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# Joint ICTP/IAEA School on Physics and Technology of Fast Reactors Systems

*9 - 20 November 2009*

## Integral Fast Reactor and Associated Fuel Cycle System

## Part 3. Status of Pyroprocess Development

YOON IL CHANG
*Argonne National Laboratory*
*USA*

Argonne
NATIONAL LABORATORY

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# *Integral Fast Reactor and Associated Fuel Cycle System*

## *Part 3. Status of Pyroprocess Development*

*IAEA/ICTP School on Physics and Technology of Fast Reactor Systems*
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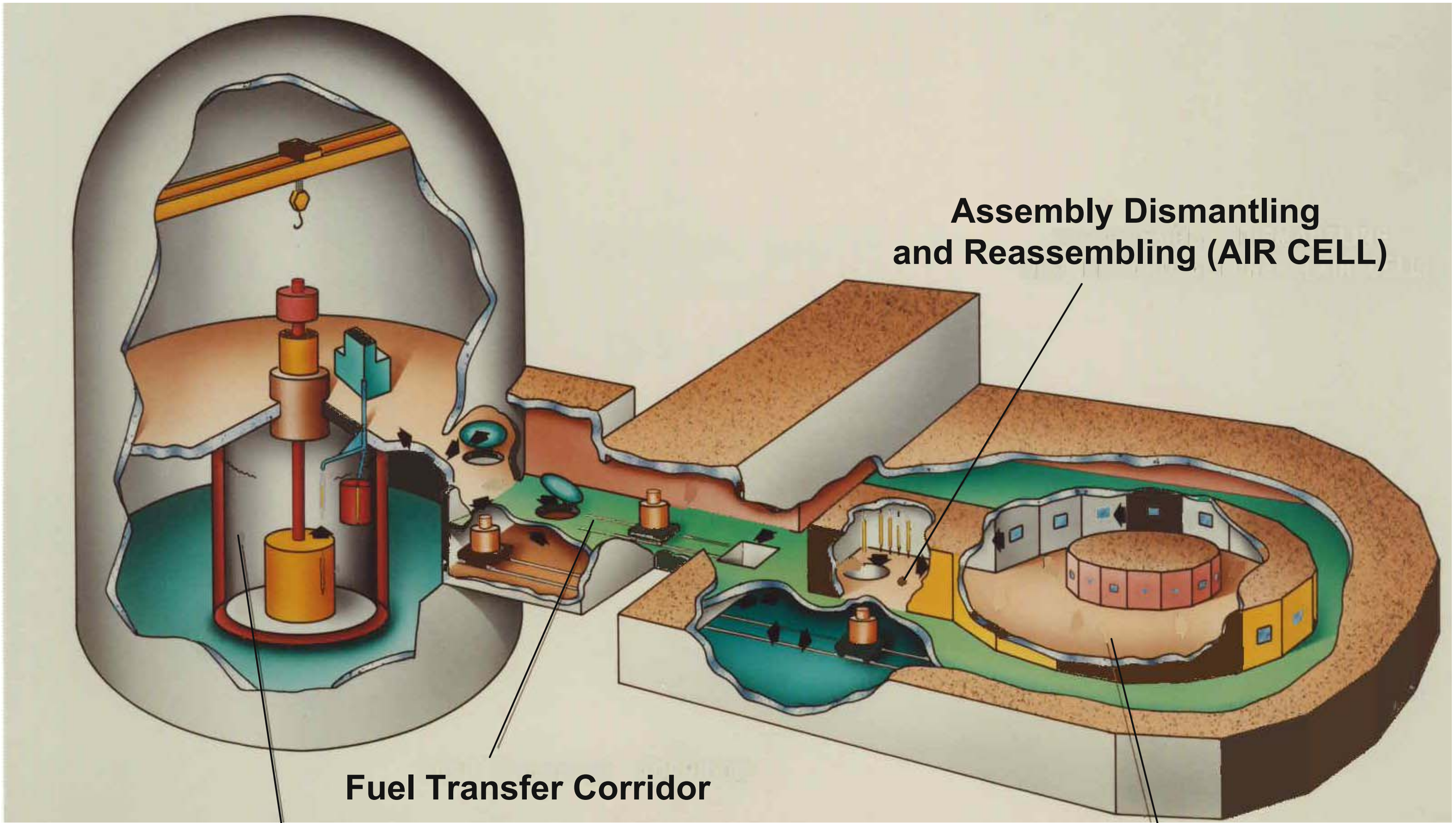
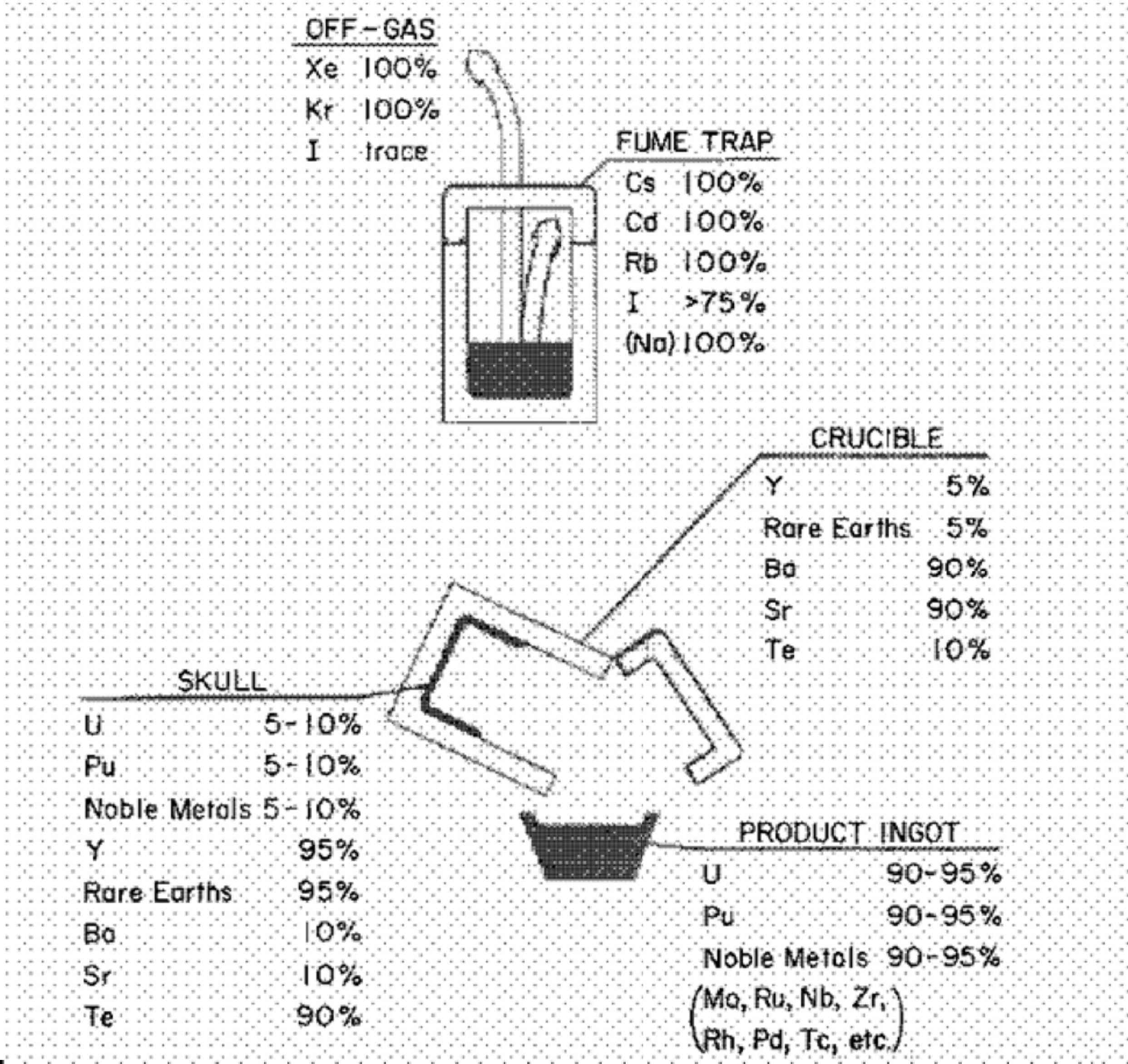
# *Early History of Pyroprocessing*

- Pyroprocessing based on melt-refining was used for the EBR-II fuel cycle closure from 1964-69.
  - Approximately 30,000 irradiated fuel pins were recycled.
  - Average turnaround time was about 2 months from discharge to reload into the reactor.
  - Average throughput rate of 100 kg/month, with peak rate reaching 245 kg/month.

