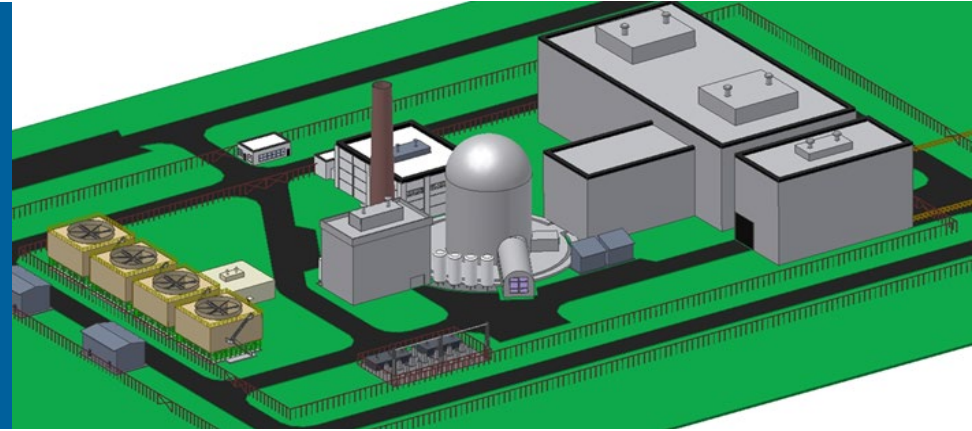


METALLIC FUEL RECYCLING



MARK A. WILLIAMSON

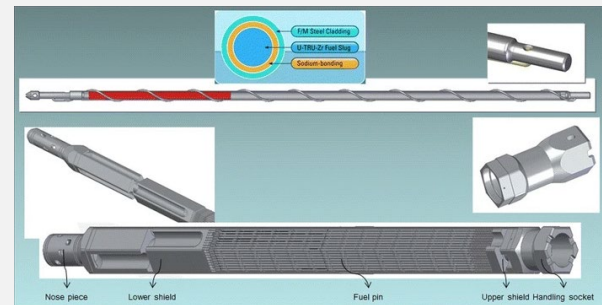
Division Director

Chemical and Fuel Cycle Technologies

Merits and Viability of Different Nuclear Fuel Cycles and Technology
Options and Waste Aspects of Advanced Nuclear Reactors
National Academy of Sciences Review Panel
December 8, 2020

USED METALLIC FUEL MANAGEMENT OPTIONS

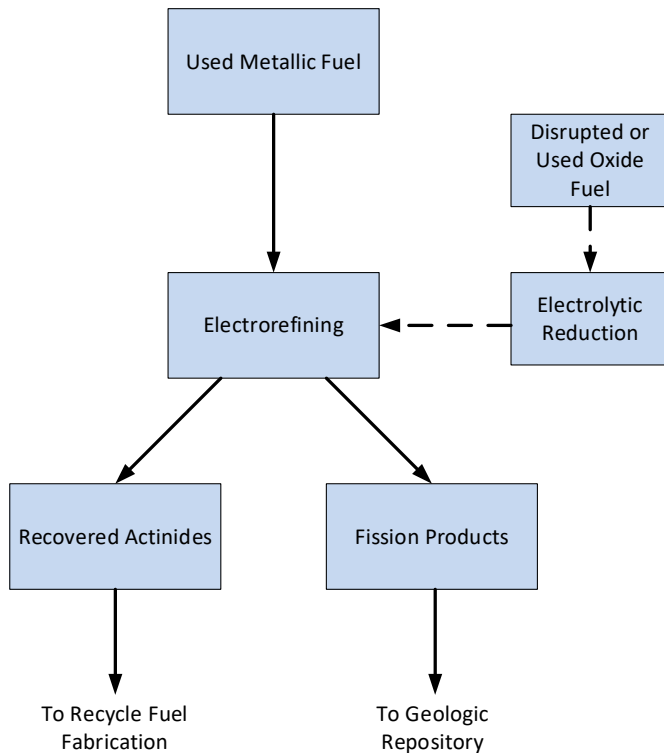
- Disposal
 - Repository requirements may not allow direct disposal of sodium-bonded fuel due to sodium reactivity
 - Develop and implement secondary packaging to mitigate concern
 - Demonstrate treatment method to stabilize fuel
 - Develop an advanced fuel form with no sodium bond, which can be qualified using the versatile test reactor
- Recycle
 - Pyrochemical processing
 - Metal feed to metal product
 - Metallic fuel recycling development activities underway
 - Aqueous processing
 - Fuels containing zirconium metal require use of hydrogen fluoride to avoid undesired reactions
 - Requires pyroprocessing back-end to produce metal product
- Technical basis for pyrochemical recycling
 - Fuel recycle tests conducted in the 1960s and 1970s at ANL
 - Integral fast reactor program in the 1980s and 1990s
 - Experimental Breeder Reactor II fuel treatment program
 - Advanced Fuel Cycle Initiative and successor programs



