# 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**

| | |
|---|---|
| **Nb** | **0.01 %** |
| **Zr** | **0.1 %** |
| **Pd** | **0.2 %** |
| **Rh** | **0.3 %** |
| **Ru** | **1.9 %** |
| **Mo** | **2.4 %** |
| **U** | **95 %** |

- **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*

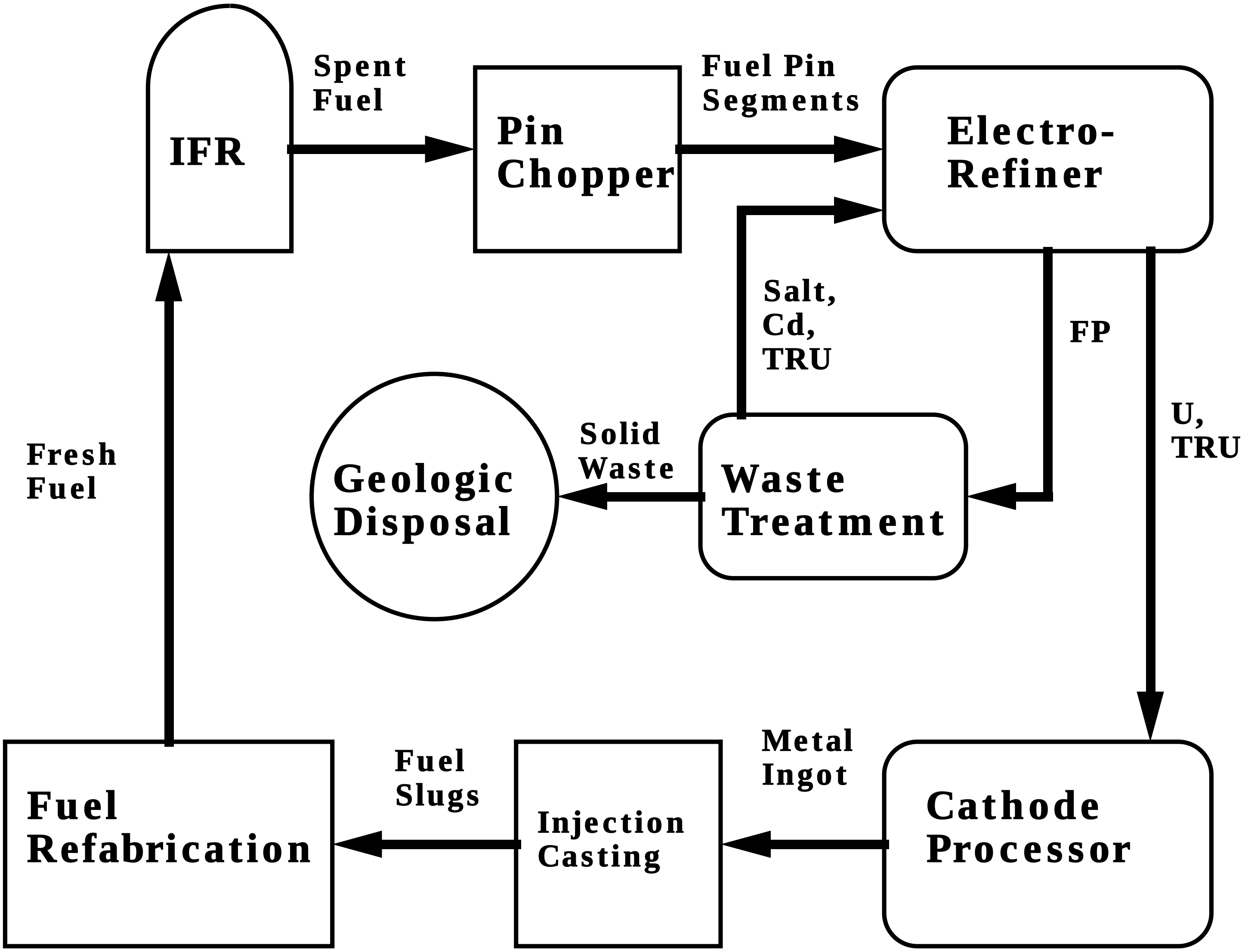
- **Passively Inherently Safe**

  *-Safe shutdown relies only on laws of physics*
  *- No complicated engineered safety systems*
  *-Long times availible foe 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



