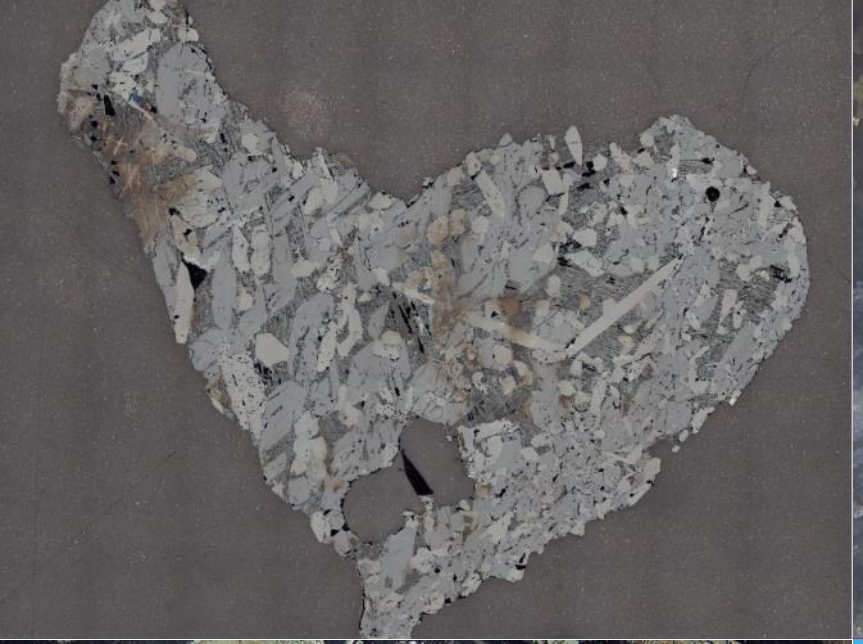

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# EBR-II Pyroprocessing Salt Waste Form Studies

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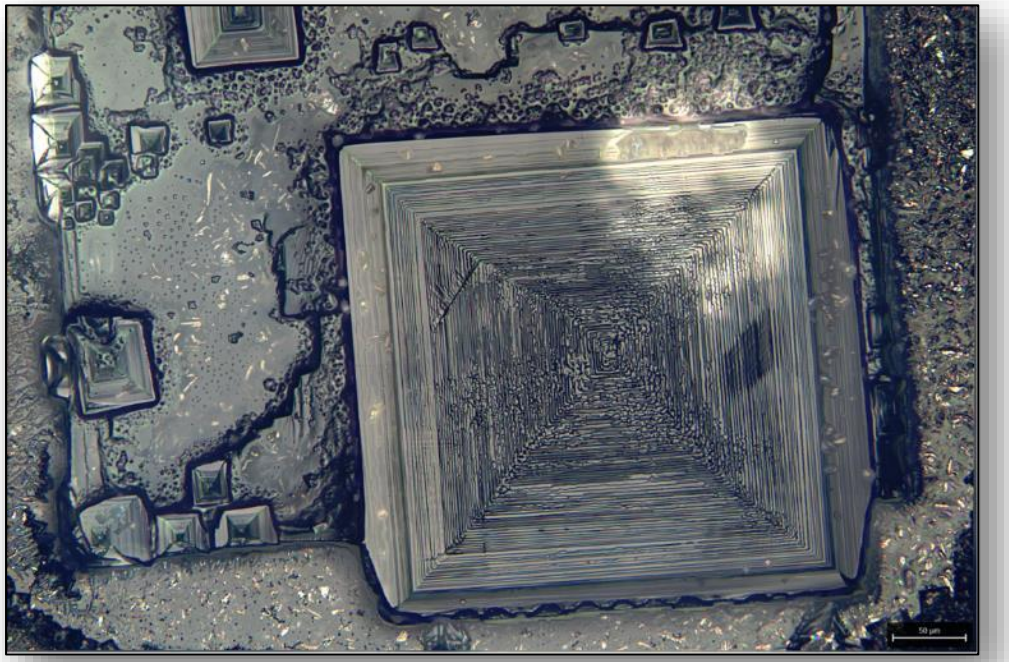
Battelle Energy Alliance manages INL for the
U.S. Department of Energy's Office of Nuclear Energy