- *Benefits of using metallic fuel in fast neutron spectrum reactors*
 - Outstanding neutronic performance and high burn-up (~20 at%)
 - Synergistic with passive approach to reactor safety
 - Simple, compact (demonstrated remote) fabrication process(es)
 - Compatible with pyrochemical recycling
- *Metallic fuel characteristics*
 - U-Pu-Zr alloy fuel shows good irradiation stability
 - Large fission gas plenum to accommodate high gas release
 - Na-bond in fuel-cladding gap to keep fuel temperatures within specifications

METALLIC FUEL RECYCLING

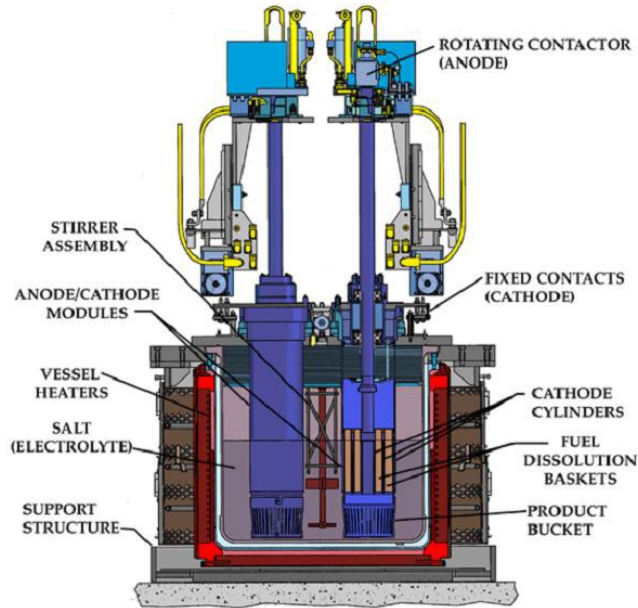
Electrorefining is the centerpiece of metallic fuel recycling



Mk IV electrorefiner circa 1996

ACTINIDE RECYCLE: NEXT-GENERATION ELECTROREFINER

Mk V Refiner



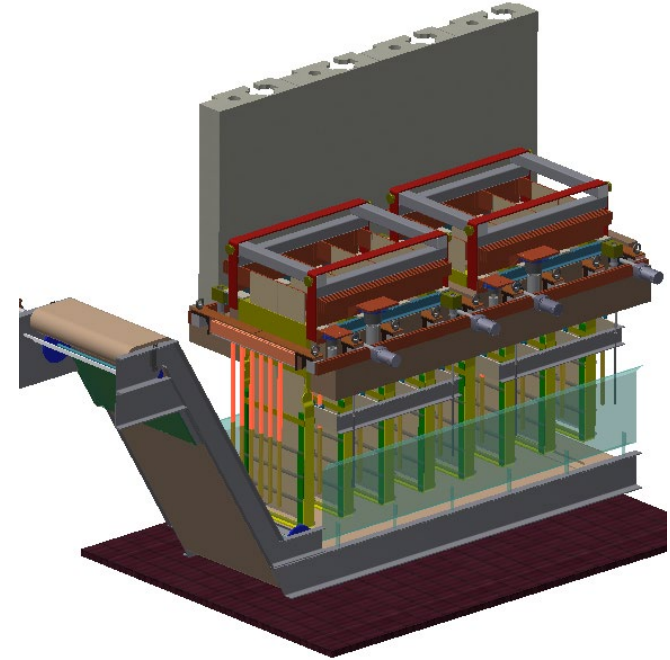
Demonstrated sustained treatment of irradiated fuel discharged from Experimental Breeder Reactor II

