Overview of Pyroprocessing

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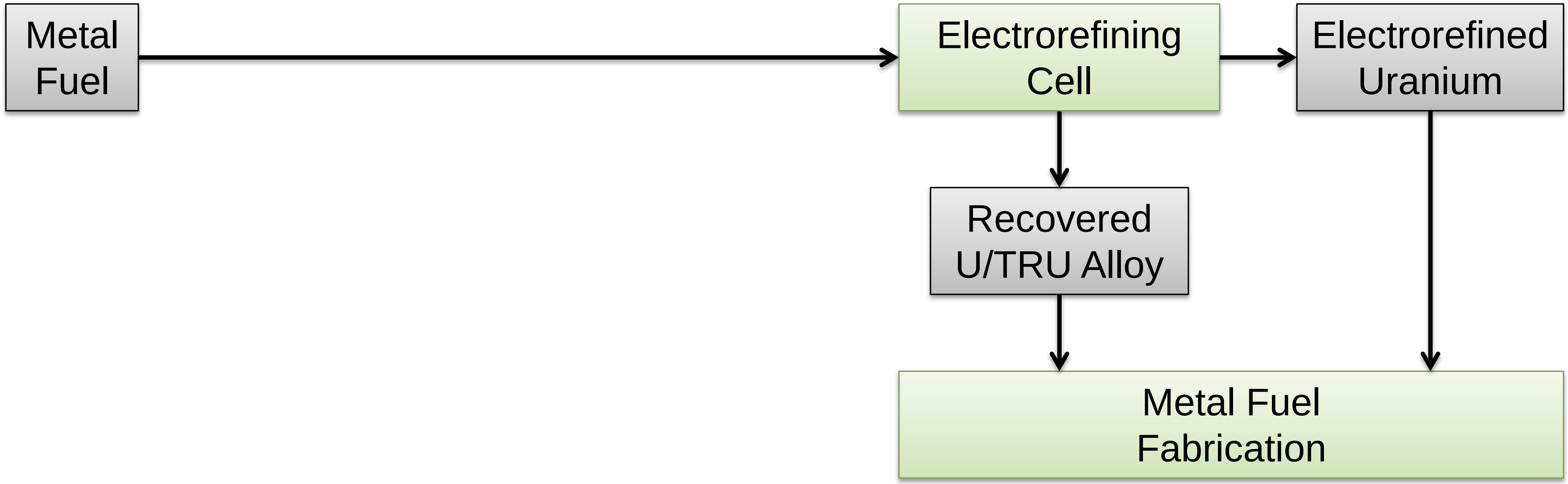
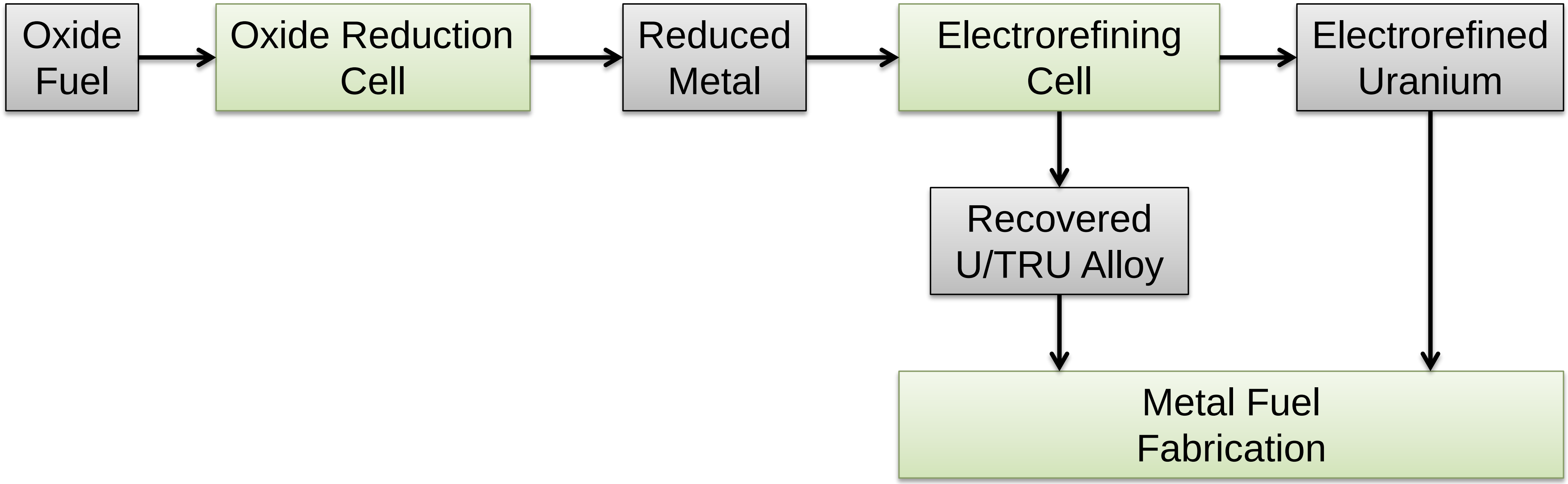
Pyroprocessing: Metal Fuels



Idaho National Laboratory

Pyroprocessing: Oxide Fuels



Pyroprocessing: Unit Operations

Head-End Operations

- Decladding
- Chopping
- Thermal Oxidation
- Sieving
- Pelletizing
- Bond-Sodium Removal

Processing Operations

- Oxide Reduction
- Uranium Electrorefining
- Transuranic Recovery
- Salt/Metal Separations
- Distillation
- Metal Casting
- Salt Management

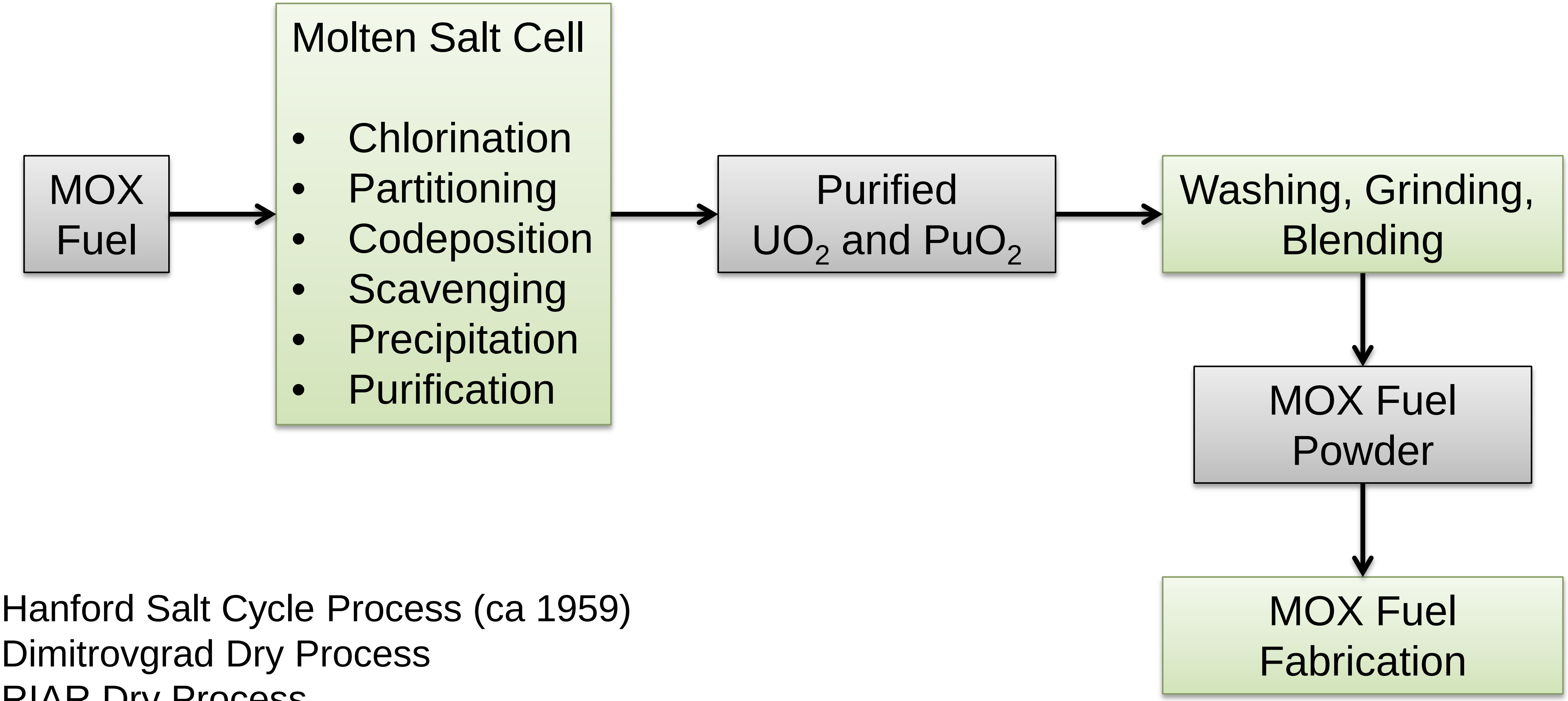
Back-End Operations

- Off-Gas Capture
- Fission Product Management
- Waste Forms
- Transportation
- Regulatory Requirements

Safeguards

- Material Control and Accountancy
- Sampling and Analyses
- Process Monitoring
- Facility Design