# Pyroprocessing was used to demonstrate the EBR-II fuel cycle closure during 1964-69



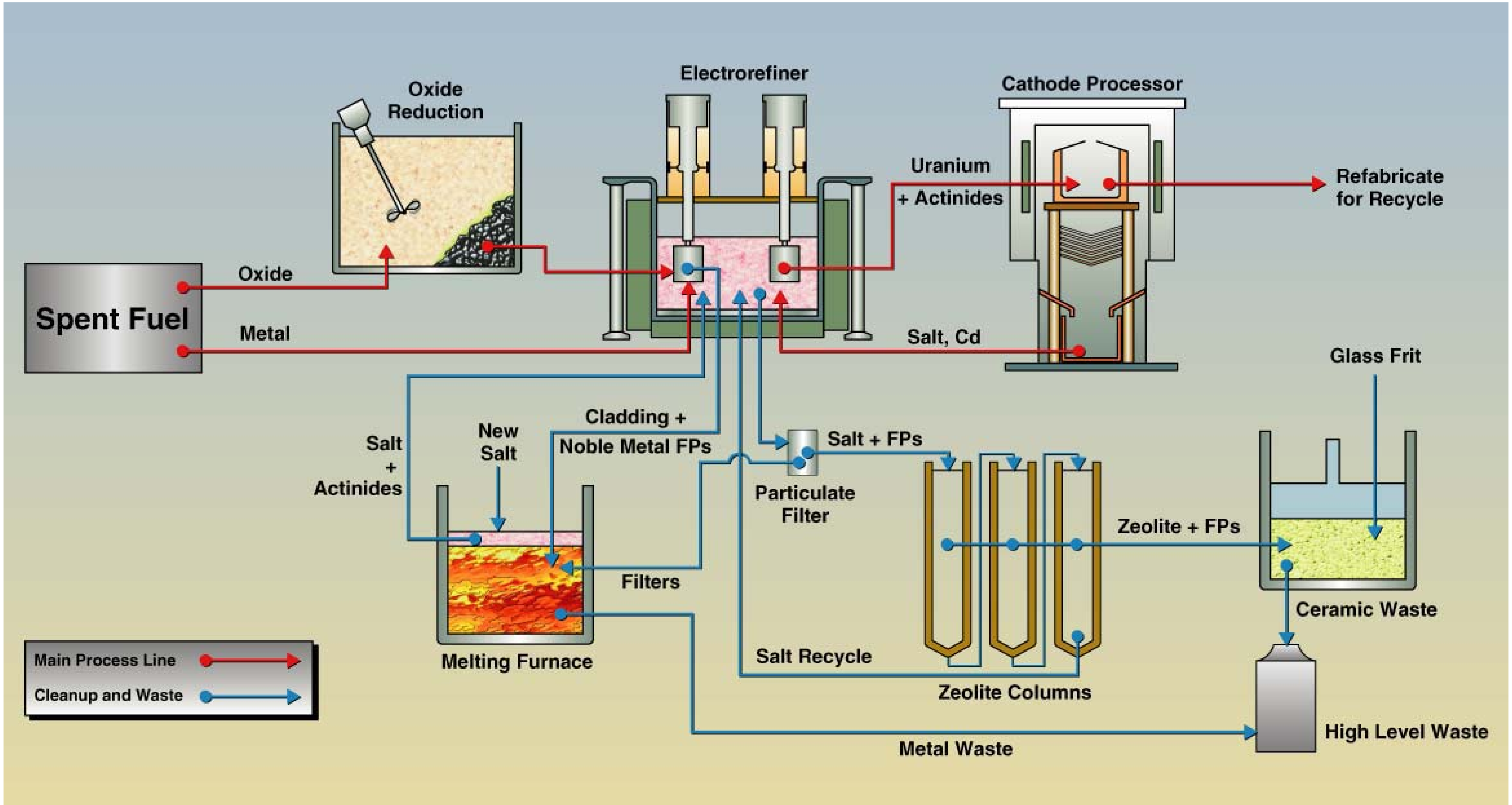
# Melt Refining



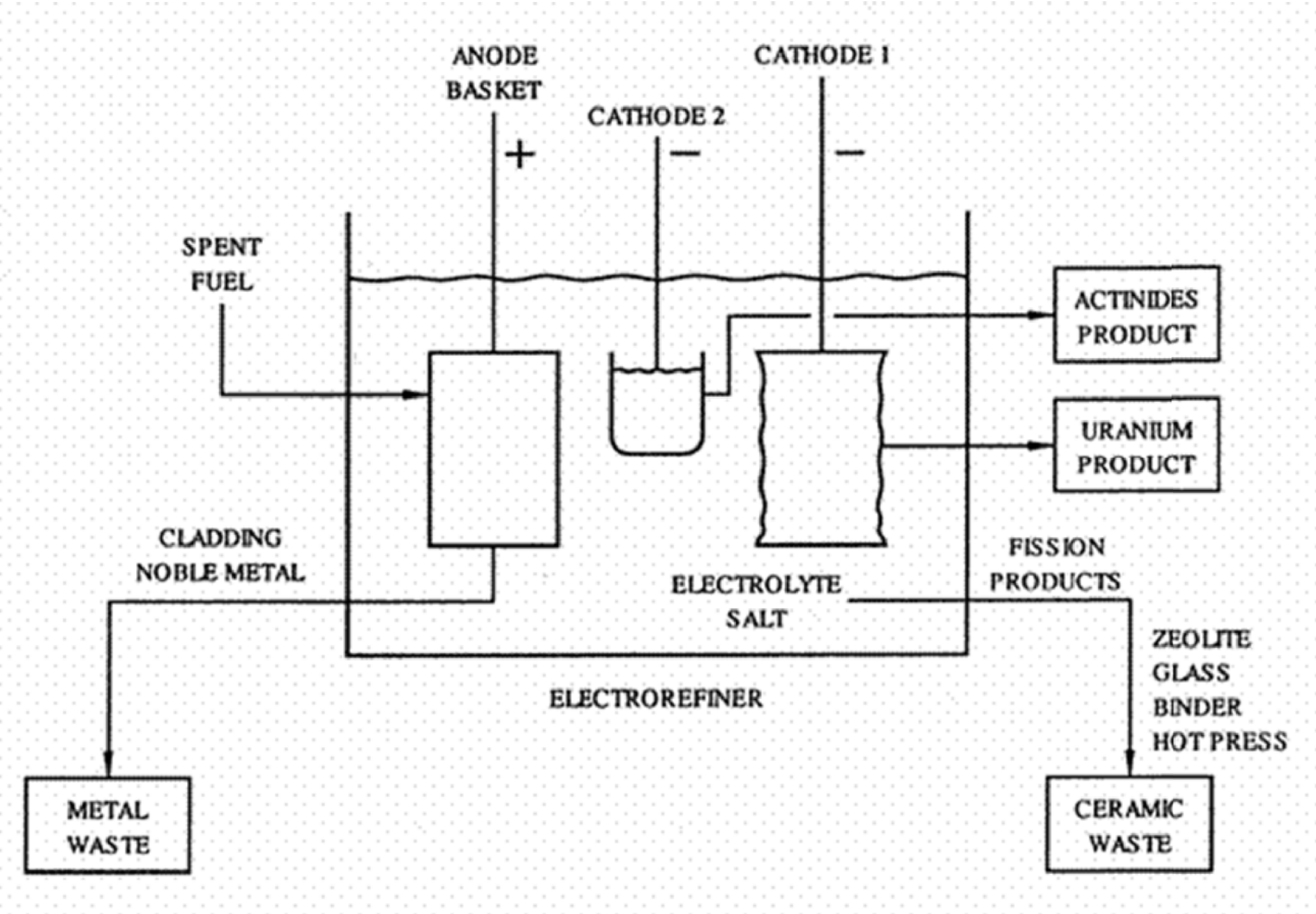
# *Pyroprocessing based on Electrorefining*

- Melt-refining had two deficiencies:
  - Noble metal fission products could not be removed. This was acceptable when the metal fuel was limited to about 1% burnup. With up to 20% burnup demonstrated, noble metal fission products have to be removed for future commercial fast reactors.
  - The early EBR-II fuel was uranium alloy fuel. Melt-refining could not recover plutonium from the blanket, which needs to be enriched for recycle along with the driver fuel.
- In the IFR Program (initiated in 1984) a pyroprocessing based on electrorefining was adopted to solve these deficiencies.

# Pyroprocessing Flowsheet



# *Electrorefining is the Key Step*



# *Two Cathode Types*

- First, uranium is electrochemically transported to a solid cathode in dendritic deposits.
- Second, all actinides are cumulated in the electrolyte salt, and then deposited in a liquid cadmium cathode together.
- The process is incapable of separating a pure plutonium product since the free energies of all minor actinides are very close to each other.