Idaho National Laboratory

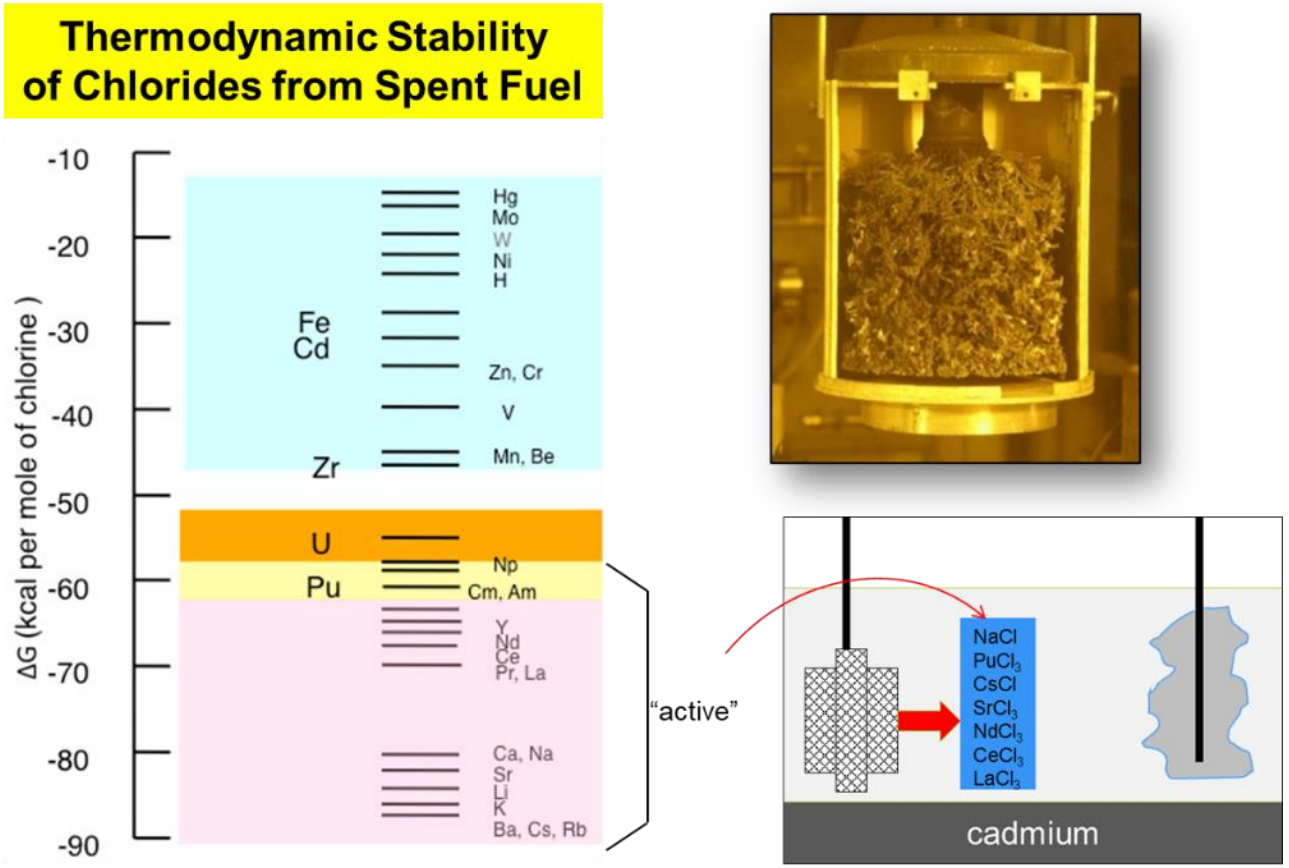
# Overview

- Electrorefiner (ER) salt chemistry
  - Fission product partitioning
  - ER salt processing
- Waste Forms
  - Advanced Ceramic
  - Silica-Alumina-Phosphate
  - Iron Phosphate Glass
- Characterization
  - Baseline vs durability testing (gamma irradiation)
    - Imaging
    - Leaching
    - Electron microscopy
    - Elemental/isotopic analysis
- Conclusion and Acknowledgements