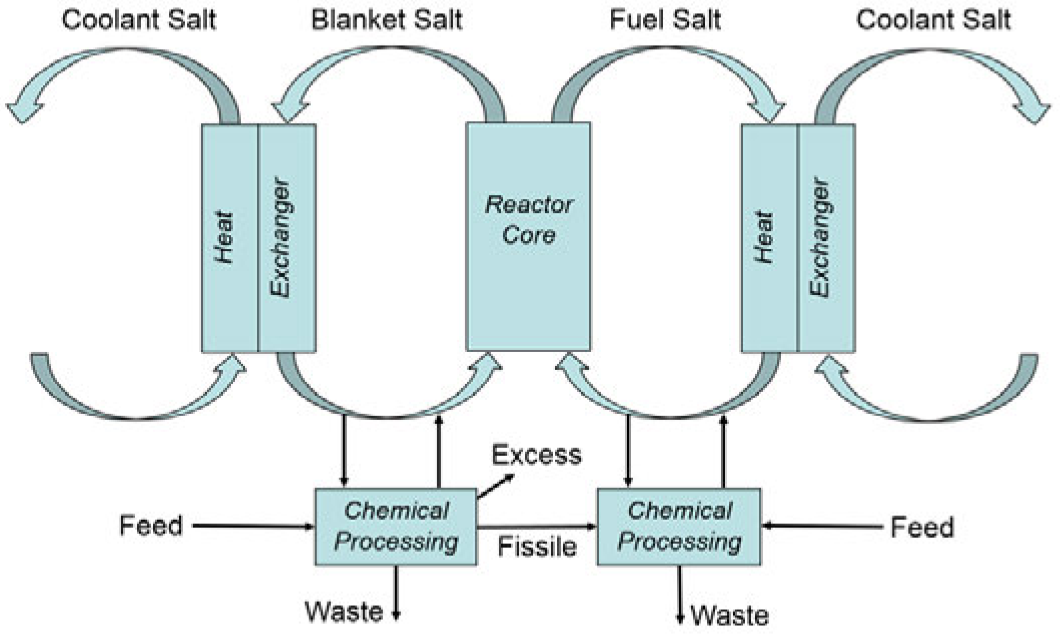
Pyroprocessing: Mixed Oxide (MOX) Fuels



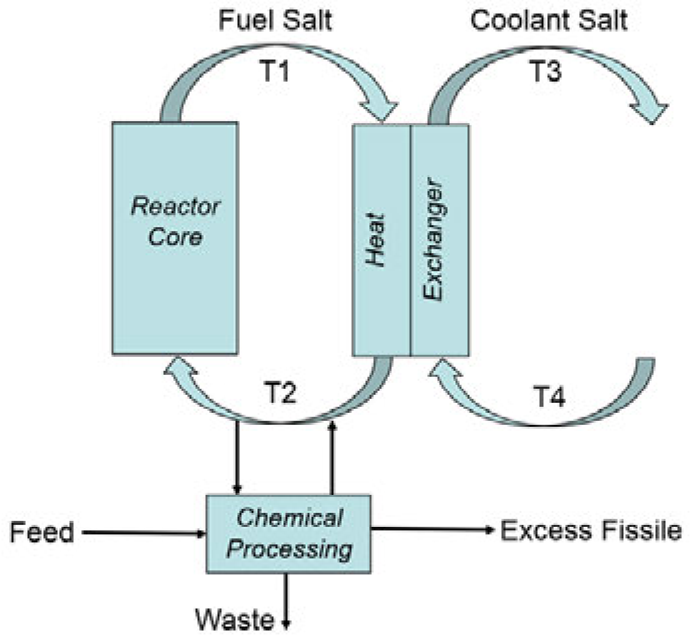
- Hanford Salt Cycle Process (ca 1959)
- Dimitrovgrad Dry Process
- RIAR Dry Process

"Pyroprocessing" to Support a $^{232}Th/^{233}U$ MSBR

Two-Fluid MSBR



Single-Fluid MSBR



Takeaway: Fuel salt processing is directly connected to reactor operations.
Primary function is Pa management.

“Pyroprocessing” to Support MSR Technologies

Head End

- Fluorination
- Chlorination
- Purification
- ^{7}Li Enrichment
- ^{37}Cl Enrichment
- Handling and Storage

Fuel Salt	Continuous Chemical Processing	Minimal Chemical Processing	No Chemical Processing
Fluoride	ORNL MSBR	---	---
Chloride	---	---	---

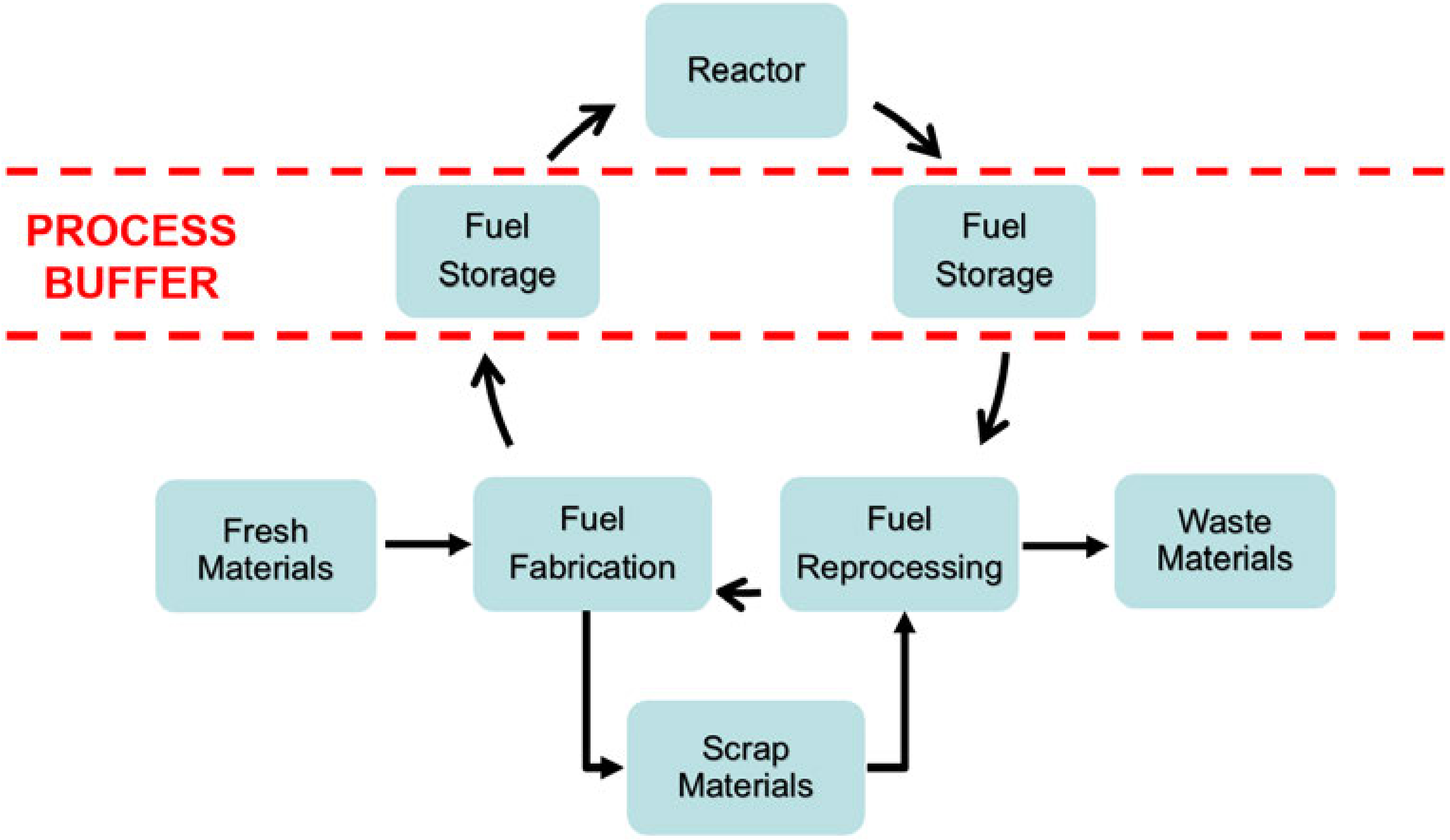
Back End

- Actinide Separations
- Fission Product Separations
- ^{7}Li Recovery
- ^{37}Cl Recovery
- Waste Form Production
- Geologic Disposition

Reactor Operations

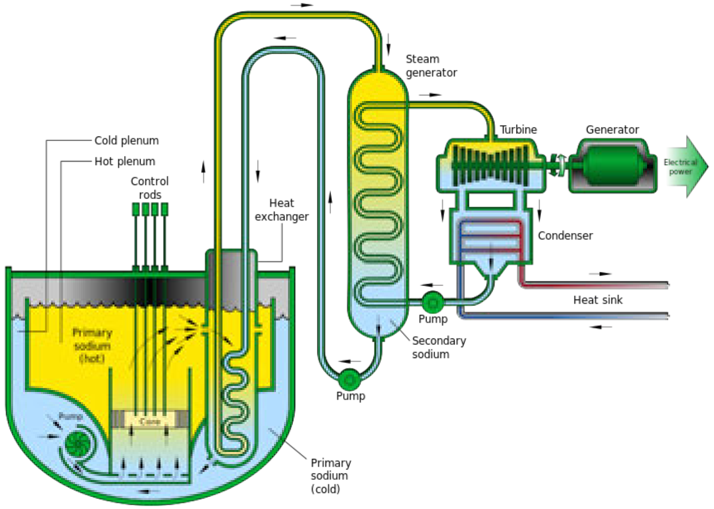
- ^{233}Pa, ^{233}U, ^{239}Pu Separations for Breeders
- Fission and Corrosion Product Management
- RedOx Measurement and Control
- Fissile Additions
- Noble Gas and Tritium Management

“Typical” Fuel Cycle



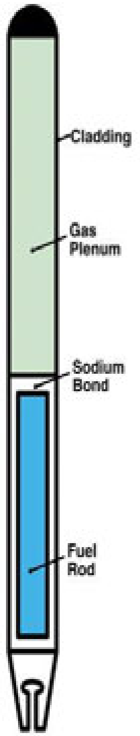
Takeaway: Fuel fabrication and fuel reprocessing are not directly connected to reactor operations. A notable exception is, in certain applications, molten salt reactors.