PEER test module



PEER viability demonstrated through testing, process inefficiencies identified in Mk V system eliminated

Commercial concept

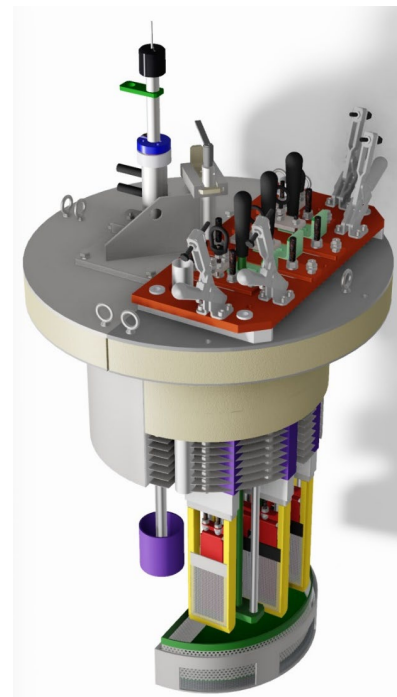


Design incorporates U and U/TRU recovery

U/TRU RECOVERY

Two technologies being developed for U/TRU recovery

- Alloy-forming liquid metal cathode (e.g., cadmium)
 - Alloying of TRU (and lanthanoid) elements shifts their deposition potential towards that of uranium
 - Process demonstrated at kilogram-scale with irradiated fuel
 - Successful recovery of U/TRU product
 - Materials compatibility issues encountered
 - Requires U/TRU separation from alloy and residual salt
- Non-alloying solid metal cathode
 - Cell current exceeds U^{3+} mass transfer current and results in co-deposition of U and TRU
 - Process demonstrated
 - U/TRU recovery demonstrated at gram-scale
 - U/Ln recovery demonstrated at kilogram-scale
 - Low melting U-TRU alloy makes metal - residual salt separation via bottom-pour feasible
- Non-alloying technology development needs include
 - Validate range of control system performance during operations
 - Demonstrate kilogram-scale co-deposition with transuranic elements and irradiated fuel



Rendering of electrorefining system used for uranium and co-deposition tests



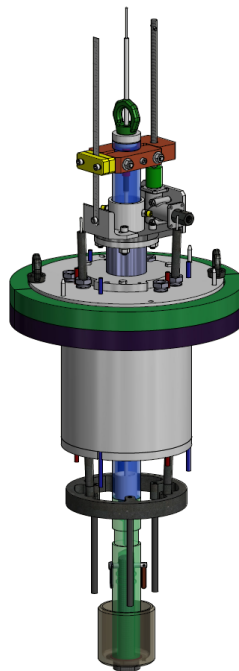
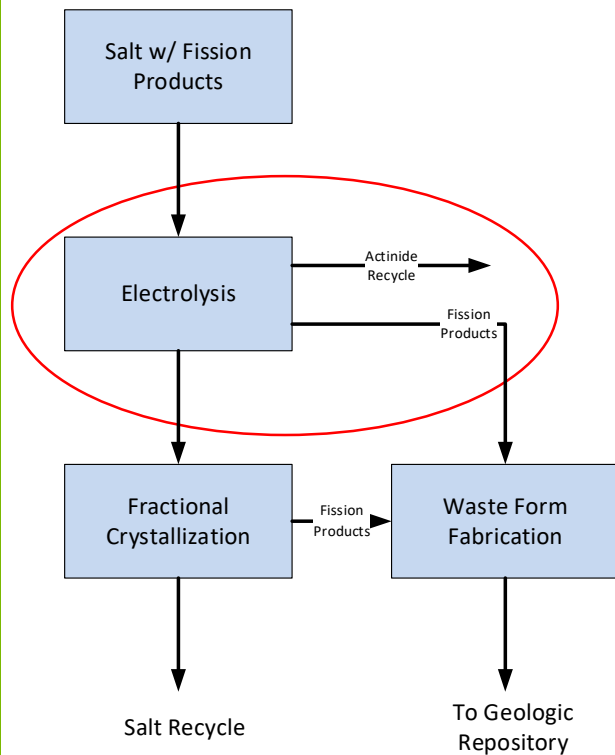
Sketch of U/TRU co-deposition module, US Patent 10,301,729