# 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.)

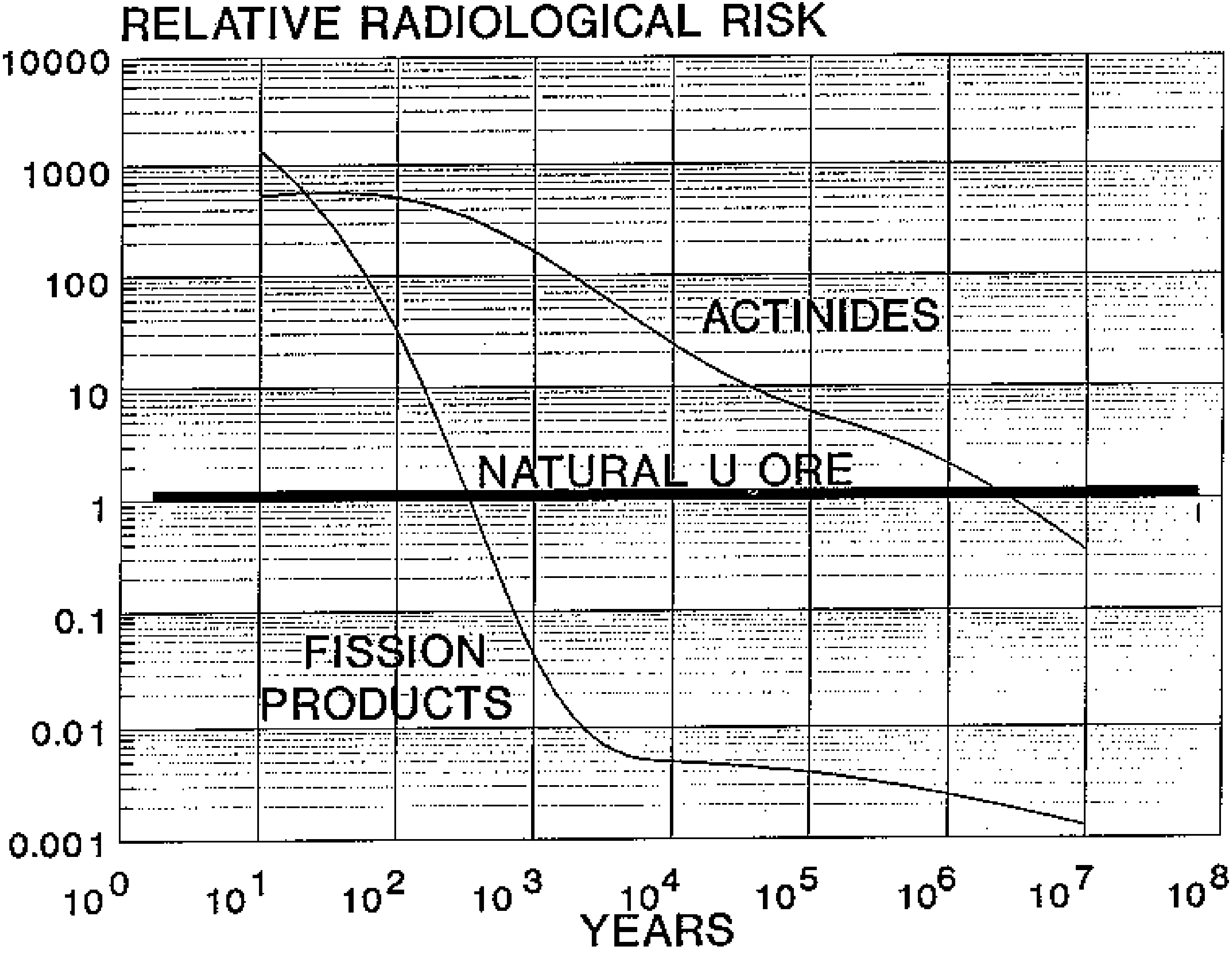
- **Improved Nuclear Waste Management**
  - *Actinide elements absent from high-level waste produced*
  - *Capability to recycle LWR spent fuel*
  - *Reduces waste volume*