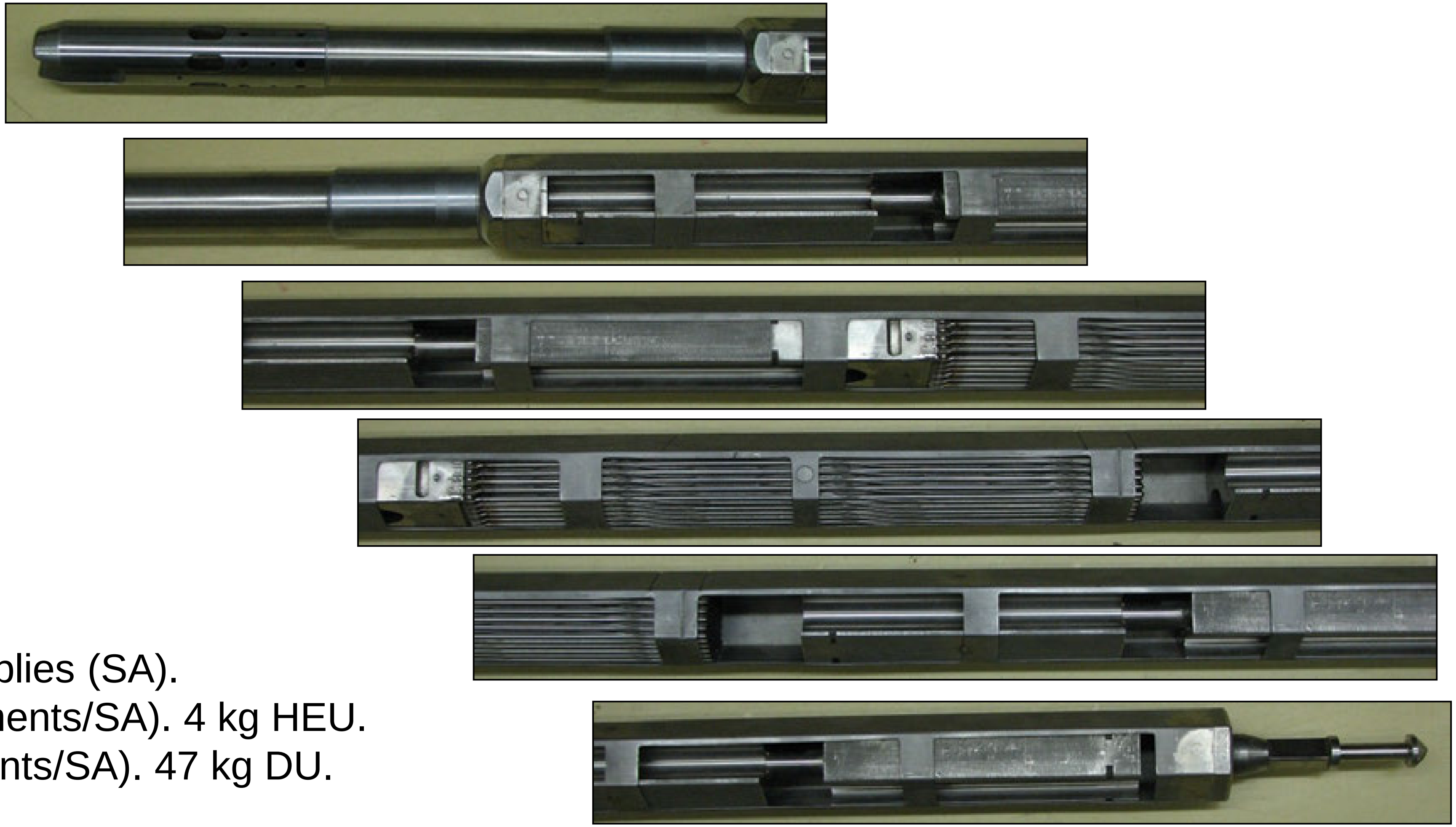
EBR-II and the Integral Fast Reactor Program



Takeaway: IFR was to demonstrate a closed fuel cycle on EBR-II using pyroprocessing.
IFR was a collaboration between US and Japan.
IFR cancelled in 1994 when EBR-II was shut down.
IFR pyroprocessing never occurred.

U.S. DOE: Integral Fast Reactor Program





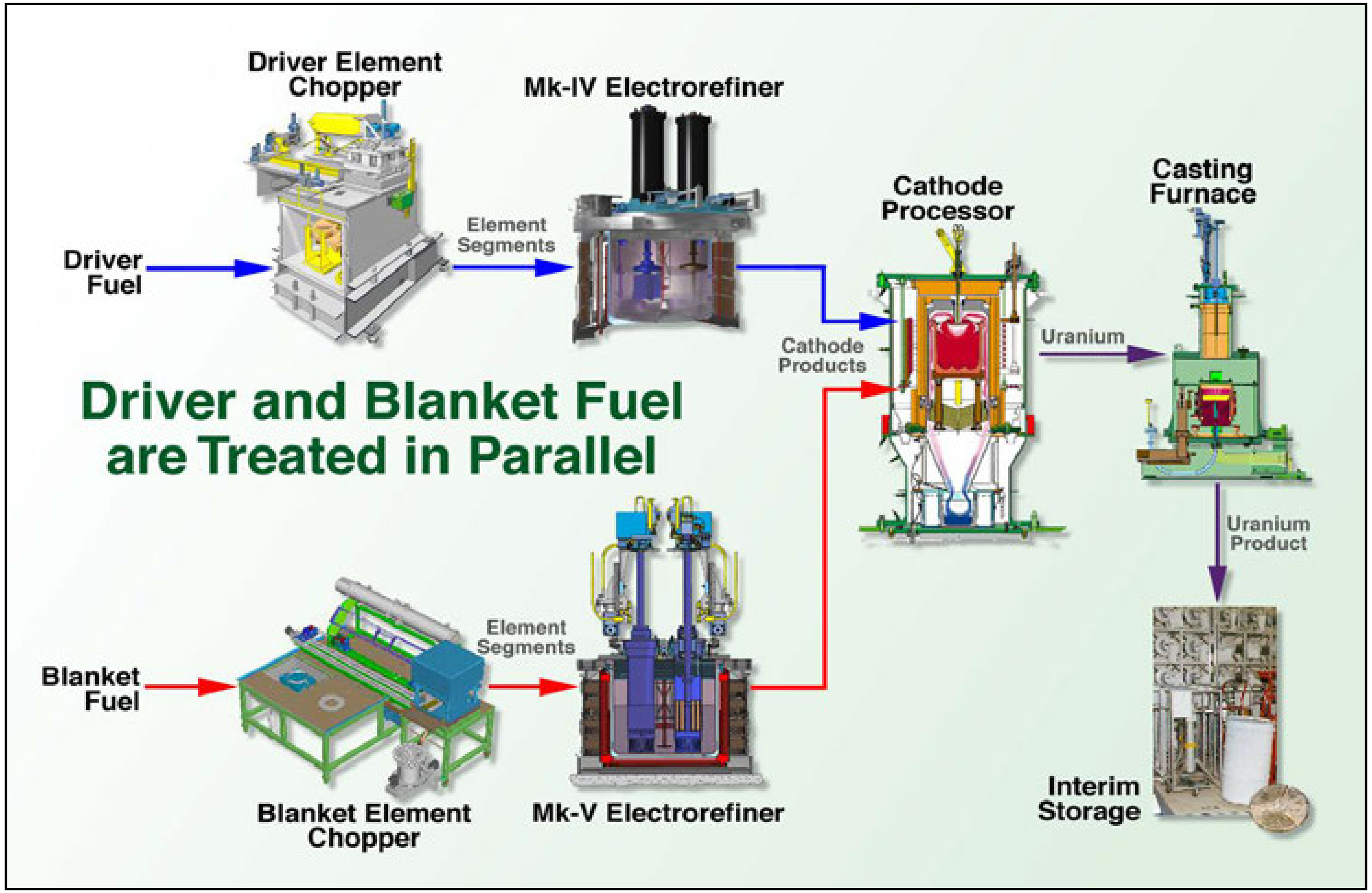
- Stainless steel subassemblies (SA).
- 67 core SA (91 or 61 elements/SA). 4 kg HEU.
- 570 blanket SA (19 elements/SA). 47 kg DU.
- 636 total SA.

Spent Fuel Treatment Program

- Driver fuel line designed for IFR.
- Blanket fuel line designed for SFT.
- Shared cathode processor.

- Driver fuel…
  - o 65 to 75% HEU.
  - o 3 MT inventory.