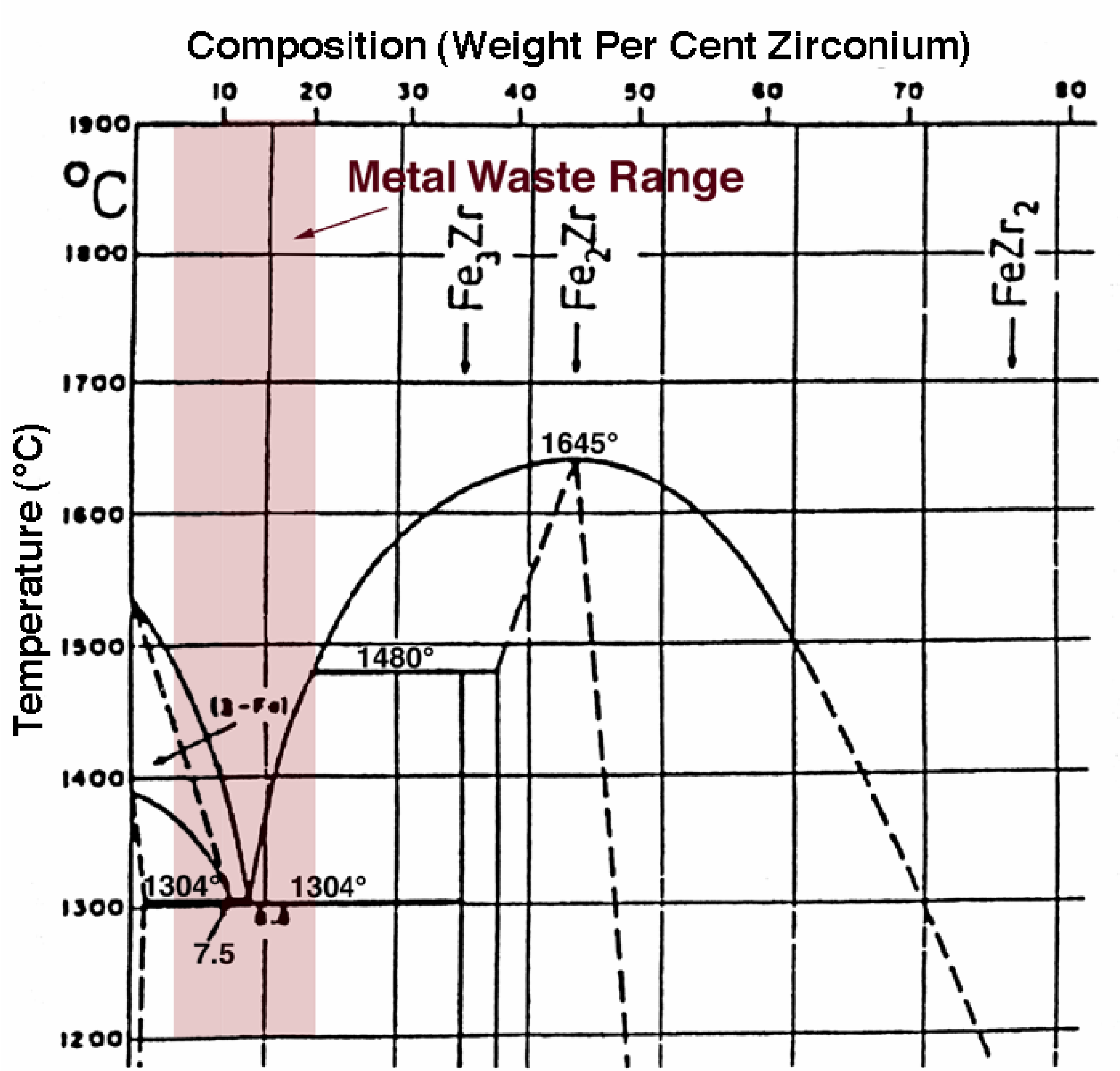
# *Free Energies of Chloride Formation at 500°C, -kcal/g-eqCl*

| Elements that remain in salt (very stable chlorides) | | Elements that can be electrotransported efficiently | | Elements that remain as metals (less stable chlorides) | |
|---|---|---|---|---|---|
| $BaCl_2$ | 87.9 | $CmCl_3$ | 64.0 | $ZrCl_2$ | 46.6 |
| $CsCl$ | 87.8 | $PuCl_3$ | 62.4 | $CdCl_2$ | 32.3 |
| $RbCl$ | 87.0 | $AmCl_3$ | 62.1 | $FeCl_2$ | 29.2 |
| $KCl$ | 86.7 | $NpCl_3$ | 58.1 | $NbCl_5$ | 26.7 |
| $SrCl_2$ | 84.7 | $UCl_3$ | 55.2 | $MoCl_4$ | 16.8 |
| $LiCl$ | 82.5 | | | $TcCl_4$ | 11.0 |
| $CaCl_2$ | 80.7 | | | $RbCl_3$ | 10.0 |
| $LaCl_3$ | 70.2 | | | $PdCl_2$ | 9.0 |
| $PrCl_3$ | 69.0 | | | $RuCl_4$ | 6.0 |
| $CeCl_3$ | 68.6 | | | | |
| $NdCl_3$ | 67.9 | | | | |
| $YCl_3$ | 65.1 | | | | |

# Metal Waste Form

- Following electrorefining, the anode basket that contains stainless cladding hulls, fuel matrix alloy zirconium, noble metal fission products (including technetium), and adhering salt is heated in the metal waste form furnace to distill the adhering salt, and then heated to higher temperature to consolidate the metal waste form.
- For fast reactors, the base alloy for metal waste will be stainless steel with zirconium concentration in the range of 5-20% to form a low melting eutectic.
- For LWRs, the base alloy will be zirconium with about 15% iron, which forms even lower temperature eutectic on the other side of the Fe-Zr phase diagram.

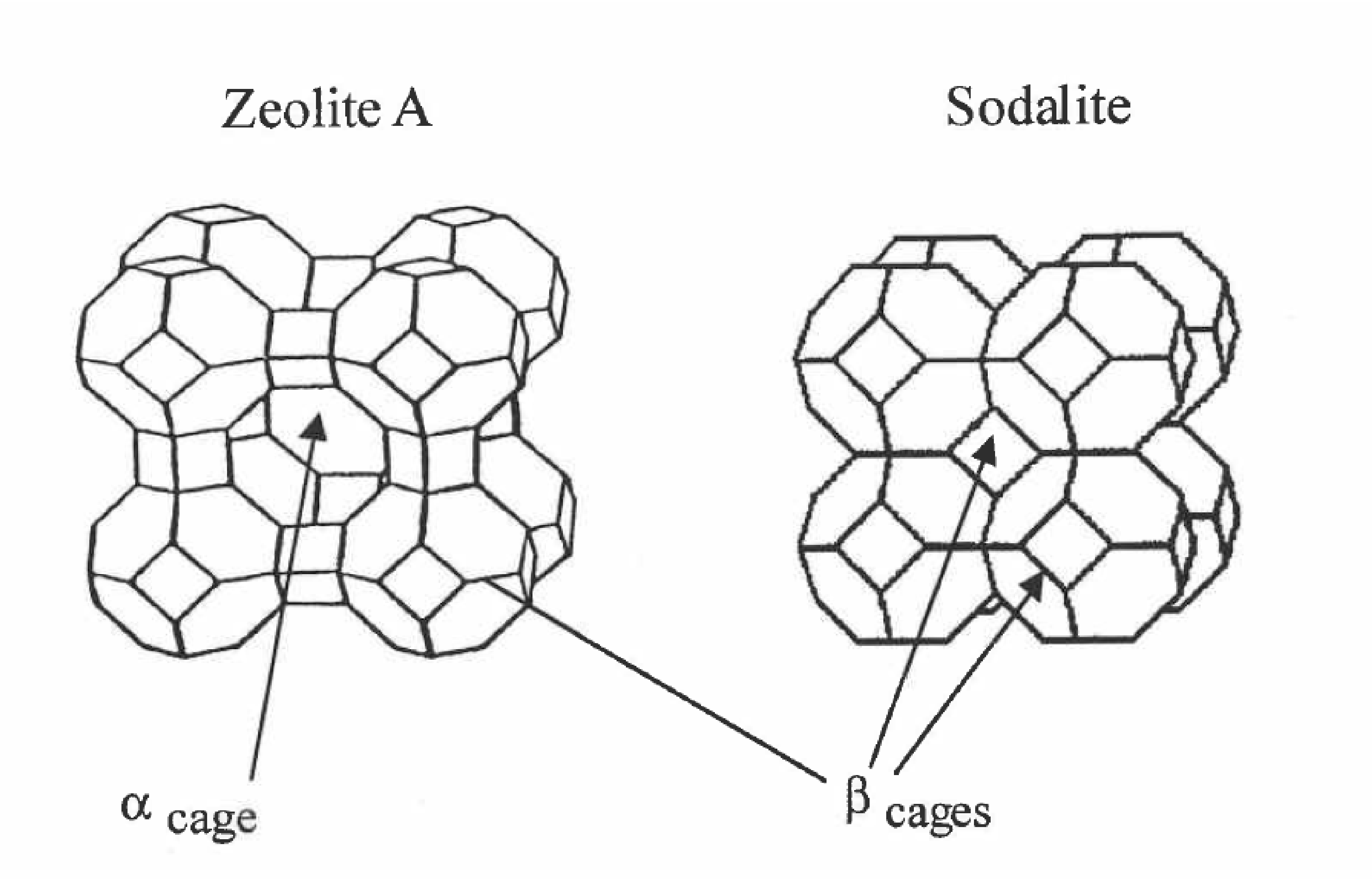
# Fe-Zr Phase Diagram



# *Ceramic Waste Form*

- Most of fission products other than noble metals cumulate in the salt phase. When saturated, salt is passed through zeolite column.
- Fission product cations are adsorbed onto zeolite by ion exchange or occuluded into molecular cages of zeolite structure.
- Zeolite with fission products immobilized is consolidated into a monolithic form by sintering at high temperatures combined with borosilicate glass as binder.
- At high temperatures, zeolite is converted into sodalite, a stable naturally occuring mineral.