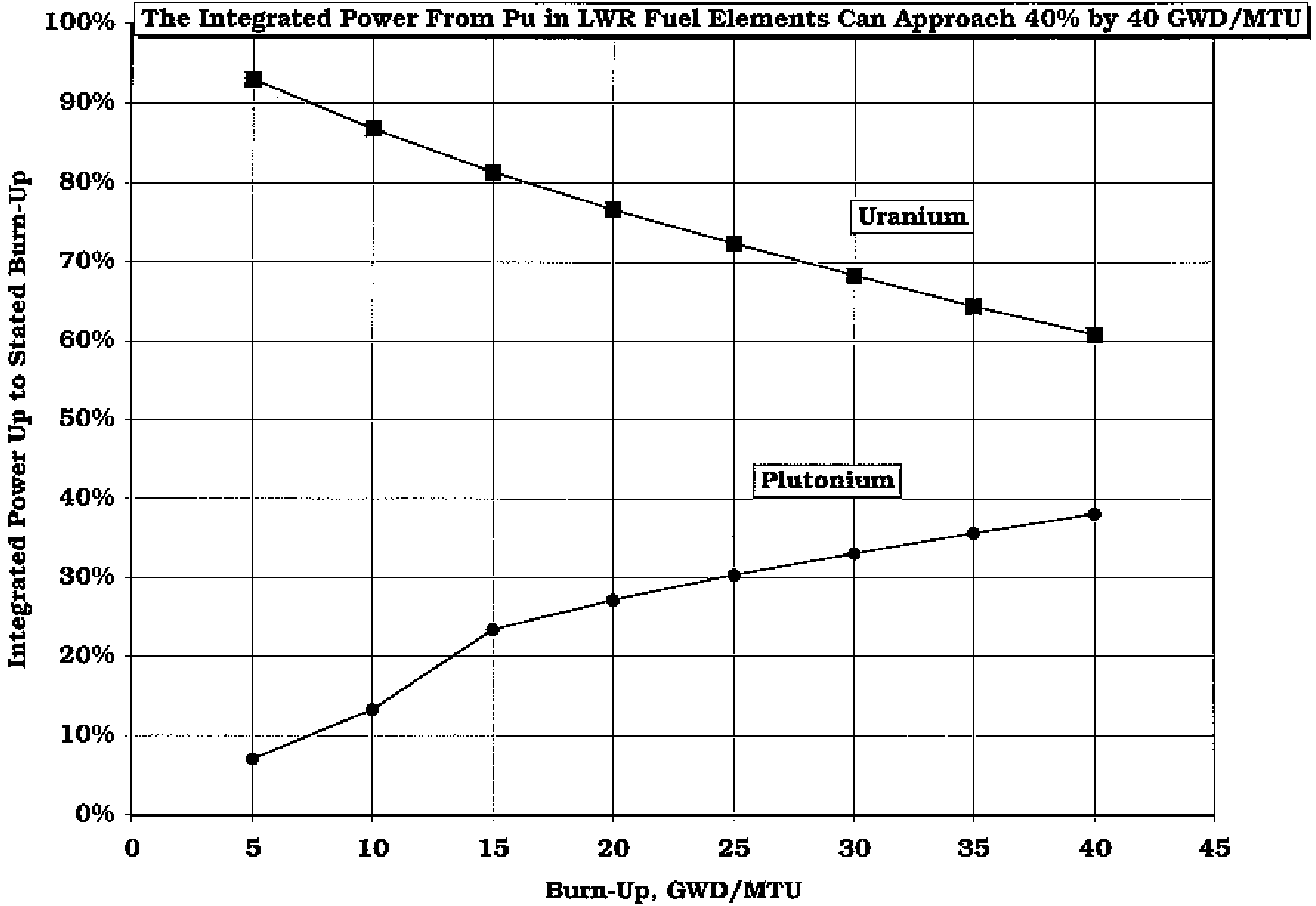
- **Efficient Utilization of Fuel Resources**
  - *Initial plants will be fissile self sufficient*
  - *Later plants can be operated as Pu breeders*