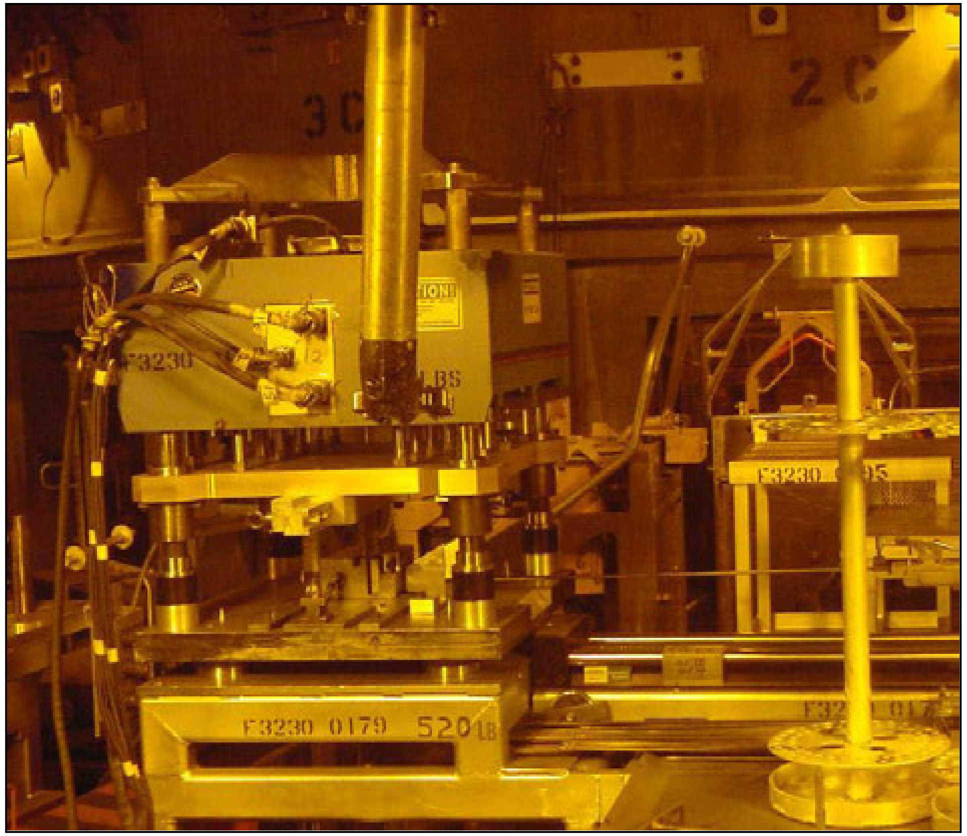
- Blanket fuel…
  - o DU w/ 1% Pu.
  - o 22 MT inventory.

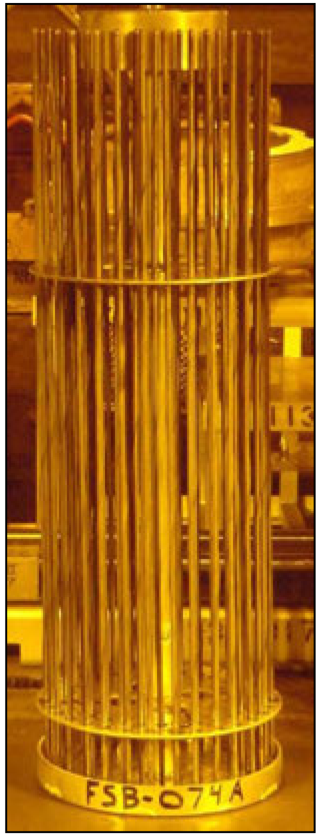


Spent Fuel Treatment Program

- Driver element chopper.
  - Five elements chopped at once.
  - 0.25-in-long segments

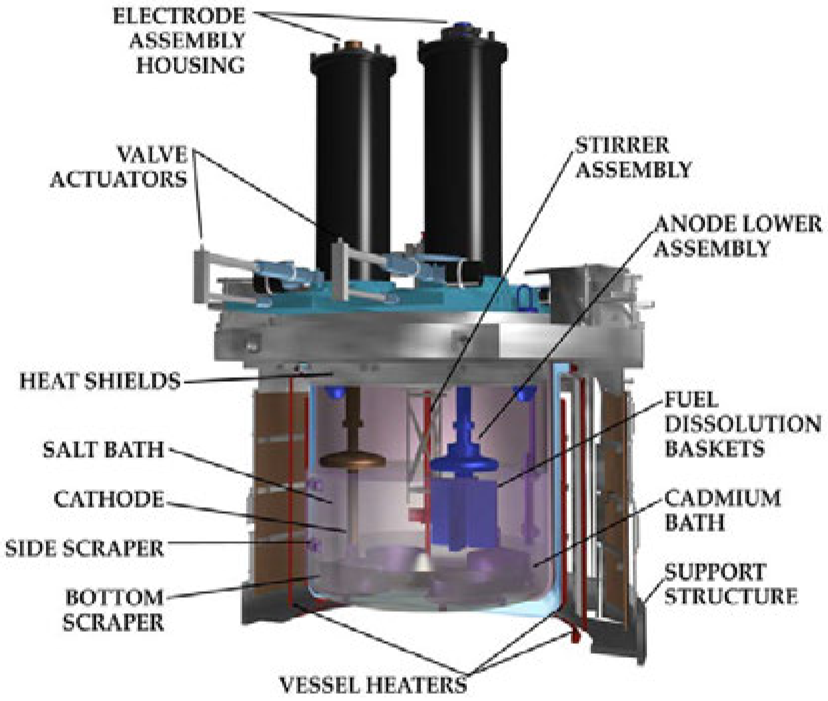
- Anode basket.
  - Holds four driver SA.
  - 16 kg HM.
  - 8,000 segments.

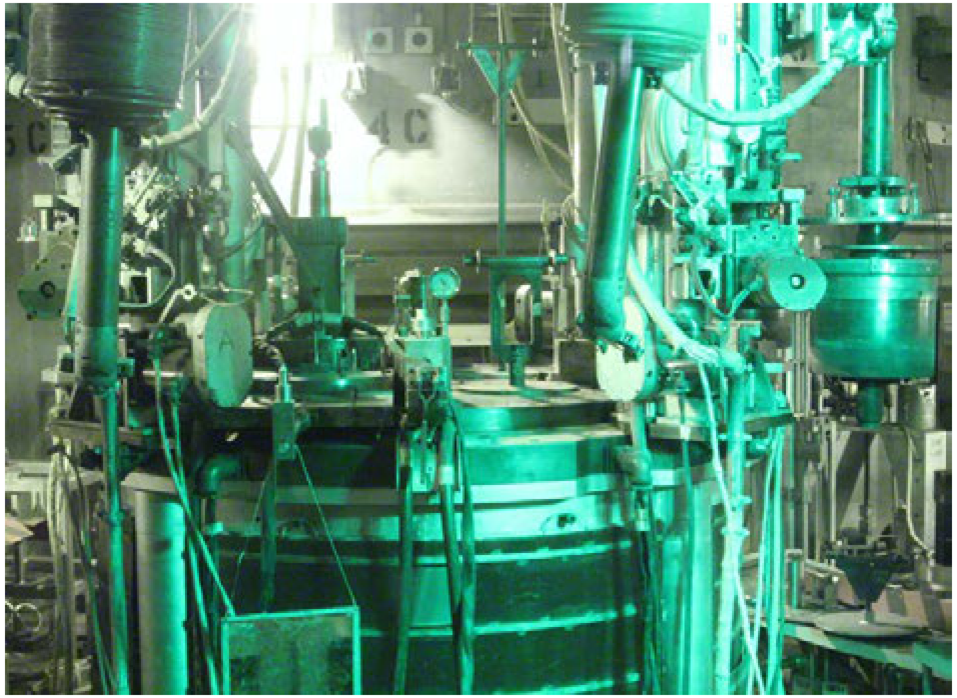




Takeaway: Engineered features for criticality safety necessary due to HEU.
Photographed through 1.5 m of glass.