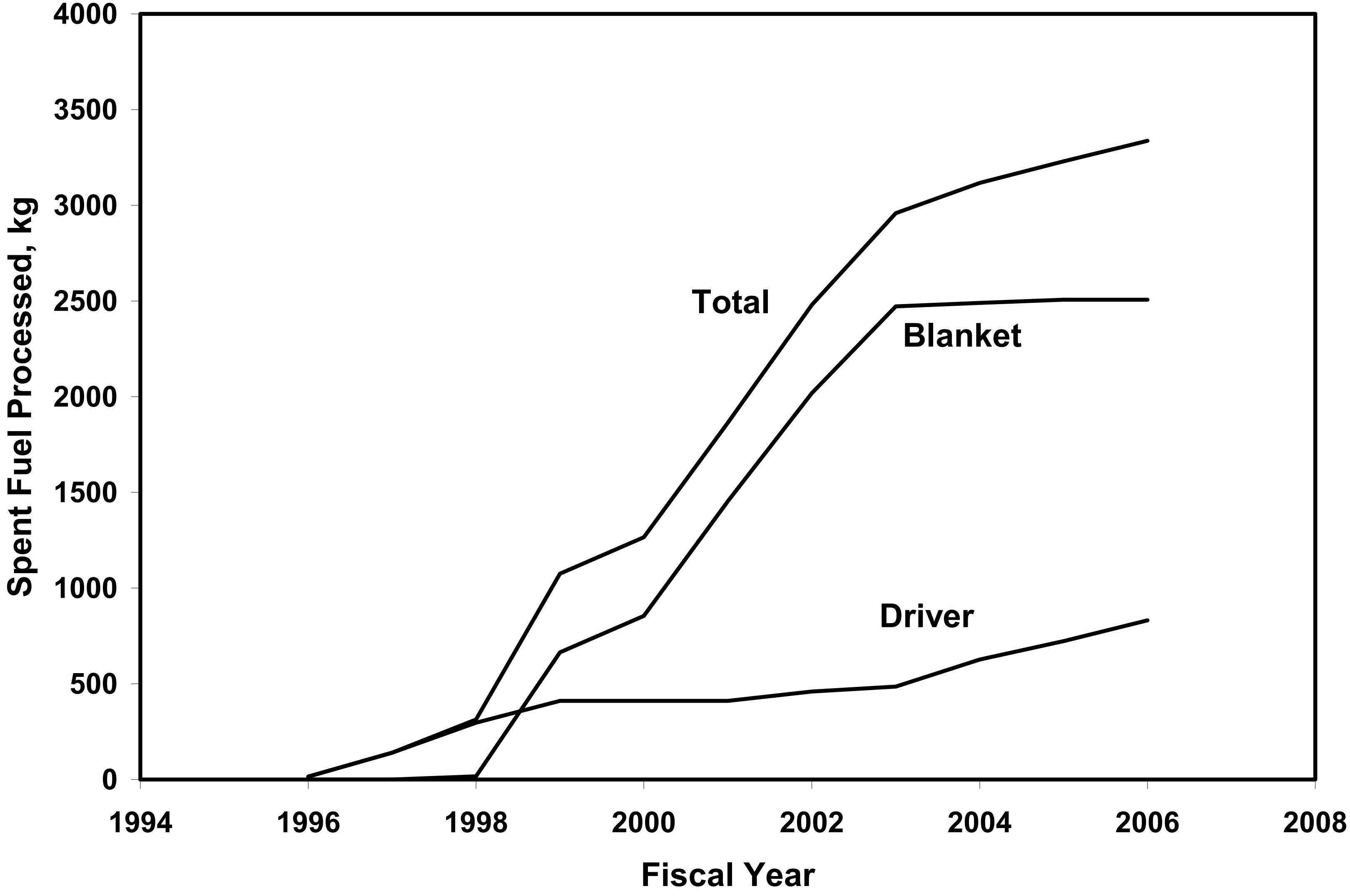
# Molecular Cages of Zeolite and Sodalite



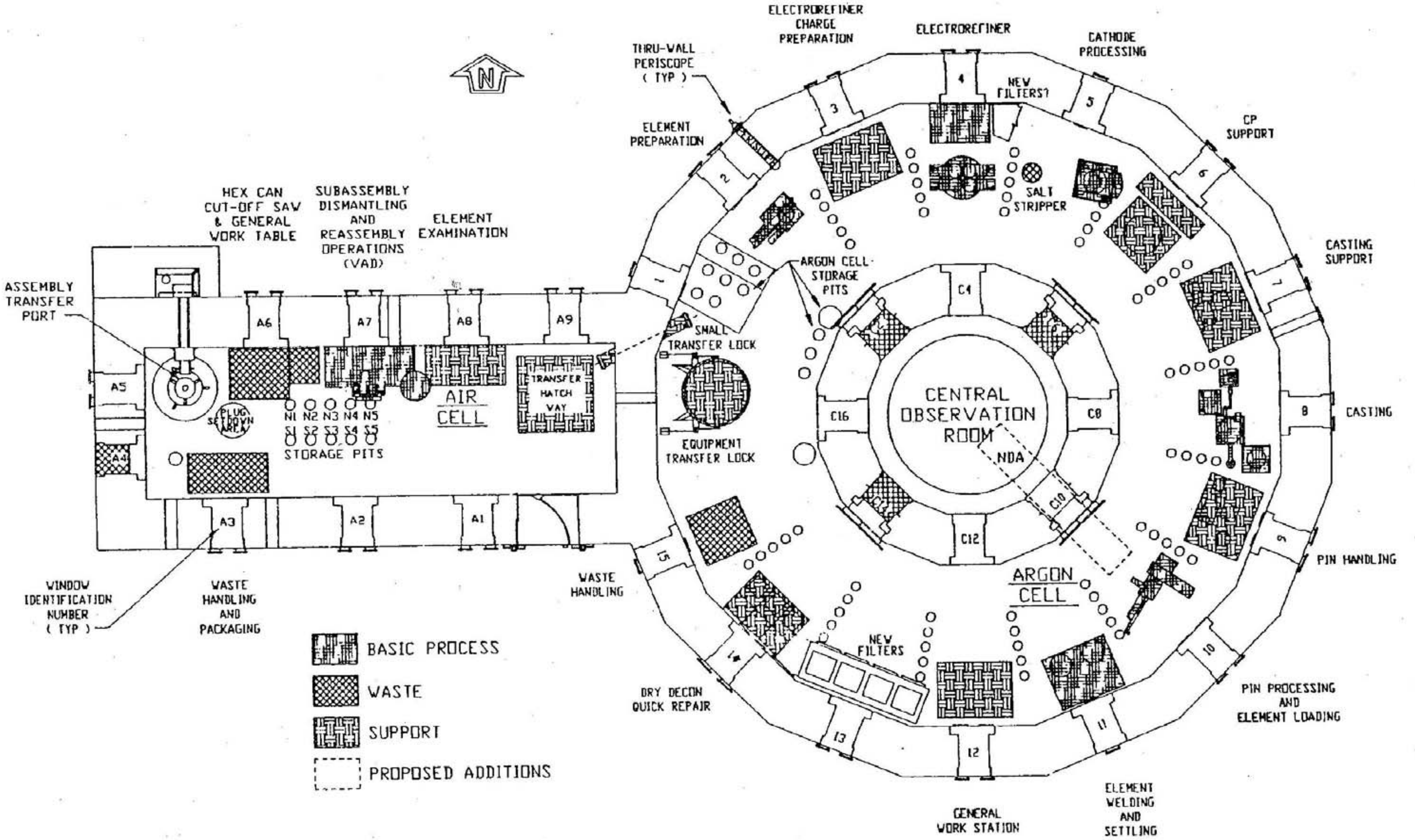
# *Progress since IFR Termination*

- EBR-II was shut down on September 30, 1994 after 30 years of very successful operation.
- EBR-II spent fuel has unique characteristics that require treatment before ultimate disposal:
  - Sodium-bonded: RCRA characteristic
  - Metal fuel: pyrophoric
  - Highly enriched uranium: criticality concerns
- Demonstration project involved treatment of 100 driver and 25 blanket assemblies over three year period at ANL-West
- Following a successful demonstration, the EBR-II spent fuel treatment started in 1998 and still ongoing.