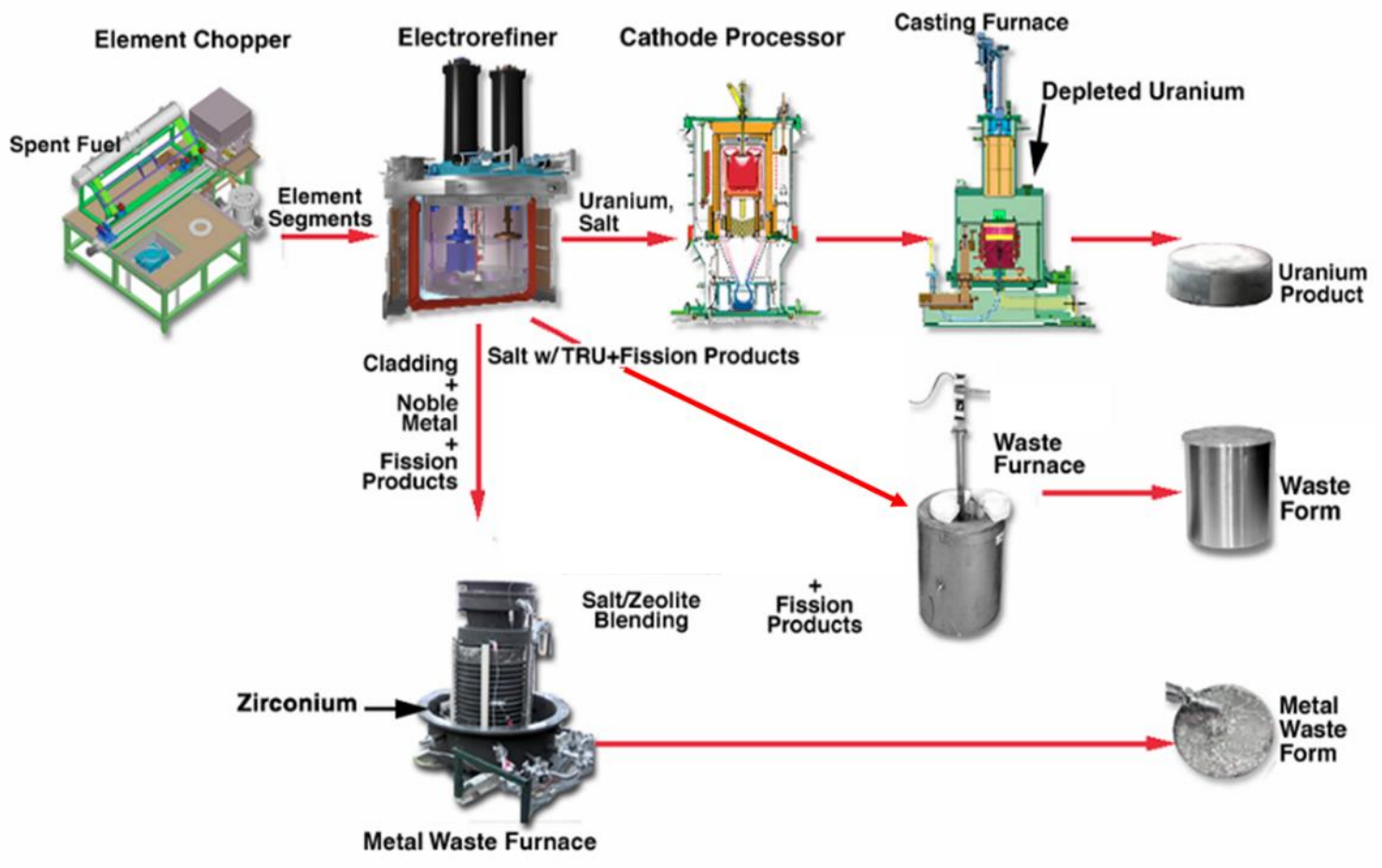


# Salt Chemistry

- Buildup of active metal chlorides limits lifetime of salt
  - Solubility
  - Melting point
  - Criticality
  - Heat generation
- When concentration limits are reached, salt needs to either be disposed or purified and recycled
- Recycling can minimize volume of waste generated
- Two steps:
  - U/TRU drawdown
  - FP separation
- This is an active area of research



