Metallic Fast Reactor Fuels

Background

- The first fuels used for the LMR's (Liquid Metal-cooled fast Reactors) in the 50's and early 60's were metallic (EBR-I, EBR-II).

- In the late 60's, world interest turned toward ceramic fuels.

- Development of metallic fuels continued into 70's because EBR-II continued to be fueled with U-5 Fs:
  
<table>
<thead>
<tr>
<th>Element</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nb</td>
<td>0.01 %</td>
</tr>
<tr>
<td>Zr</td>
<td>0.1 %</td>
</tr>
<tr>
<td>Pd</td>
<td>0.2 %</td>
</tr>
<tr>
<td>Rh</td>
<td>0.3 %</td>
</tr>
<tr>
<td>Ru</td>
<td>1.9 %</td>
</tr>
<tr>
<td>Mo</td>
<td>2.4 %</td>
</tr>
<tr>
<td>U</td>
<td>95 %</td>
</tr>
</tbody>
</table>

- Events in the 80's caused a reassessment of reactor technology

  1.) Cancellation of CRBR  
     *(fuel cycle costs)*  
  2.) Three Mile Island/Chernobyl  
     *(Public Safety Demands)*  
  3.) Radioactive Waste "logjam"

- 1983 IFR (Integral Fast Reactor) Concept Start
The Integral Fast Reactor (IFR)

- Na Cooled Fast Reactor
  - Ambient-pressure cooling system

- Metallic Fuel (U-Pu-Zr)
  - High thermal conductivity
  - Superior compatibility with coolant

- Innovative Process for Recycling Fuel
  - Pyrometallurgical processing ("pyroprocessing")
  - Simple, compact, economical process

- Passively Inherently Safe
  - Safe shutdown relies only on laws of physics
  - No complicated engineered safety systems
  - Long times available for operator response

- Over 29 y of Operating Experience With the IFR Prototype, EBR-II
  - High capacity factor, over 75%
  - Low personnel exposures
  - No component failures
IFR Fuel Cycle

IFR

Spent Fuel → Pin Chopper → Fuel Pin Segments → Electro-Refiner

Salt, Cd, TRU → Waste Treatment → Solid Waste → Geologic Disposal

Fresh Fuel → Fuel Refabrication

Fuel Slugs → Injection Casting → Metal Ingot → Cathode Processor

U, TRU
Advantages of the IFR Concept

• Improved Reactor Safety

- Proven passively inherently safe

On 4/3/86 reactor shutdown w/o operator or mechanical intervention in two tests:

1.) Loss of flow without scram from full power(simulated conditions in Chernobyl accident)

2.) Loss of heat sink without scram from full power (simulated conditions existing in TMI-2)

- In both tests, inherent feedbacks enabled the reactor to respond to the abnormal events and return to a safe and coolable state

1.) Thermal expansion of the core
2.) Doppler reactivity feedback

- Atmospheric pressure of primary coolant

- Large thermal inertia of Na pool

- High thermal conductivity of metallic fuel

1.) Low fuel temperature
2.) less stored energy

-Large margin between operating temperature (340-510 °C) and Na boiling temperature ( 900°C)
Advantages of the IFR Concept (cont.)

• Improved Nuclear Waste Management
  
  - Actinide elements absent from high-level waste produced
  - Capability to recycle LWR spent fuel
  - Reduces waste volume

• Efficient Utilization of Fuel Resources
  
  - Initial plants will be fissile self sufficient
  - Later plants can be operated as Pu breeders

• Potential Economic Parity With Other Energy Sources
  
  - Limited safety-grade construction
  - Very long plant life (low pressure, low corrosion)
  - Reduced fuel cycle costs via reprocessing
  - Flexible deployment: large or small, modular plants

• Proliferation Resistant
  
  - No separation of Pu (tied up with U and non-fissile actinides)
  - Fuel processed and refabricated remotely due to presence of fission products
INCENTIVE FOR ACTINIDE REMOVAL

RELATIVE RADIOLOGICAL RISK

ACTINIDES

NATURAL U ORE

FISSION PRODUCTS

YEARS

10^0 10^1 10^2 10^3 10^4 10^5 10^6 10^7 10^8
The Integrated Power From Pu in LWR Fuel Elements Can Approach 40% by 40 GWD/MTU.
IFR Operations Proven in EBR-II

- Personnel exposure is 1-2% of LWR's

- EBR-II annual capacity factor (75-80%) over the average for operating commercial plants in the U.S. (~70%)

- EBR-II steam generators have operated without leaks for over 25 years of continuous service

Metal Fuel is the Foundation of the IFR Concept

- Key factor contributing to passive safety characteristics

- Metal fuel fabrication is simple and compact

- Compact, simple pyroprocessing of metallic fuel promises dramatic improvements in fuel cycle economics

- Pyroprocessing facilitates significant improvements in waste management
Performance of IFR Fuel Has Been Demonstrated Successfully

- Ongoing tests of U-Pu-Zr and U-Zr fuels have now achieved burnups of 20 a/o, well in excess of their design target burnup level of 100,000 MWd/T (10 a/o burnup), assuring excellent fuel cycle economics.

- Metal assemblies have been operated for up to 223 days beyond cladding failure without any degradation, providing utility operators with assurance of reliable, efficient plant operation.