MOLTEN SALT MANAGEMENT

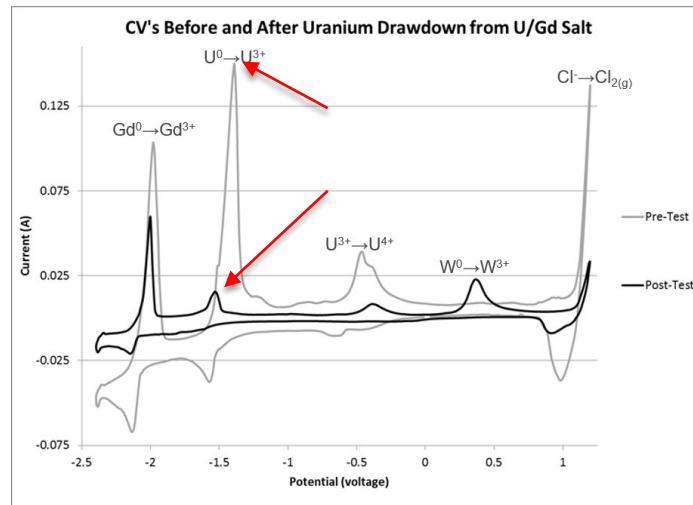
Fission product drawdown enables molten salt recycle

- Clean-up and recycle of process salt is important to implementing pyrochemical processing
 - Process salt becomes contaminated with alkali, alkaline earth, lanthanoid elements and other fission products over time
 - Maximize actinide recovery
 - Recover sufficient fission products from process salt to maintain actinide product quality and minimize waste destined for repository
 - Processes should not add complexity or significant cost to the fuel treatment facility
 - Same technologies applicable to solid and liquid fuel recycle (e.g., molten salt reactor fuels)
- Numerous process options available for fission product recovery
 - Electrolysis can be used to extract lanthanoid elements from process salt
 - Fractional crystallization technique to concentrate alkali and alkaline earth fission products in a discharge salt stream while the majority of the input salt stream is reused
 - Liquid metal – molten salt extraction for lanthanoid element recovery
- Fission product recovery test requirements
 - Identify operational parameters and their effects on the degree of separation
 - Demonstrate the extent to which fission products can be recovered from the process salt as a function of time
 - Provide a test-bed for in-situ process monitoring