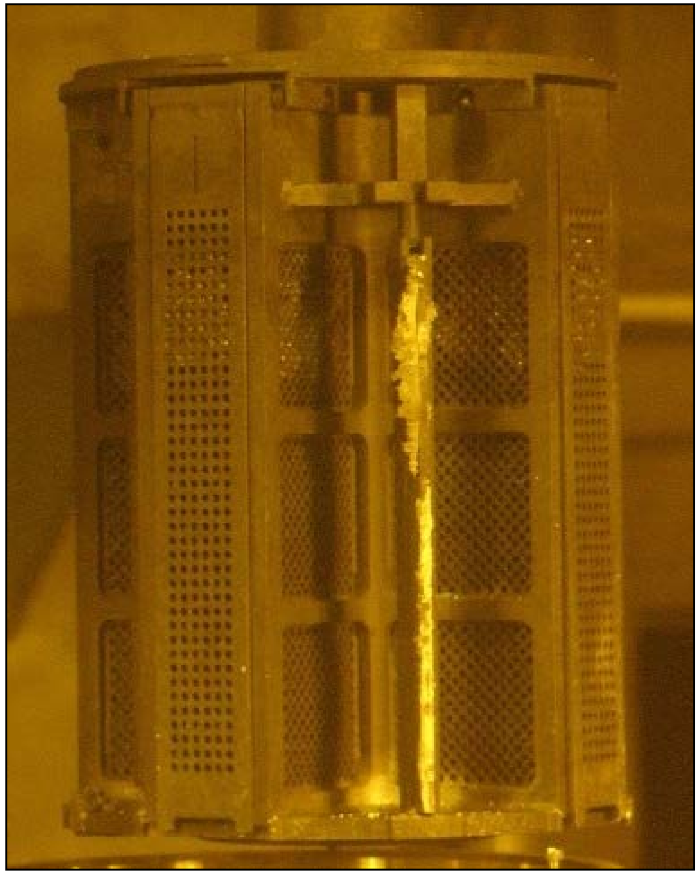
Spent Fuel Treatment Program



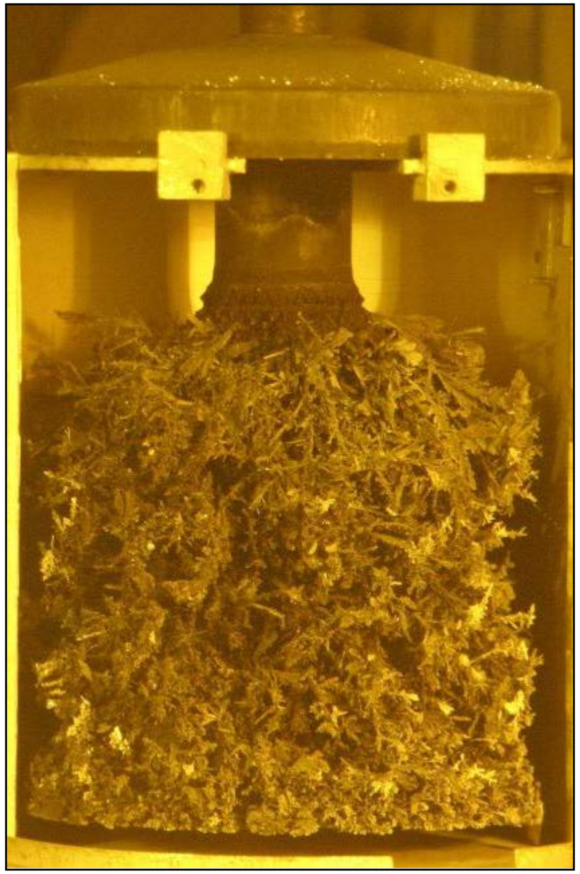


- Mk-IV electrorefiner for driver fuel.
  - Four electrode ports.
  - Two independent power supplies.
  - Two anode/cathode pairs.
  - 1-m-dia. by 1-m-tall.
  - Cr-Mo steel vessel.
  - 650 kg of salt.

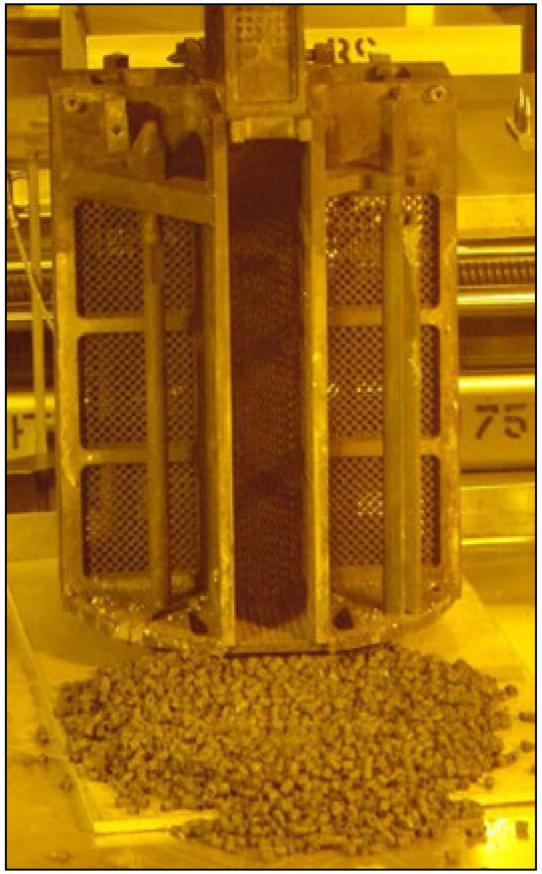
Spent Fuel Treatment Program



Full Anode Basket

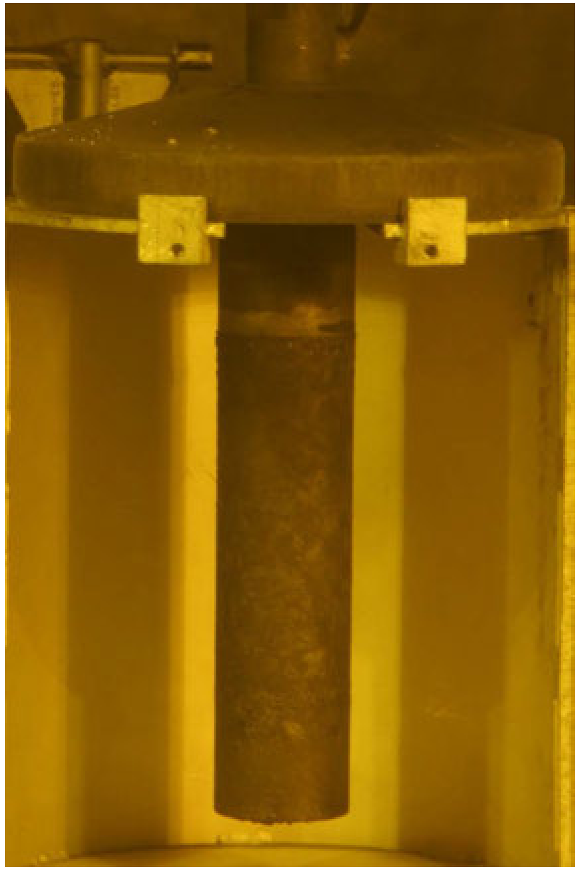


Electrorefined Uranium

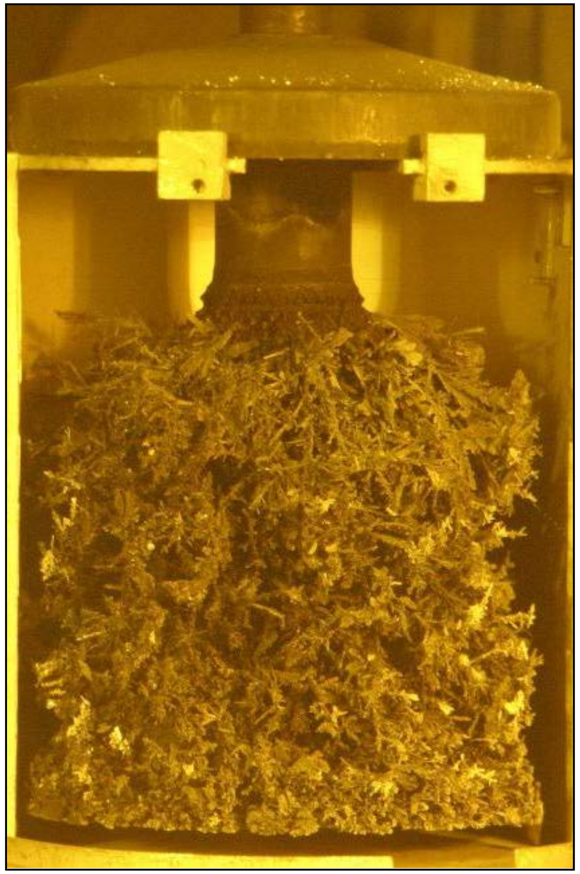


Empty Anode Basket

Spent Fuel Treatment Program



Bare Cathode Mandrel



Uranium Morphology

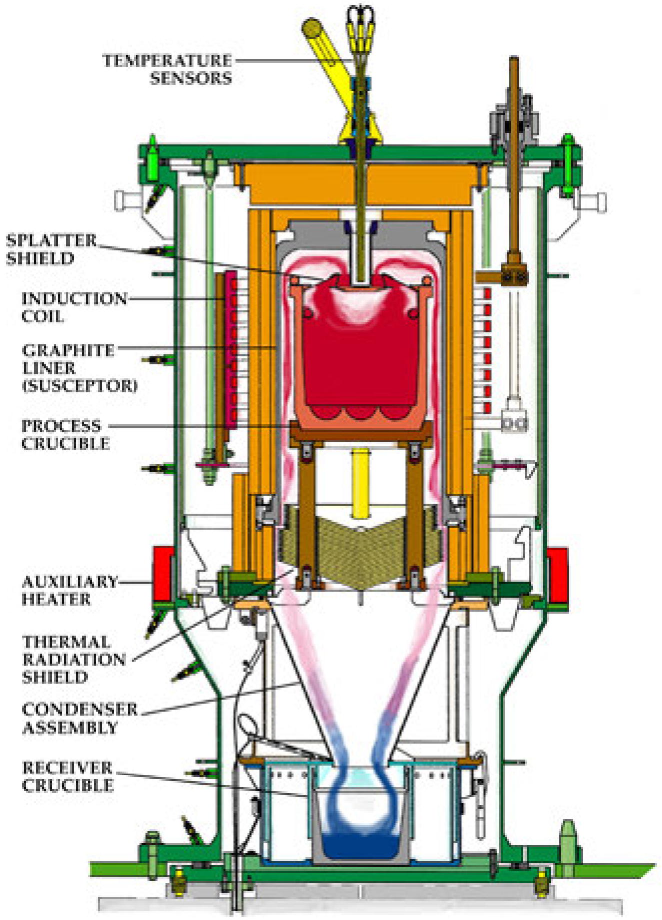
Zirconium Morphology

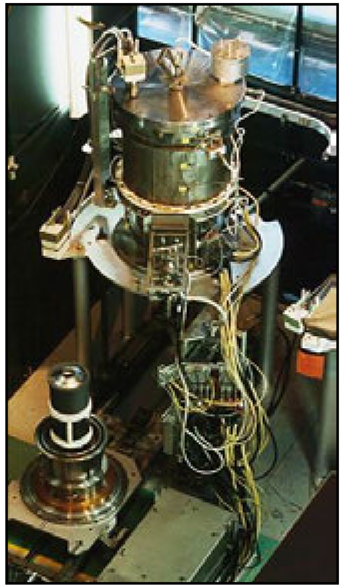
Spent Fuel Treatment Program

- Cathode processor (CP).
  - Vacuum retort furnace.
  - Separate salt and metal.
  - Salt distillate collected in lower receiver.
  - Metal consolidate into ingot.

- Casting furnace.
  - Vacuum retort furnace.
  - Re-melt ingot from CP.
  - Sample to verify enrichment target.

- Final uranium ingot.
  - Must be LEU, < 20% ^{235}U.



