuel discharge monitors
Two detectors monitoring top of CORE

Surveillance camera inside containment
Two detectors monitoring refueling machine

One detector monitors transfers between storage and refueling machine
Entrance Gate Monitor, Seals

Entrance gate monitor

Seals on fresh fuel storage

Fresh fuel storage

Seal on fresh fuel storage room access portal
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