EXAMPLE: ELECTROLYSIS SYSTEM



Rendering of electrolysis system for actinide and lanthanide drawdown

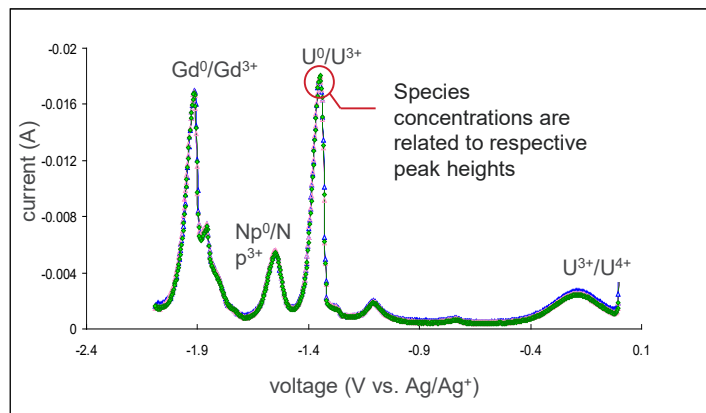


Uranium product from drawdown test

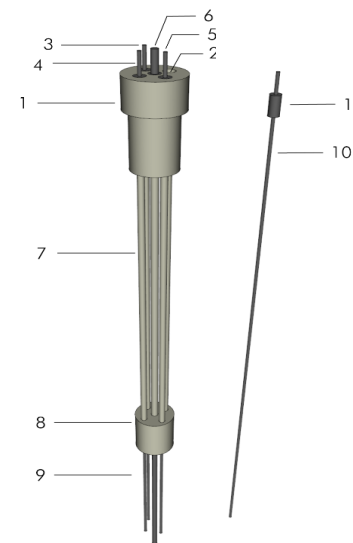
IN-SITU PROCESS MONITORING

Electrochemical techniques enable process monitoring and control, and supplement nuclear material accounting methods

- Actinide concentration and other salt characteristics determined from current response to voltage waveforms
 - Allow rapid, real-time measurements
 - Equipment not affected by high radiation background
 - Technique based on electrochemical properties of salt and does not require use of standards
 - Well-developed theory and methods for voltammetric response for a given redox reaction are available¹⁻⁵



Square-wave voltammogram for molten salt containing UCl_3 , NpCl_3 and GdCl_3

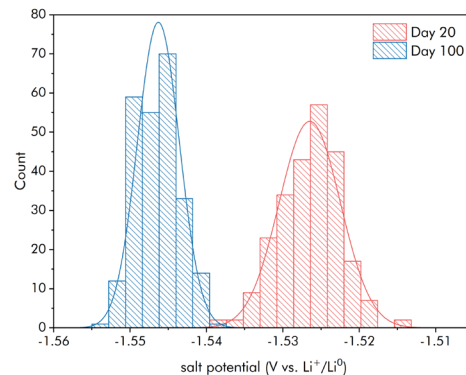
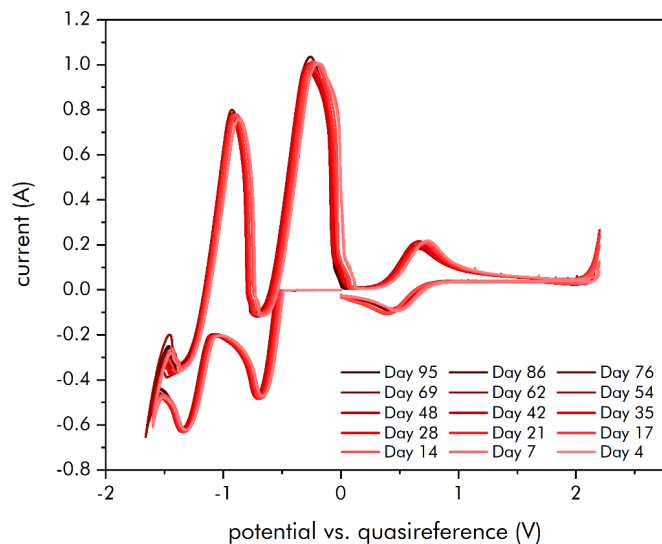