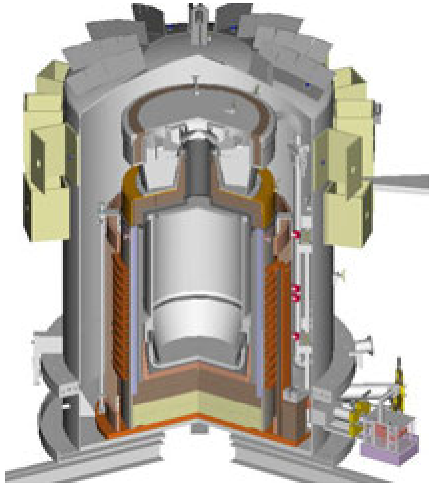


Spent Fuel Treatment Program

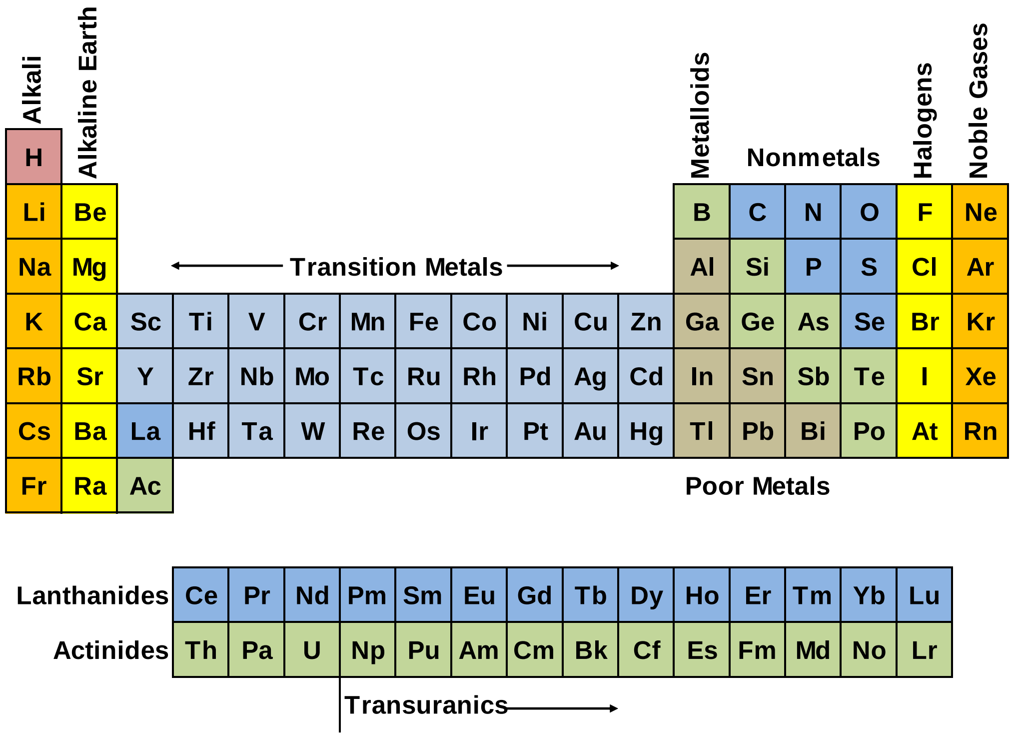

- Ceramic Waste Form.
  - Blend of zeolite, glass, and salt.
  - 200 kg waste form holds 10 kg salt.
  - Research continues for alternate waste forms.

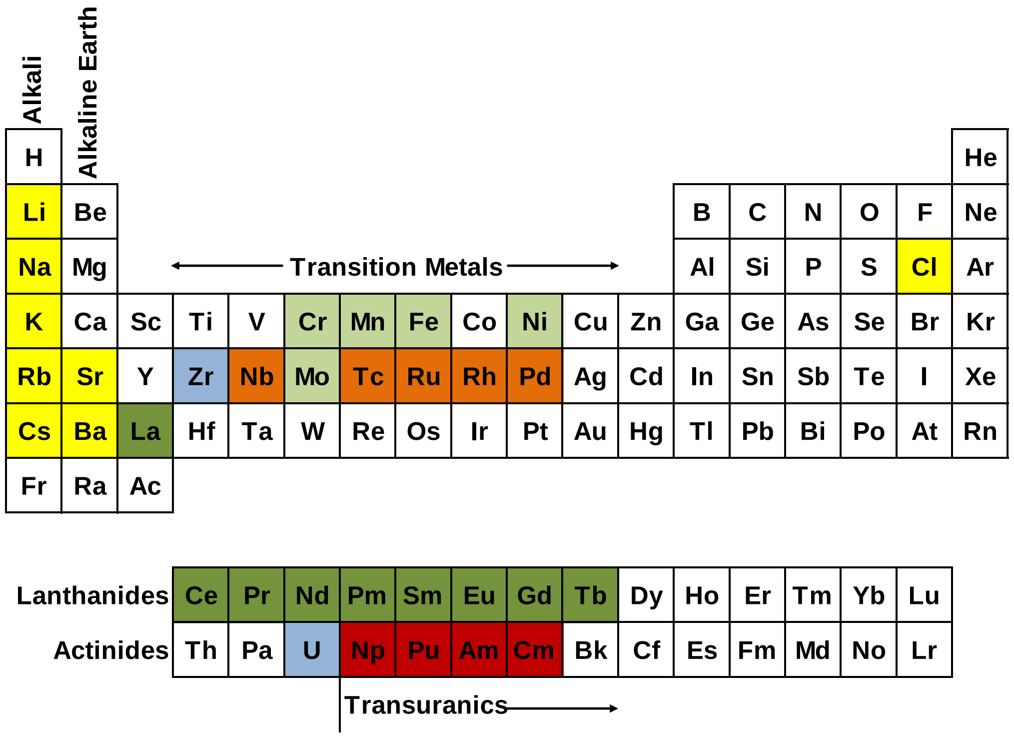
- Metal Waste Form.
  - Alloy of stainless steel and zirconium.
  - Waste forms being produced.
  - Research continues for alternate waste forms.

- Waste form qualified for Yucca Mountain.




Separations Chemistry





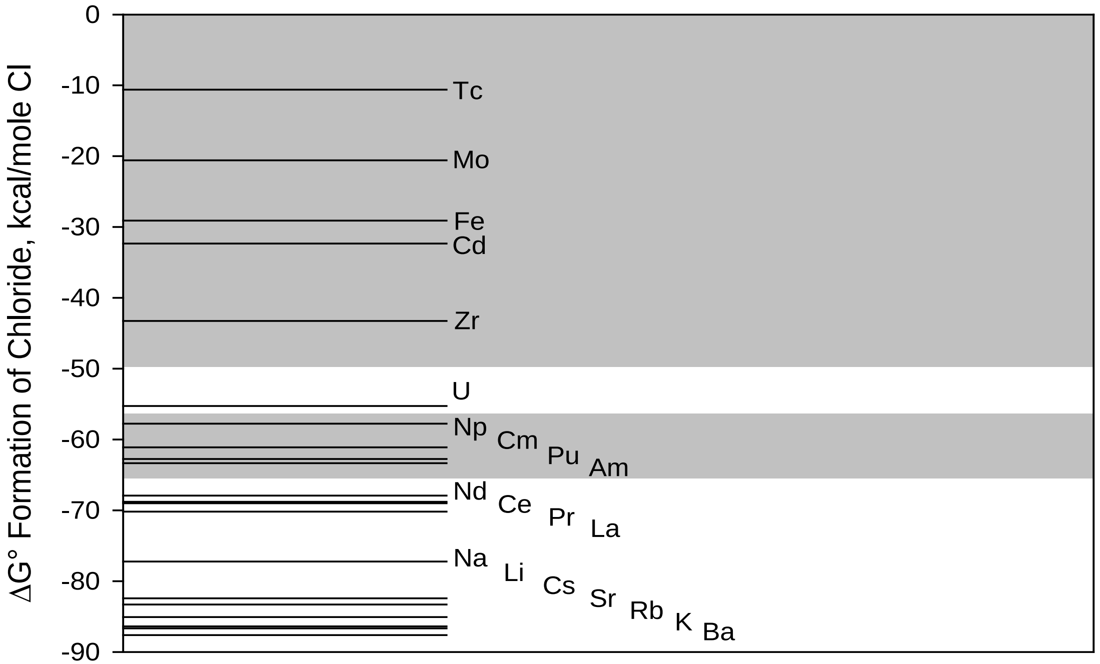
Takeaway: “Group” fission products have similar chemical properties.

Separation Chemistry

- The following FPs accumulate in salt.
  - o Alkali.
  - o Alkaline earth.
  - o Lanthanides.
  - o Transuranics.