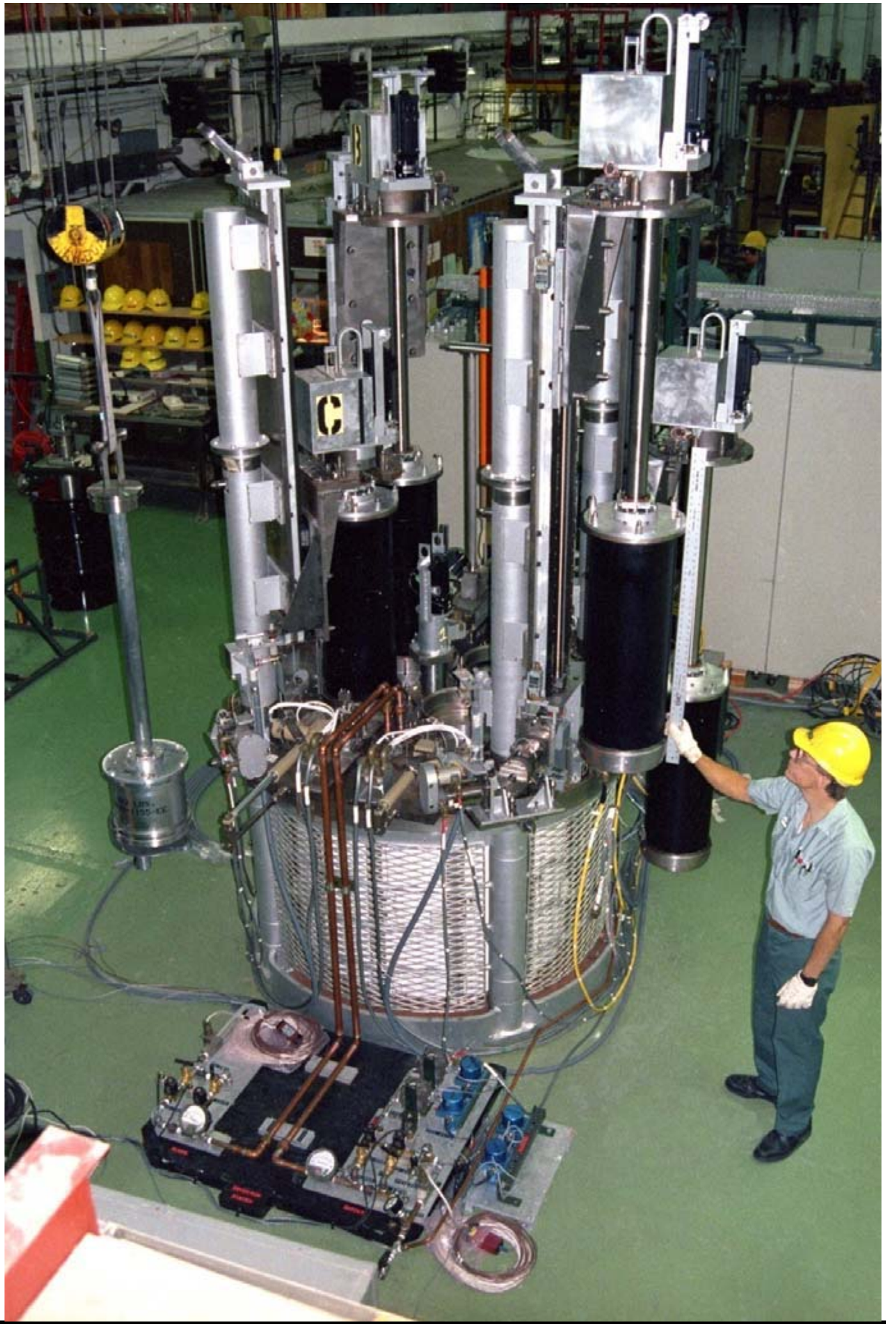
# *Cumulative Quantities of EBR-II Spent Fuel Treatment*