Multi-electrode sensor

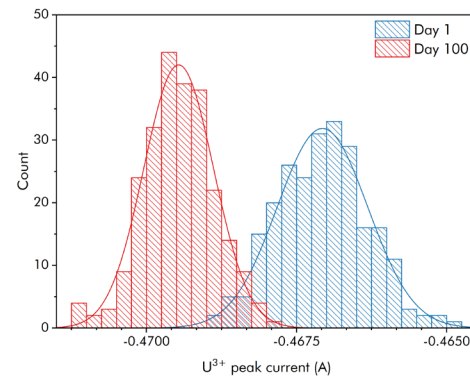
¹Hill, Perano, Osteryoung, *JES* **107** (8) 698 (1960)
²Mamantov, Manning, *Anal. Chem.* **38** (11) 1494 (1966)
³Tylka, et al. *JES* **162** (9) H625 (2015)
⁴Tylka, et al. *JES* **162** (12) H852 (2015)
⁵Hoyt, et al. *JES* **164** (2) H134 (2017)

100-DAY SENSOR DEMONSTRATION

Data collected over 100 day test in the pilot-scale electrorefiner reveals outstanding reproducibility and repeatability



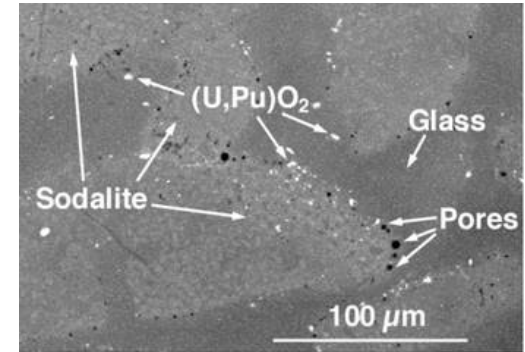
Salt potential measurement mean absolute deviation over the entire test of less than 15 mV



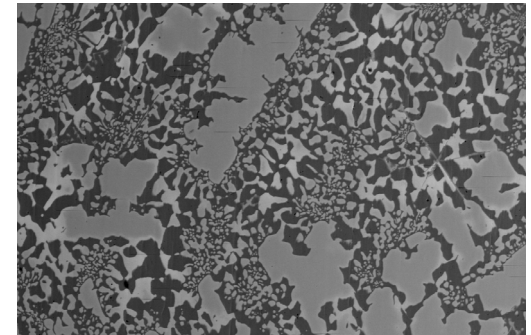
UCl_3 peak current deviation over the entire test of 1.08%

WASTE FORMS

- Ceramic Waste Form (CWF)
 - Chemistry of CWF production, waste loading limits, and process control are well understood
 - Test methods to optimize waste forms and assess performance have been developed and demonstrated
 - Performance model based on binder glass durability has been developed
- Metal Waste Form (MWF)
 - MWF formulation ensures stable passivated surface under range of environmental conditions possible during disposal (primarily Eh, pH, Cl)
 - Test methods to optimize production and assess performance have been developed and demonstrated for steel-based MWF
 - Performance model based on corrosion resistance being developed for steel-based MWF with additives
- Off-gas capture
 - Off-gases captured in process hot cell atmosphere system
 - Radioactive fission product gases packaged for decay storage



Glass-encapsulated sodalite with Actinides



*HT9-based MWF (for steel cladding)
EBR II experience*

TECHNOLOGY DEVELOPMENT

Metallic fast reactor used fuel recycling RD&D needs

- Recycle technology industrialization
 - Demonstrate actinide and fission product recovery processes to facilitate salt recycle and waste minimization
 - Method and sensor development for process monitoring and control, and nuclear material accounting
 - Close-coupled testing of unit operations to assess product recovery efficiency and product quality
 - Demonstrate performance of unit operations with irradiated used nuclear fuel
 - Update flowsheet modeling capabilities with process models developed from experimental tests
 - Develop conceptual design for fuel recycle facility
- Develop and demonstrate disruptive technologies that may exceed recycle mission goals

ACKNOWLEDGMENTS

- Financial support provided by
 - U.S. Department of Energy, Office of Nuclear Energy
 - Landmark Foundation
- Government License Notice - the manuscript has been created by UChicago Argonne, LLC, Operator of Argonne National Laboratory (“Argonne”). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357.



Argonne National Laboratory is a
U.S. Department of Energy laboratory
managed by UChicago Argonne, LLC.