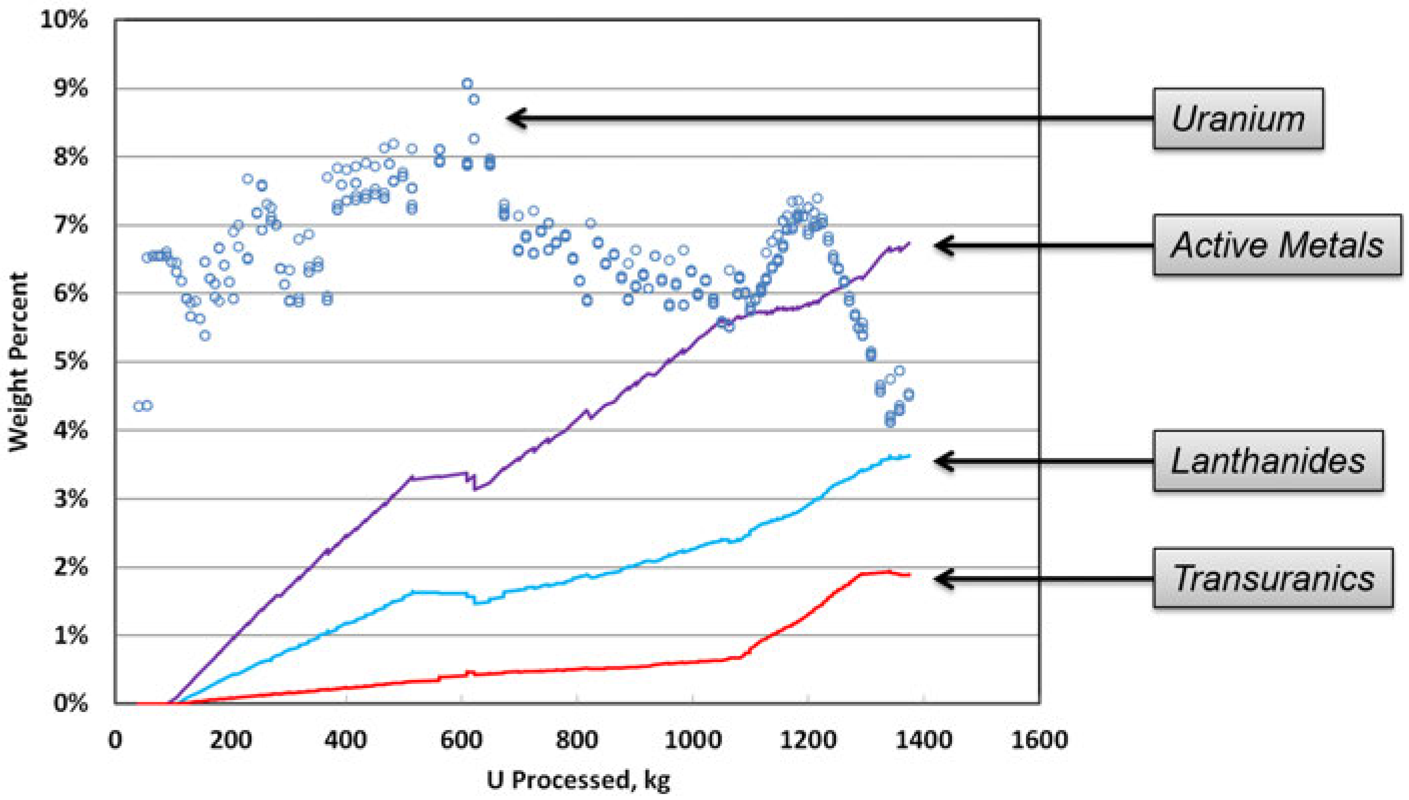
- The following FPs remain in the anode basket.
  - o Transition metals.

- UCl_3 is consumed.
  - o e.g., Ce + UCl_3 = $CeCl_3$ + U.
  - o e.g., Pu + UCl_3 = $PuCl_3$ + U.
  - o e.g., 3 Na + UCl_3 = 3 NaCl + U.



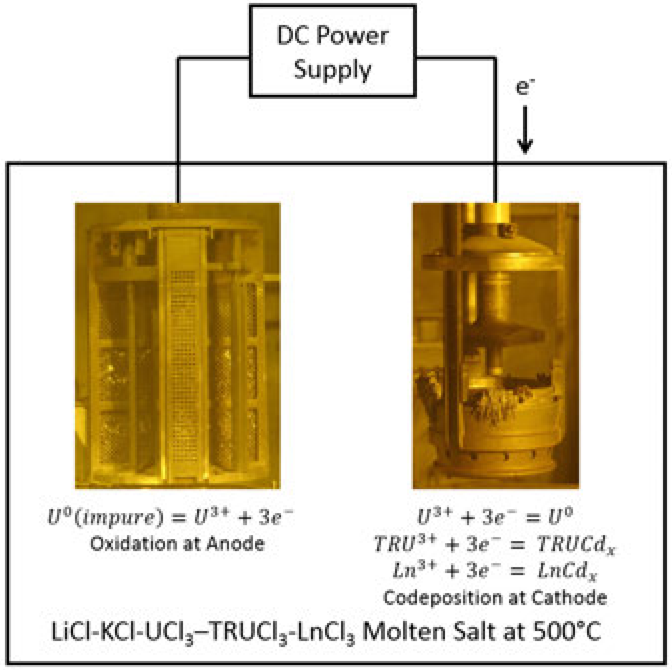
Separations Chemistry

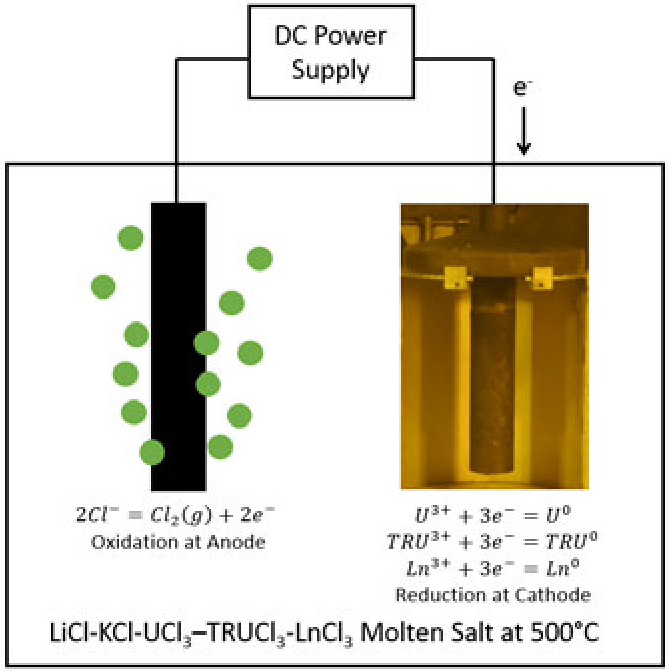
- Composition of the Mk-IV ER salt.
  - o Treatment began in 1996.
- Active metals include.
  - o Alkali.
  - o Alkaline earth.
  - o Sodium-bond.
- UCl_3 maintained by $CdCl_2$ additions.
  - o $3/2\ CdCl_2 + U = UCl_3 + 3/2\ Cd$

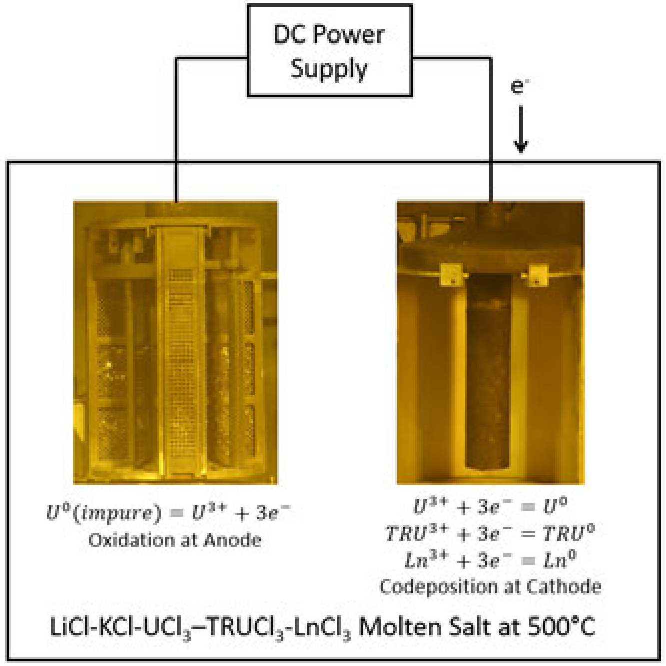


Takeaway: Salt composition changes during every batch of fuel treated.
UCl_3 concentration must be maintained.

TRU Recovery Technologies



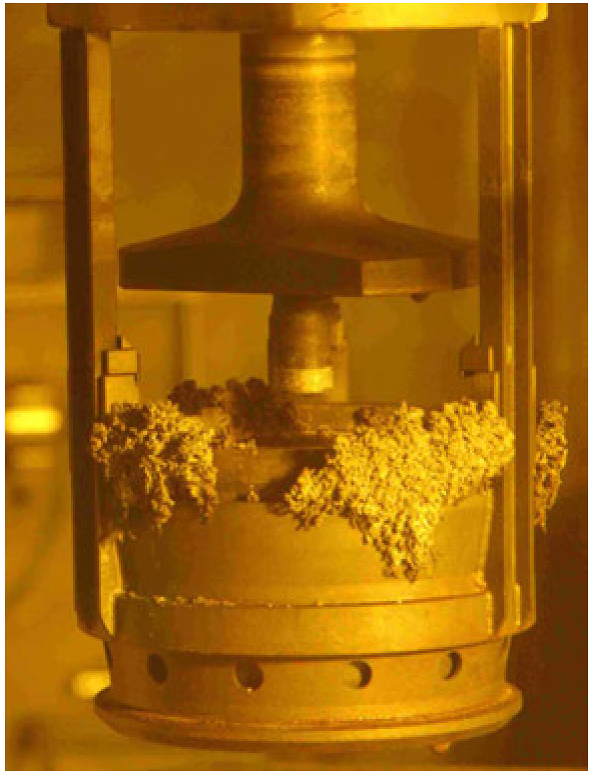




- Alloying metal cathode codeposition (e.g., liquid cadmium, solid aluminum).
- Inert anode codeposition (e.g., chlorine gas evolving graphite anode).
- Reactive anode codeposition (e.g., anode containing spent metallic fuel).