- **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




The Integrated Power From Pu in LWR Fuel Elements Can Approach 40% by 40 GWD/MTU
Integrated Power Up to Stated Burn-Up
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
Uranium
Plutonium
0
5
10
15
20
25
30
35
40
45
Burn-Up, 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:**

***Pyro***metallurgical 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:*** $M \dashrightarrow M^{+3} + 3e^-$

***Cathode:*** $M^{+3} + 3e^- \dashrightarrow M$

# 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**

$$M + M'Cl_3 \longleftrightarrow M' + MCl_3$$

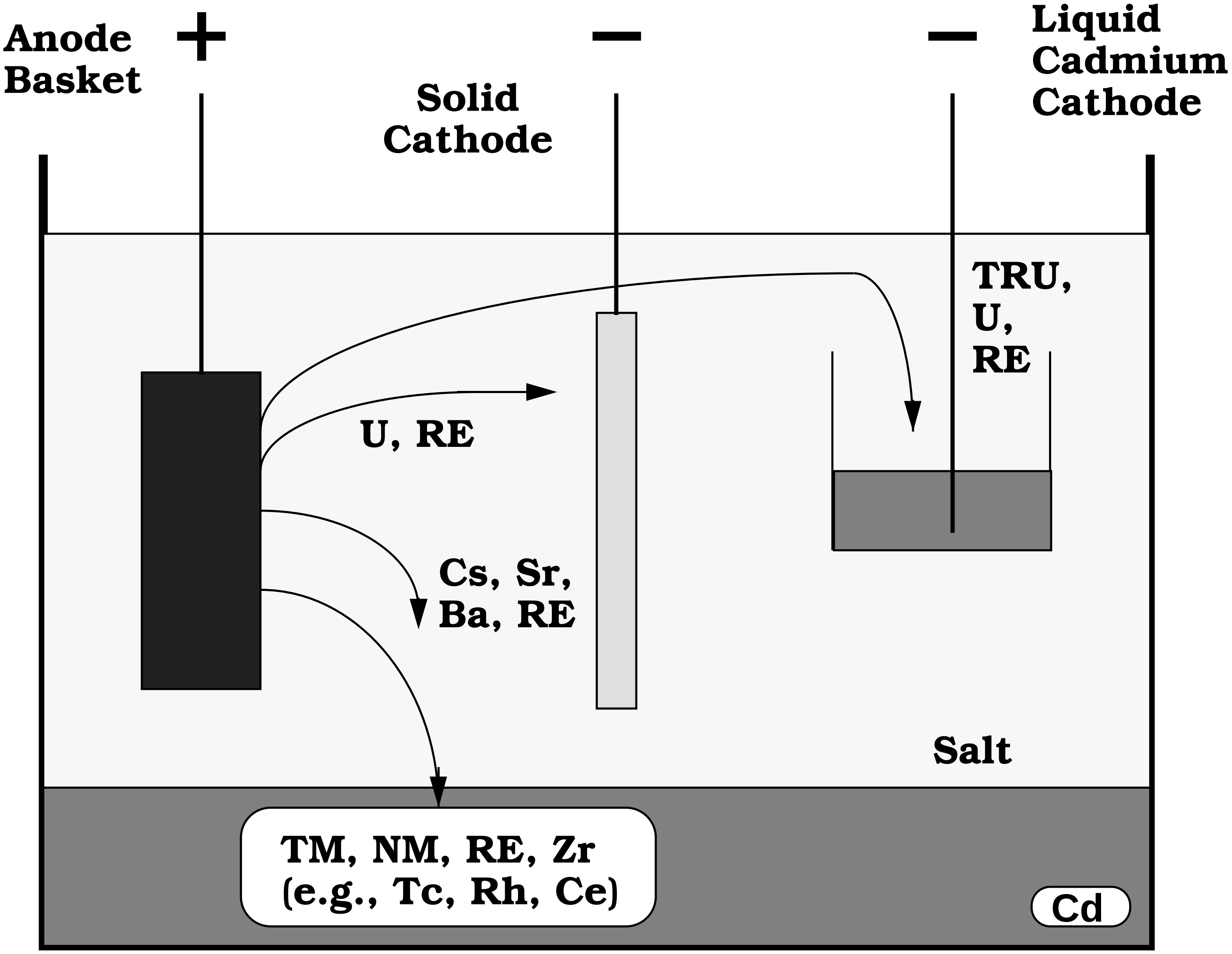
- **$\Delta G$ = Free Energy Change**

$$\Delta G = \Delta G_f^0(MCl_3) - \Delta G_f^0(M'Cl_3)$$

- $K_{eq} = \exp\left(\frac{-\Delta G}{RT}\right) = \frac{\left(a_{M'} a_{MCl_3}\right)}{\left(a_M a_{M'Cl_3}\right)}$

**Process is Controlled by Adjusting Redox State of Electrorefining Cell**

# Electrorefining-Schematic



# Free Energies of Formation

| $-\Delta G_f^0$ in kcal/g-mole equiv. Cl @ 500°C | | | | | |