- EBR-II was fully converted for operation with the IFR-type fuel alloys (U-Zr and U-Pu-Zr).
Terminology

• Pyroprocessing:
  *Pyrometallurgical and electrochemical processing*

• Key Step: Electrorefining
  Electrotransport in a molten salt (LiCl-KCl) electrolyte

• Electrorefining:
  Metal is electronically dissolved at an anode made of impure metal and re deposited at a cathode in a condition of greater purity

  Anode: \( M \rightarrow M^{+3} + 3e^- \)
  Cathode: \( M^{+3} + 3e^- \rightarrow M \)

Chemical Basis of Pyroprocessing

• Separations based on the relative ease of oxidation into a molten salt

  -Free energy of formation of metal chloride is primary determinant of ease of oxidation

• Some Separations are Chemically Complete

  -Halides remain in salt as anions
  -Alkali metal and alkaline earth metals (plus Sm and Eu) are completely oxidized and remain in salt
  -Noble metals not oxidized; remain as metals
• Actinide and Rare Earth Metals
  Partition Between Salt and Metal Phases
  -Can be transferred to salt by oxidation, or to metal by reduction

Pyroprocess Chemistry

• Treat as a Series of Equilibrium Reactions

\[ M + M' \text{Cl}_3 \leftrightarrow M' + M\text{Cl}_3 \]

• \( \Delta G = \text{Free Energy Change} \)

\[ \Delta G = \Delta G_f^0 (\text{MCl}_3) - \Delta G_f^0 (M'\text{Cl}_3) \]

• \( K_{eq} = \exp \left( \frac{-\Delta G}{RT} \right) = \frac{a_{M'}a_{\text{MCl}_3}}{a_Ma_{M'\text{Cl}_3}} \)

Process is Controlled by Adjusting Redox State of Electrorefining Cell
Free Energies of Formation

\[ -\Delta G_f^0 \text{ in kcal/ g-mole equiv. Cl @ 500°C} \]

<table>
<thead>
<tr>
<th>Elements that remain in salt (very stable chlorides)</th>
<th>Materials that can be electrotransported efficiently</th>
<th>Elements that remain in Cd pool as metals (less stable chlorides)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaCl(_2) 87.9</td>
<td>CmCl(_3) 64.0</td>
<td>CdCl(_2) 32.2</td>
</tr>
<tr>
<td>CsCl 87.8</td>
<td>PuCl(_3) 62.4</td>
<td>FeCl(_2) 29.2</td>
</tr>
<tr>
<td>RbCl 87.0</td>
<td>AmCl(_3) 62.1</td>
<td>NbCl(_5) 26.7</td>
</tr>
<tr>
<td>KCl 86.7</td>
<td>NpCl(_3) 58.1</td>
<td>MoCl(_4) 16.8</td>
</tr>
<tr>
<td>SrCl(_2) 84.7</td>
<td>UCl(_3) 55.2</td>
<td>TcCl(_4) 11.0</td>
</tr>
<tr>
<td>LiCl 82.5</td>
<td>ZrCl(_2) 46.6</td>
<td>RhCl(_3) 10.0</td>
</tr>
<tr>
<td>NaCl 81.2</td>
<td></td>
<td>PdCl(_2) 9.0</td>
</tr>
<tr>
<td>CaCl(_2) 80.7</td>
<td></td>
<td>RuCl(_4) 6.0</td>
</tr>
<tr>
<td>LaCl(_3) 70.2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PrCl(_3) 69.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>CeCl(_3) 68.6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NdCl(_3) 67.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>YCl(_3) 65.1</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Plutonium Recovery

• Chemical Reaction at Solid Cathode

- \( \text{UCI}_3 + \text{Pu} \ (s) \rightleftharpoons \text{PuCl}_3 + \text{U} \ (s) \)
- \( \text{PuCl}_3 \) is more stable than \( \text{UCI}_3 \)
- Deposition of Pu is favored if

\[
\frac{[\text{PuCl}_3]}{[\text{UCI}_3]} > 10^5 \quad \text{(not practical)}
\]

• In the Presence of Cd, the Pu Chemical Activity is Greatly Lowered and It Behaves as though Its Chloride were Only Very Slightly More Stable Than \( \text{UCI}_3 \)

• Chemical Reaction-Liquid Cd Cathode

- \( \text{UCI}_3 + \text{Pu} \ (\text{Cd}) \rightleftharpoons \text{PuCl}_3 + \text{U} \ (\text{Cd}) \)
- Deposition of Pu is favored only if

\[
\frac{[\text{PuCl}_3]}{[\text{UCI}_3]} > 2
\]

- Deposition of TRU elements occurs as the intermetallic compound; e.g., \( \text{PuCd}_6 \)

- U also deposits in a quantity roughly equal to the TRU elements
ADVANTAGES OF PYROPROCESSING

- Simple, compact system
  - Low capital and operating costs

- Small high-level waste volumes
  - About 300-500 liters (0.3-0.5 m³) per MTIHM spent fuel processed

- Limited secondary wastes
  - Only contaminated equipment, tools, other indirect process wastes such as gloves, rags, etc.

- Actinide elements virtually absent from waste streams

- Pyroprocessing can be applied to the treatment of a wide variety of spent fuel and waste types, offering a common solution to the disposition of nuclear wastes
  - Metal fuel
  - Graphite fuel
  - Pu processing scrap and waste
  - Oxide fuel
  - Naval fuel
  - Test/Research Reactor fuel