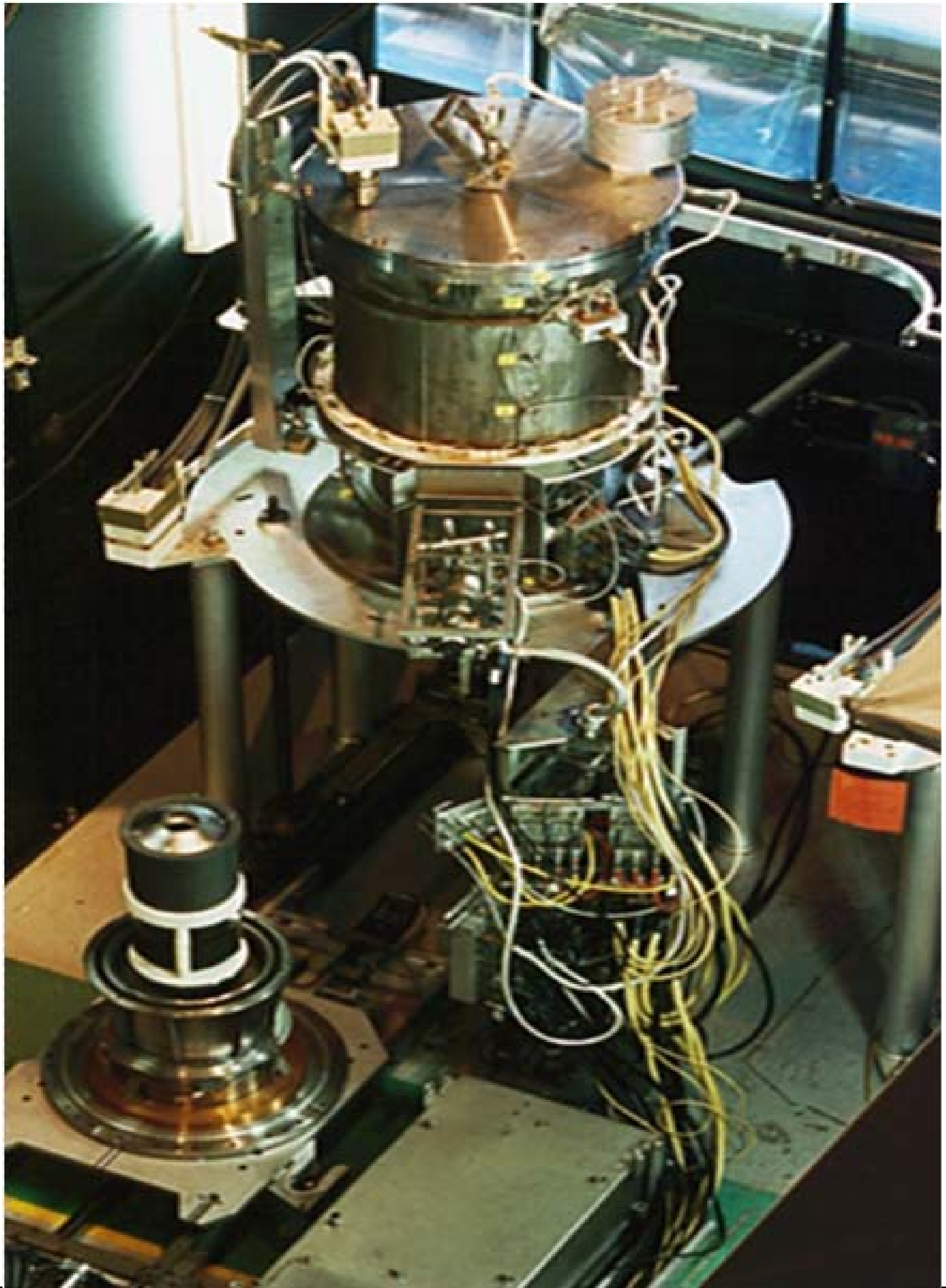
# EBR-II Fuel Cycle Facility



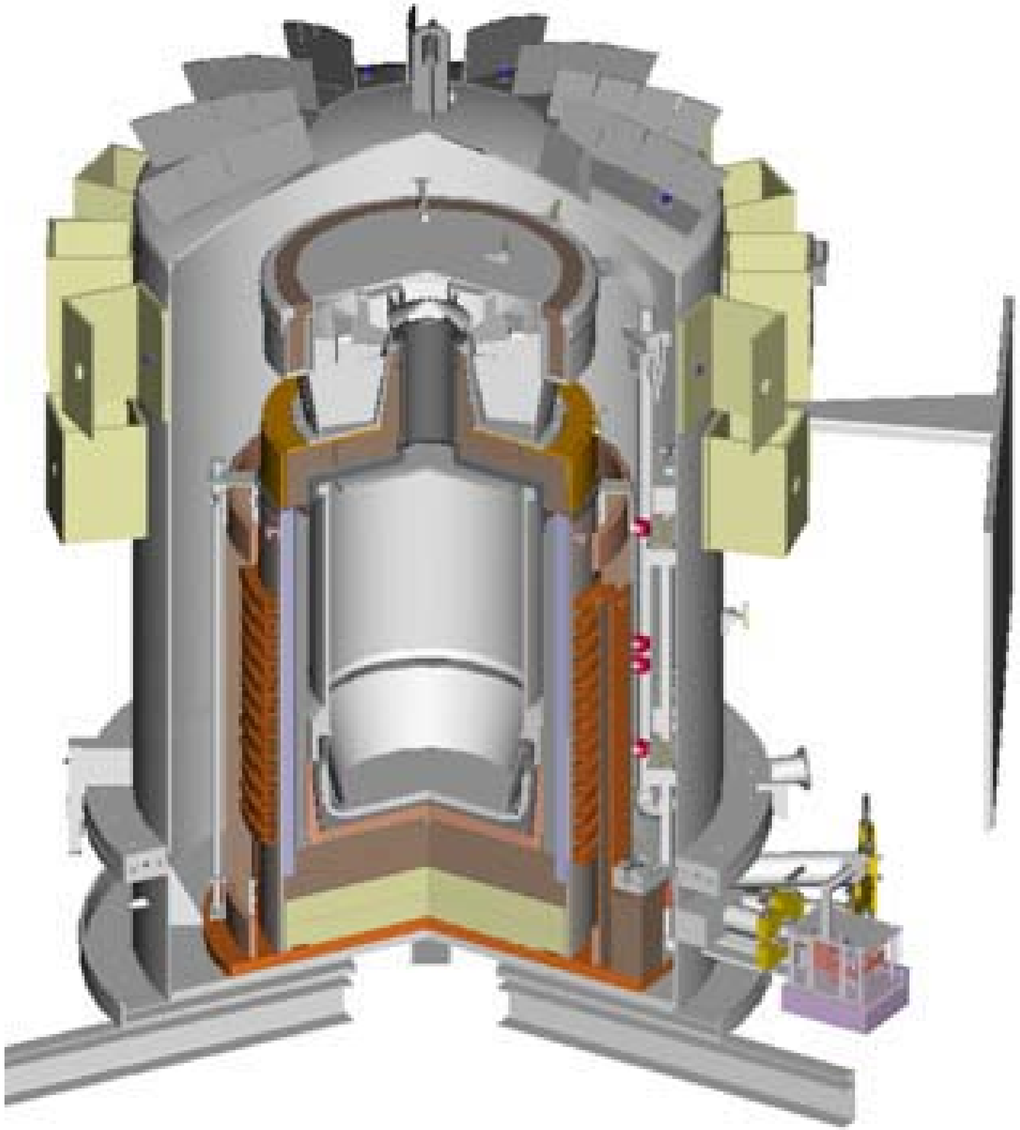
# *Electrorefiner*



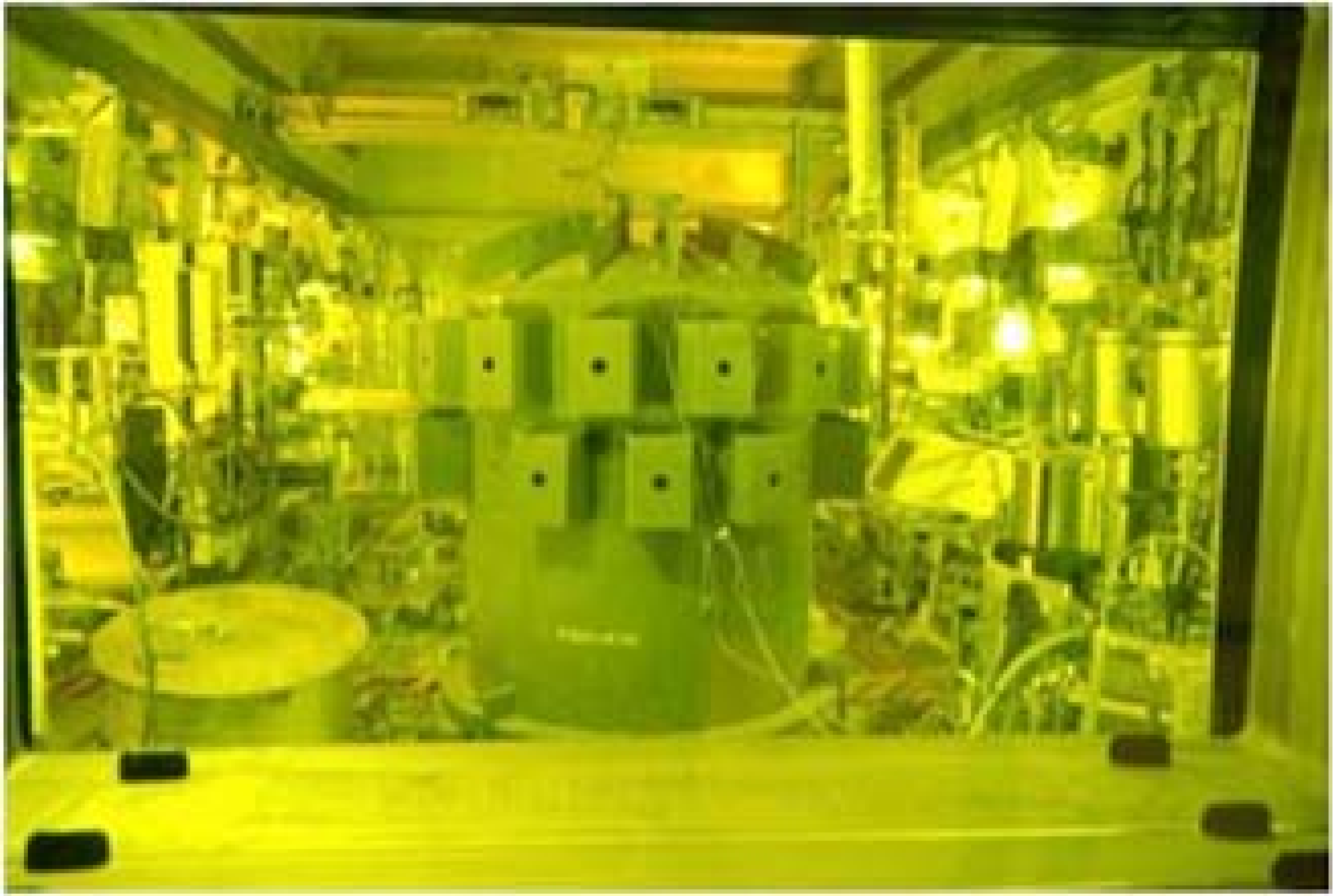
# *Cathode Processor*



# *Metal Waste Furnace*



# *Metal Waste Furnace Installed in Hot Cell*



# *Key Attributes of Pyroprocessing*

- Compact equipment systems based on electrorefining.
- All actinides are recovered together, and hence there is no need to develop additional partitioning processes.
- Direct waste processing and no liquid low level waste streams.
- Intrinsic proliferation-resistance characteristics.
- All of the above characteristics combine to a potentially drastic improvement in economics.

# *Capital Cost Comparison ($million)*
# *Fuel Cycle Facility for 1400 MWe Fast Reactor*

| | Pyroprocessing | Aqueous Reprocessing |
|---|---|---|
| Size and Commodities | | |
| Building Volume, ft3 | 852,500 | 5,314,000 |
| Volume of Process Cells, ft3 | 41,260 | 424,300 |
| High Density Concrete, cy | 133 | 3,000 |
| Normal Density Concrete, cy | 7,970 | 35-40,000 |
| | | |
| Capital Cost, $million | | |
| Facility and Construction | 65.2 | 186.0 |
| Equipment Systems | 31.0 | 311.0 |
| Contingencies | 24.0 | 124.2 |
| Total | 120.2 | 621.2 |

# Weapons Usability Comparison

| | Weapon Grade Pu | Reactor Grade Pu | IFR Grade Actinide |
|---|---|---|---|
| Production | Low burnup PUREX | High burnup PUREX | Fast reactor Pyroprocess |
| Composition | Pure Pu 94% Pu-239 | Pure Pu 65% Pu-fissile | Pu + MA + U 50% Pu-fissile |
| Thermal power w/kg | 2 - 3 | 5 - 10 | 80 - 100 |
| Spontaneous neutrons, n/s/g | 60 | 200 | 300,000 |
| Gamma radiation r/hr at ½ m | 0.2 | 0.2 | 200 |

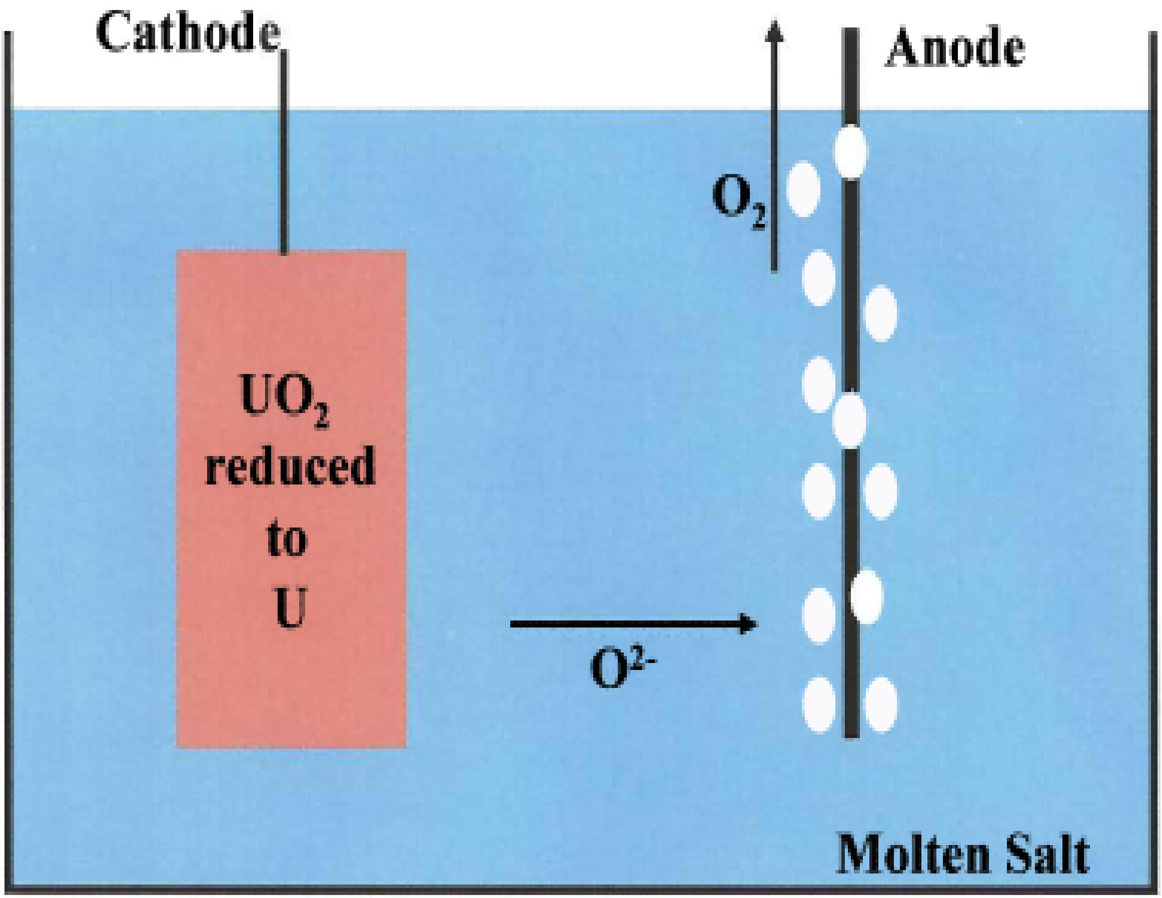
# *Joint Program on Pyroprocessing with Japan*