|---|---|---|---|---|---|
| *Elements that remain in salt (very stable chlorides)* | | *Materials that can be electrotransported efficiently* | | *Elements that remain in Cd pool as metals (less stable chlorides)* | |
| **$BaCl_2$** | **87.9** | **$CmCl_3$** | **64.0** | **$CdCl_2$** | **32.2** |
| **CsCl** | **87.8** | **$PuCl_3$** | **62.4** | **$FeCl_2$** | **29.2** |
| **RbCl** | **87.0** | **$AmCl_3$** | **62.1** | **$NbCl_5$** | **26.7** |
| **KCl** | **86.7** | **$NpCl_3$** | **58.1** | **$MoCl_4$** | **16.8** |
| **$SrCl_2$** | **84.7** | **$UCl_3$** | **55.2** | **$TcCl_4$** | **11.0** |
| **LiCl** | **82.5** | **$ZrCl_2$** | **46.6** | **$RhCl_3$** | **10.0** |
| **NaCl** | **81.2** | | | **$PdCl_2$** | **9.0** |
| **$CaCl_2$** | **80.7** | | | **$RuCl_4$** | **6.0** |
| **$LaCl_3$** | **70.2** | | | | |
| **$PrCl_3$** | **69.0** | | | | |
| **$CeCl_3$** | **68.6** | | | | |
| **$NdCl_3$** | **67.9** | | | | |
| **$YCl_3$** | **65.1** | | | | |