Engineering-Scale Liquid Cadmium Cathode

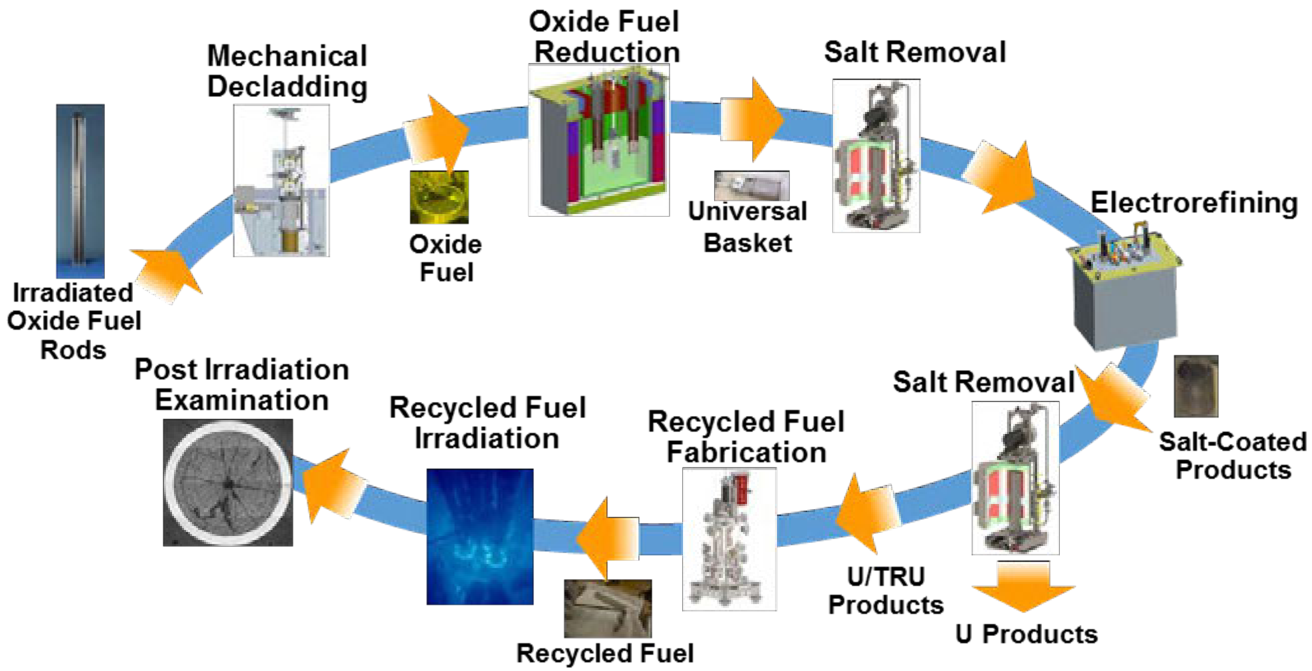
LCC Test	Pu, g	U, g	Pu, %	U, %
1	1,037	367	74	26
2	1,102	652	63	37
3	495	894	36	64
4	552	809	41	59



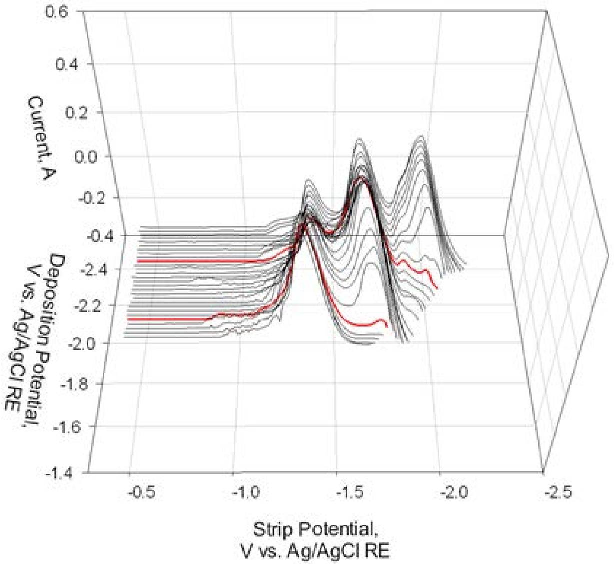
- 24-kg cadmium electrode.
- Beryllia crucible.
- Pu returned to the Mk-V electrorefiner.

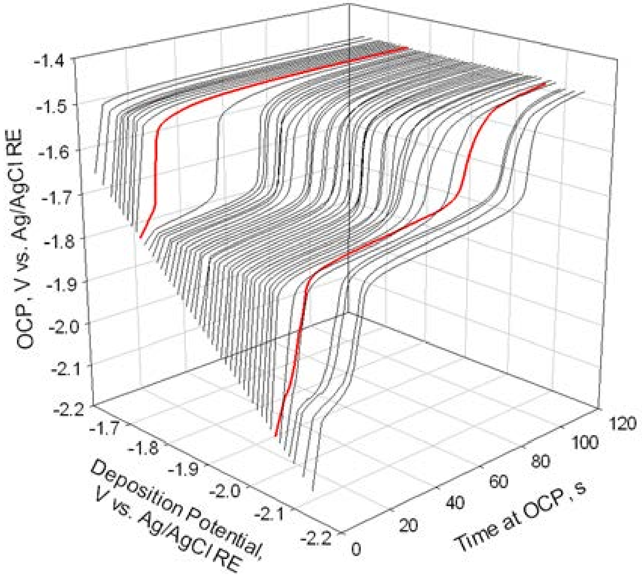
International Collaborations on Pyroprocessing Research

- Goal: Provide data on the technical and economic feasibility and nonproliferation acceptability of electrochemical recycling.
- Multiple research laboratory partners.
- Major unit operations fully integrated.
- Safeguards research incorporated.



Gram-Scale Codeposition

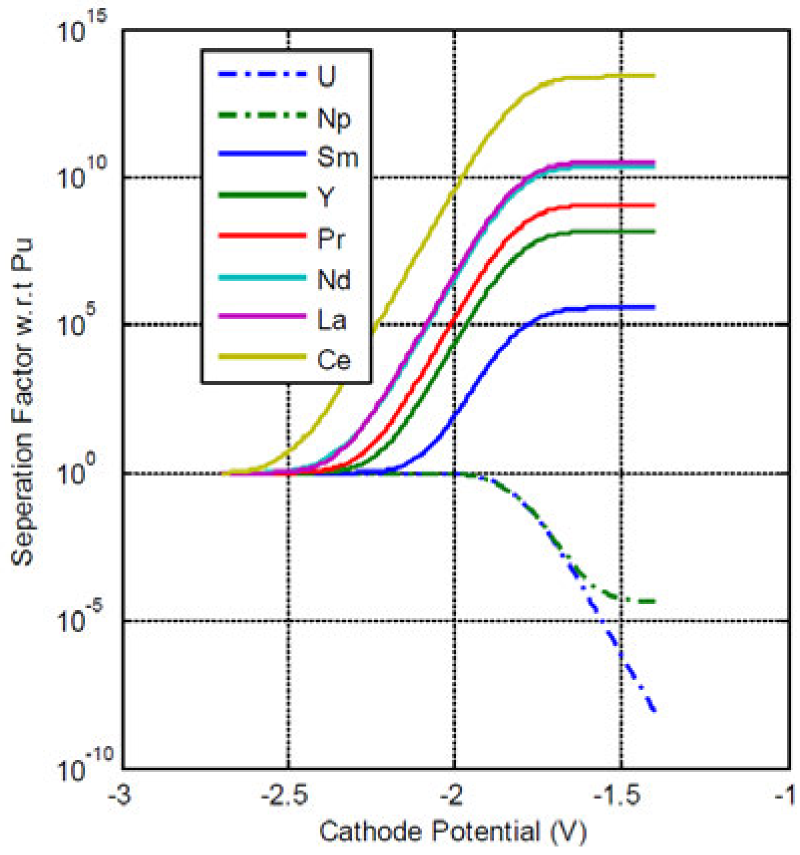




- Electrochemical measurements on 1-mm-dia. tungsten electrode.
- LiCl-KCl eutectic salt (at 500^{o}C) containing UCl_3, $PuCl_3$, and $LnCl_3$.

Gram-Scale Codeposition: Separation Factors

$$SF_i = \frac{x_{i(salt)}}{x_{i(alloy)}} \Bigg/ \frac{x_{Pu(salt)}}{x_{Pu(alloy)}}$$



- SF based on thermodynamic and kinetic modeling.
- SF if a function of cathode potential.
- Separations are never perfect.

Research Publication Trends

Country/Year	2016	2017	2018
Europe	6	17	18
Japan	4	5	2
India	12	4	3
China	42	33	36
ROK	16	12	10
USA	18	19	25
Other	2	10	6

- Percentage of total pyroprocessing related publications (e.g., molten salt electrochemistry).
- Data from: Supathorn Phongikaroon, “Academic Research Paths on Pyroprocessing Technology in the United States with Respect to Other Nations”, 2018 IPCR, Tokai, Japan, October 24-26, 2018

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  - Robert O. Hoover
  - Ken C. Marsden
  - Michael D. Patterson
  - J. C. Price
  - Brenda E. Serrano
  - Michael R. Shaltry
  - Terry A. Todd
  - Prabhat K. Tripathy
  - DeeEarl Vaden
  - Dale R. Wahlquist
  - Dennis L. Wahlquist
  - Stephen A. Warmann
  - Brian R Westphal
  - Tae-Sic Yoo

Summary

- “Pyroprocessing” goes by many names.
    - Electrometallurgical, Electrochemical, Non-Aqueous, Dry-Process, Echem.
- Pyroprocessing flowsheet design is highly application specific.
- Pyroprocessing research is applied to metal fuel, oxide fuel, nitride fuel, aluminum-clad fuel, fluoride MSRs, chloride MSRs.