- Central Research Institute of Electric Power industry (CRIEPI): $20 million cost sharing signed in July 1989.
- CRIEPI and Japan Atomic Power Company jointly representing Federation of Electric Power Companies (FEPC): Additional $20 million added in October 1992.
- Tokyo, Kansai, and Chubu Electric Power Companies: $6 million for LWR feasibility study signed in July 1992.
- Power Reactor and Nuclear Fuel Development Corporation (PNC): $60 million cost sharing program agreed to in February 1994, but canceled by DOE.
- These joint programs ended when the IFR Program was terminated in October 1994.

# A key question is whether pyroprocessing can be applied to LWR spent fuel as well

- For LWR spent fuel application, oxide-to-metal reduction front-end step is required:
  - Electrolytic reduction process is proven most promising.
- For economic viability, the electrorefining batch size and throughput rate have to be increased: this should be straightforward with planar electrode concept.
- Hence, the application of pyroprocessing for the LWR spent fuel is straightforward, and may provide drastic improvements in economics, waste management, repositories, etc.

# Schematics of Electrolytic Reduction Process



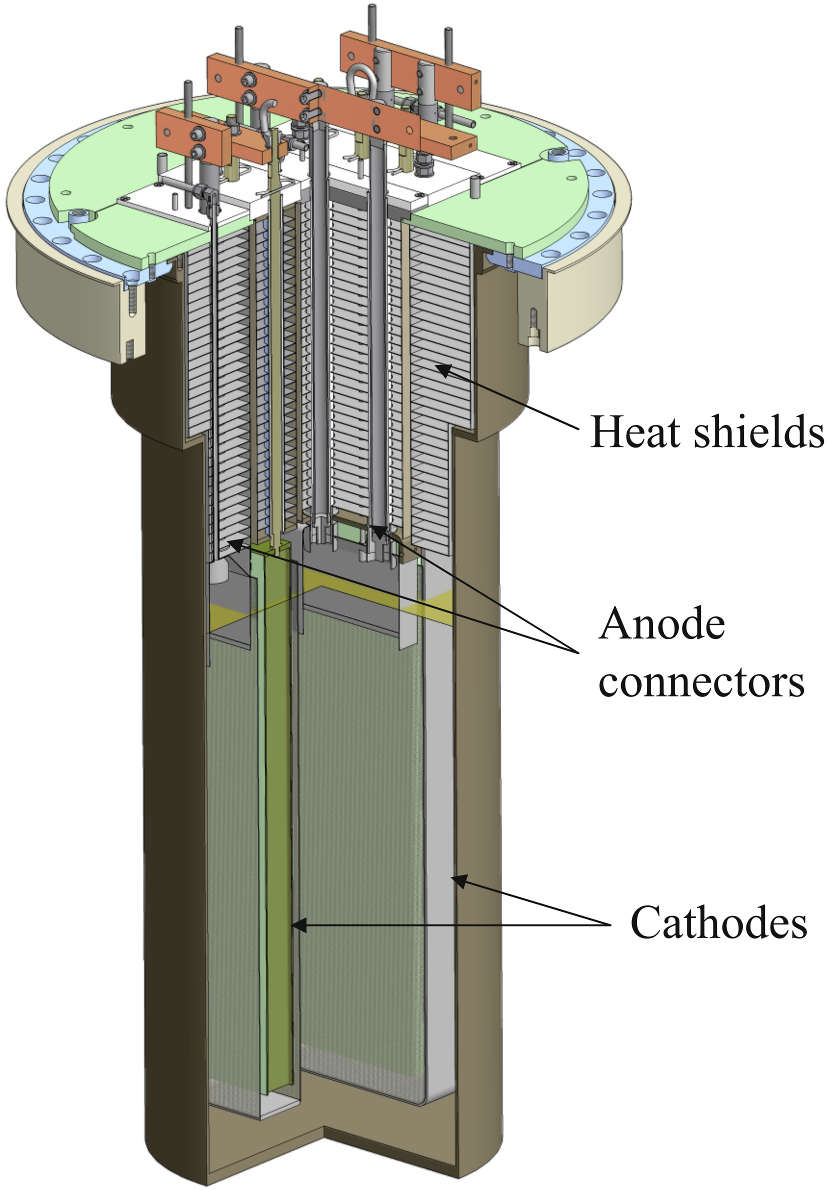
# *Chemical principles of electrolytic reduction process have been satisfactorily demonstrated*

- Complete reduction of $UO_2$ (>99.7%) routinely demonstrated up to the kilogram scale.
- Complete reduction of $U/PuO_2$ demonstrated at gram scale.
- Alkali and alkaline earth fission products have no effect on conversion process and no show stoppers identified in other fission products, e.g., lanthanides and iodine.
- High reduction rate and efficient oxygen removal demonstrated.

*Sample reduced uranium product*

# Engineering-scale electrolytic reduction system is ready for fabrication and testing

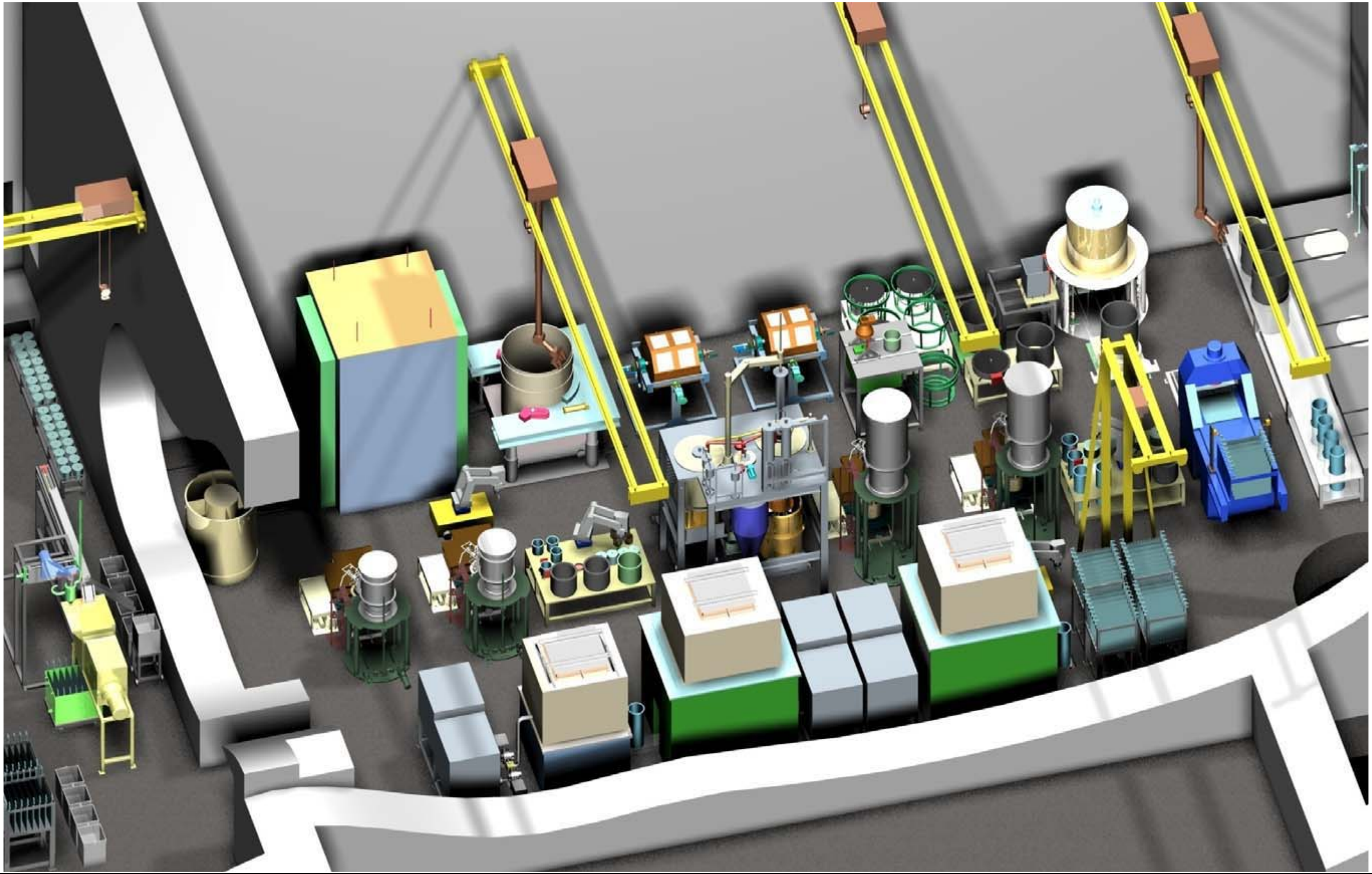
- Engineering-scale system (100 kg capacity) provides test bed for process development, optimization and scale-up
- Interleaved planar anode – cathode module
- Cathode module compatible with planar electrorefiner for follow-on uranium recovery tests
- Adjustable spacing between anodes and cathodes allows for process optimization (e.g., maximize current efficiency)
- Anode designed to allow testing of various anode materials (e.g., metals, ceramics)