# Plutonium Recovery

- **Chemical Reaction at Solid Cathode**

  *-$UCl_3$ + Pu (s) <-----> $PuCl_3$ + U (s)*

  *-$PuCl_3$ is more stable than $UCl_3$*

  *- Deposition of Pu is favored if*

$$\frac{\lfloor PuCl_3 \rfloor}{\left[UCl_3\right]} > 10^5$$

*_ (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 $UCl_3$**

- **Chemical Reaction-Liquid Cd Cathode**

  *-$UCl_3$ + Pu (Cd) <---> $PuCl_3$ + U (Cd)*

  *- Deposition of Pu is favored only if*

$$\frac{\lfloor PuCl_3 \rfloor}{\left[UCl_3\right]} > 2$$

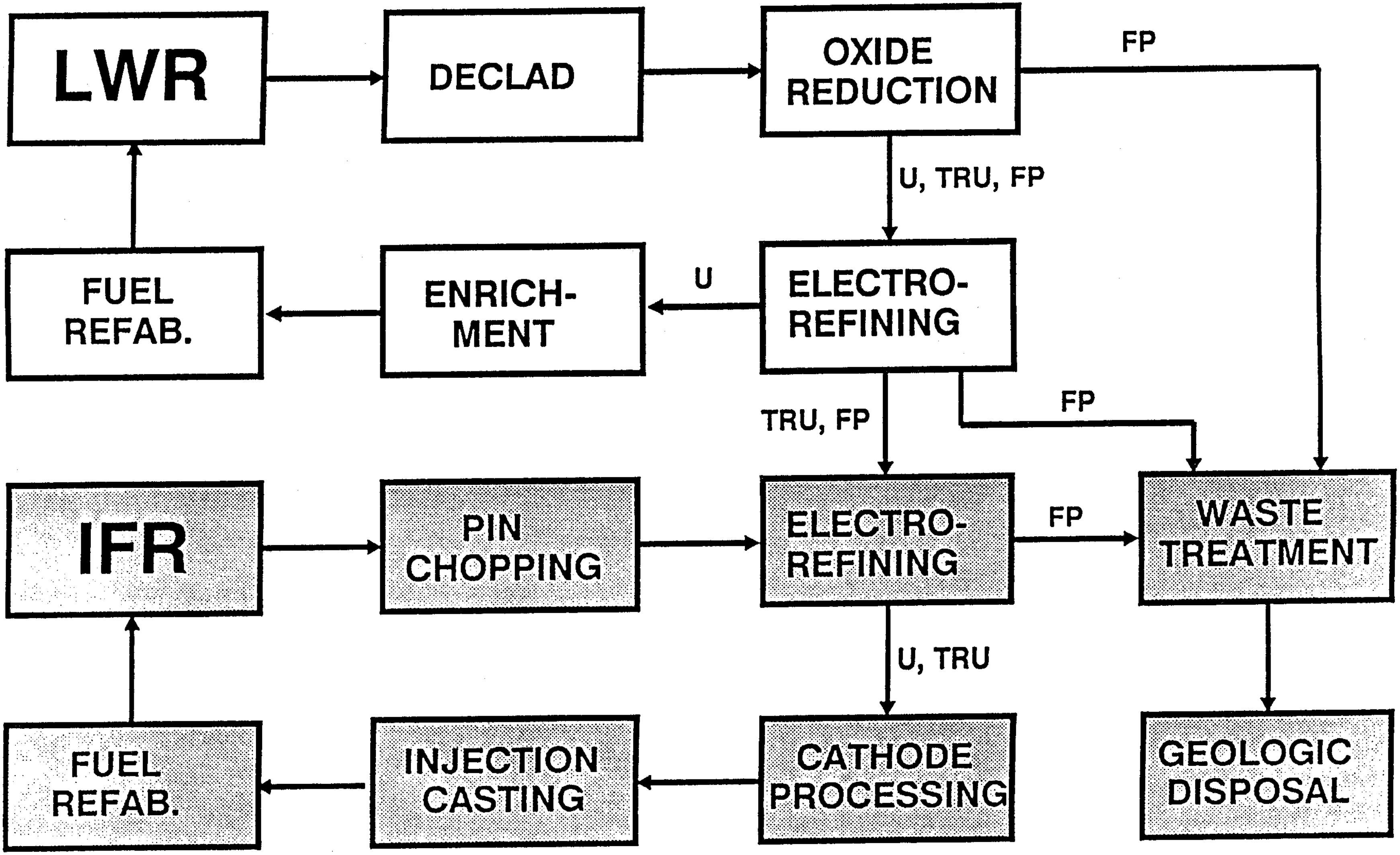
*- Deposition of TRU elements occurs as the intermetallic compound; e.g., $PuCd_6$*

*- 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 $m^3$) 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

# INTEGRATED IFR AND LWR SPENT FUEL PYROPROCESSING



LWR

DECLAD

OXIDE REDUCTION

FP

U, TRU, FP

FUEL REFAB.

ENRICHMENT

U

ELECTROREFINING

TRU, FP

FP

IFR

PIN CHOPPING

ELECTROREFINING

FP

WASTE TREATMENT

U, TRU

FUEL REFAB.

INJECTION CASTING

CATHODE PROCESSING

GEOLOGIC DISPOSAL