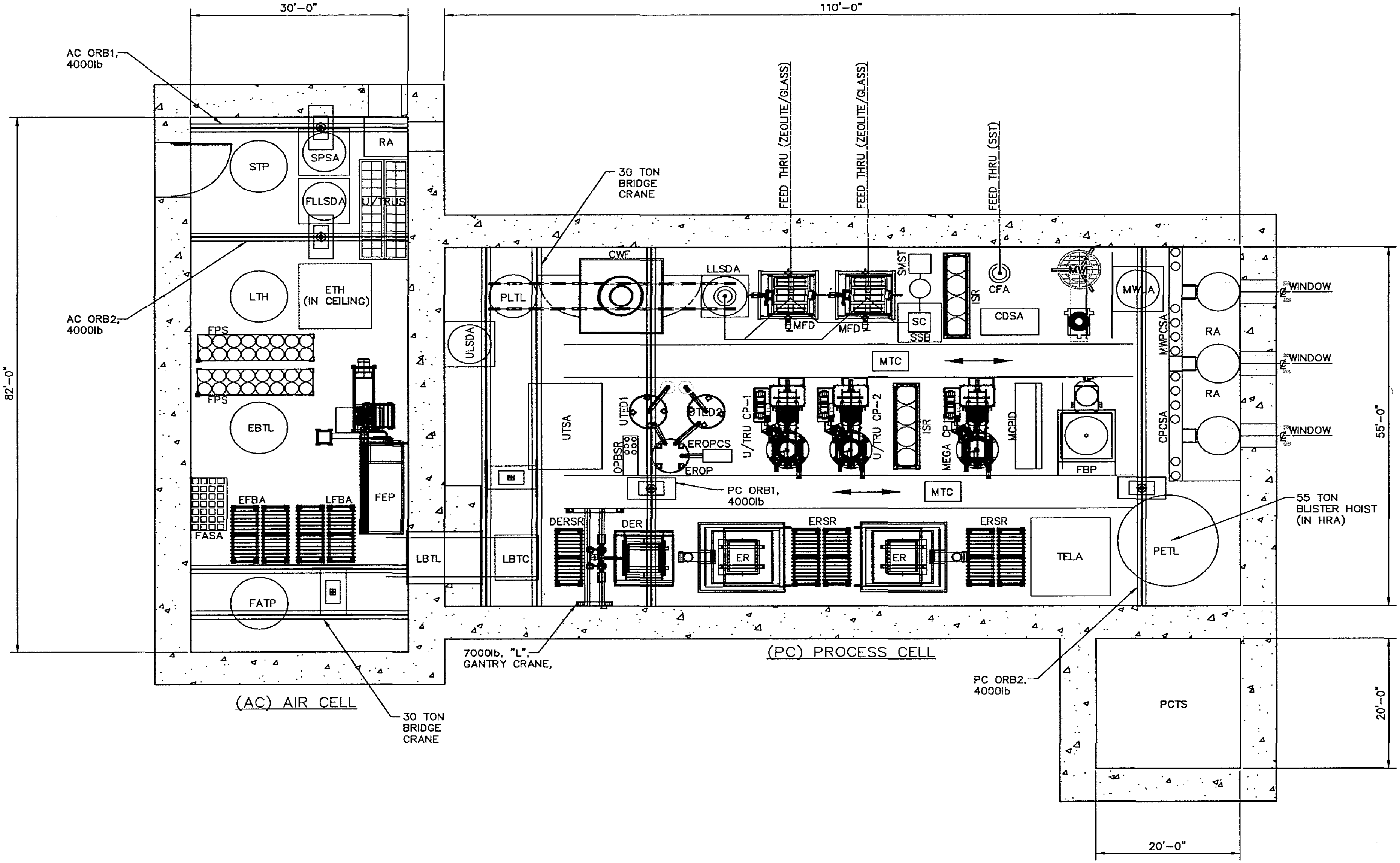
# *Pilot-scale (100 T/tr) Pyroprocessing Facility*

- ANL developed a pre-conceptual design of a 100 T/yr pyroprocessing facility for LWR spent fuel:
  - Detailed flowsheet
  - Equipment concepts
  - Operational process models
- Potentially a major economic advantage.
- Long-term repository implications

# *Pre-conceptual design of a pilot-scale (100 T/yr) LWR Pyroprocessing Facility*



# Plan View of LWR Pyroprocessing Facility (100 T/yr)



## *Capital Cost for LWR Pyroprocessing Facility*

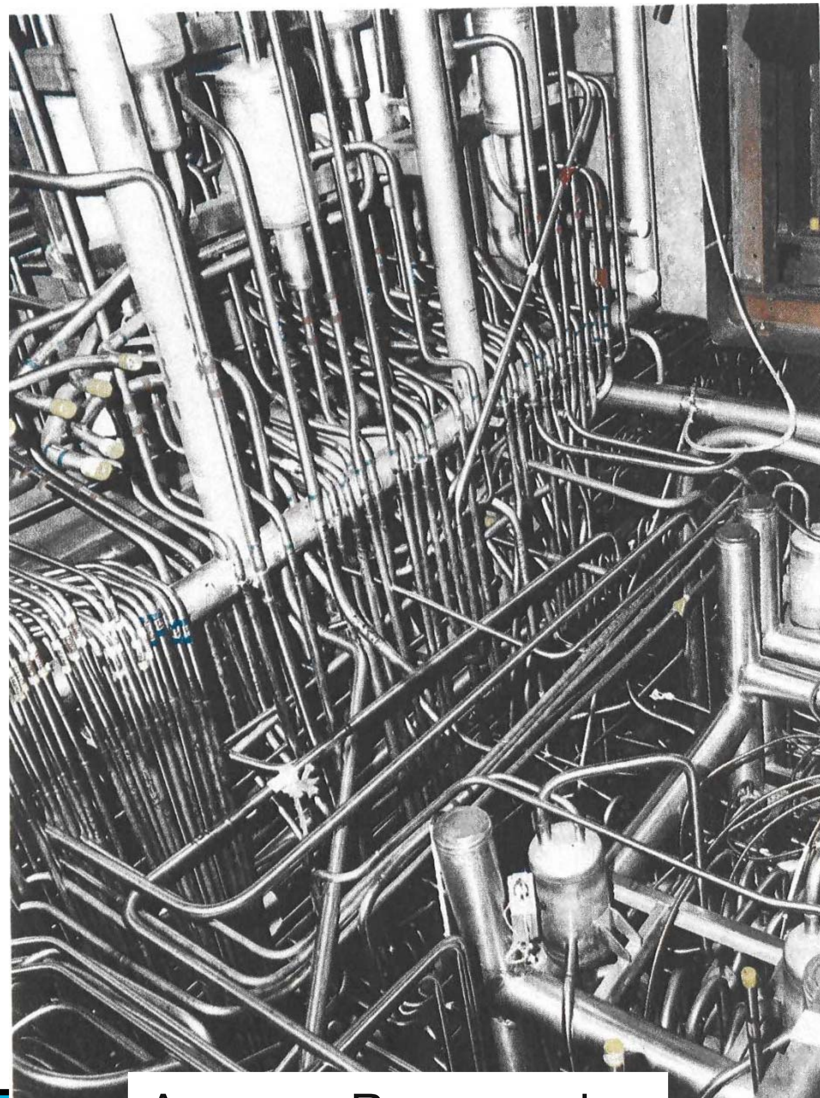
- The capital cost for the 100 ton/yr LWR pyroprocessing is estimated at:

| | |
|---|---|
| Engineering | 100 |
| Construction | 120 |
| Equipment systems | 100 |
| Contingencies | 80 |
| Total | $400 million |

- Even if the equipment systems are duplicated without any further scaleup, a commercial scale (800 T/yr) would cost about $2.5 billion, which is an order of magnitude less than the Rokkasho reprocessing plant.
- The above is a very rough estimate based on experiences of the EBR-II FCF refurbishment (<$50 million) and the Fuel Manufacturing Facility ($4 million).

# *Pyroprocessing is drastically different from conventional aqueous reprocessing*

Pyroprocessing



Aqueous Reprocessing

# *Summary on Pyroprocessing*

- Pyroprocessing has been demonstrated in engineering scale for the IFR application with significant improvements in:
  - Economics
  - Proliferation-resistance
  - Waste management
- With electrolytic reduction front-end step, pyroprocessing can be applied for the LWR spent fuel as well, but this requires a pilot-scale (~100 T/yr) demonstration to establish economics viability.
- The pyroprocessing product stream (mixture of all actinides) is not suitable for recycling in LWRs.