# Salt Chemistry



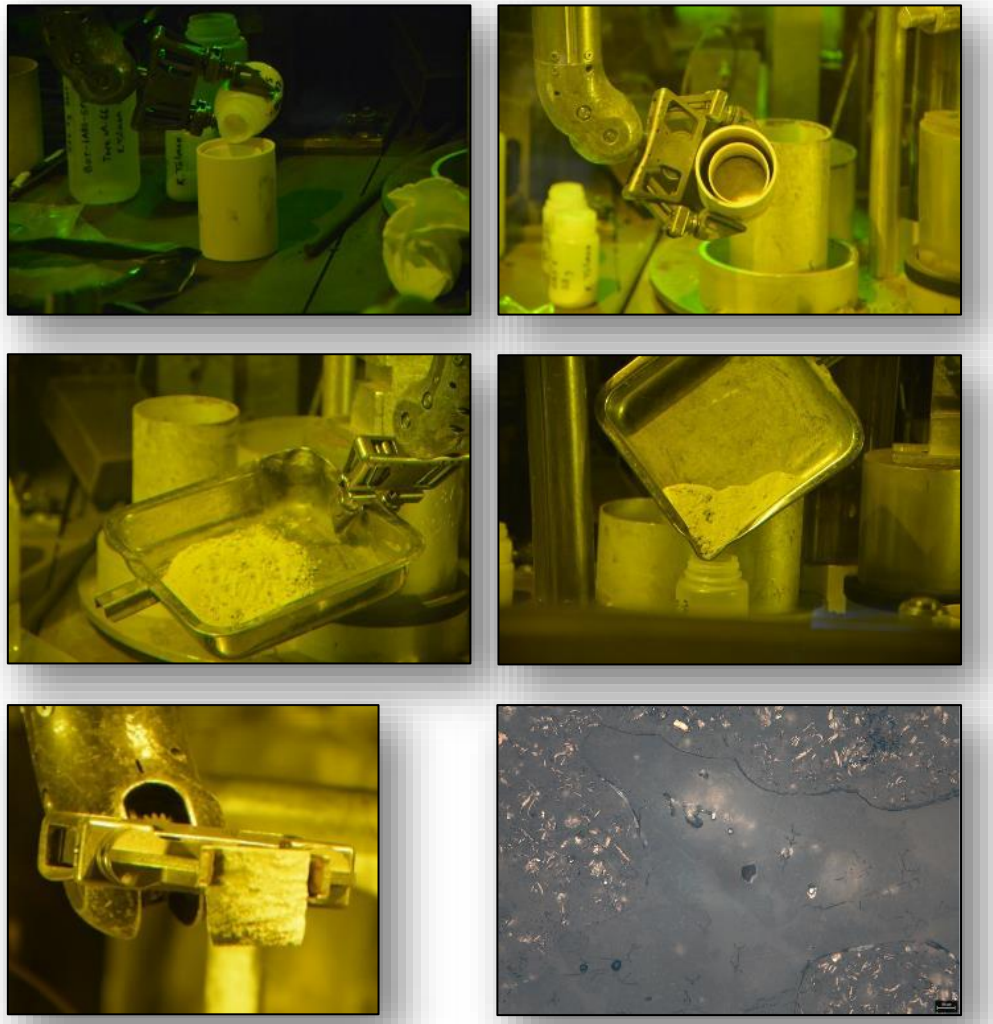

Flowsheet for EBR-II Spent Fuel Treatment

| | Mark-IV ER (driver) |
|---|---|
| | Wt % of chloride salt |
| LiCl/KCl | 69.14 |
| NaCl | 12.16 |
| RbCl | 0.23 |
| $SrCl_2$ | 0.65 |
| $YCl_3$ | 0.47 |
| CsCl | 1.66 |
| $BaCl_2$ | 0.86 |
| $LaCl_3$ | 0.82 |
| $CeCl_3$ | 1.55 |
| $PrCl_3$ | 0.77 |
| $NdCl_3$ | 2.63 |
| $PmCl_3$ | 0.03 |
| $SmCl_3$ | 0.50 |
| $EuCl_3$ | 0.03 |
| $NpCl_3$ | 0.08 |
| $UCl_3$* | 5.81 |
| $PuCl_3$* | 2.57 |

*Concentration dependent on electrorefiner operating conditions and are typically lower

# Advanced Ceramic Waste Form (ACWF)

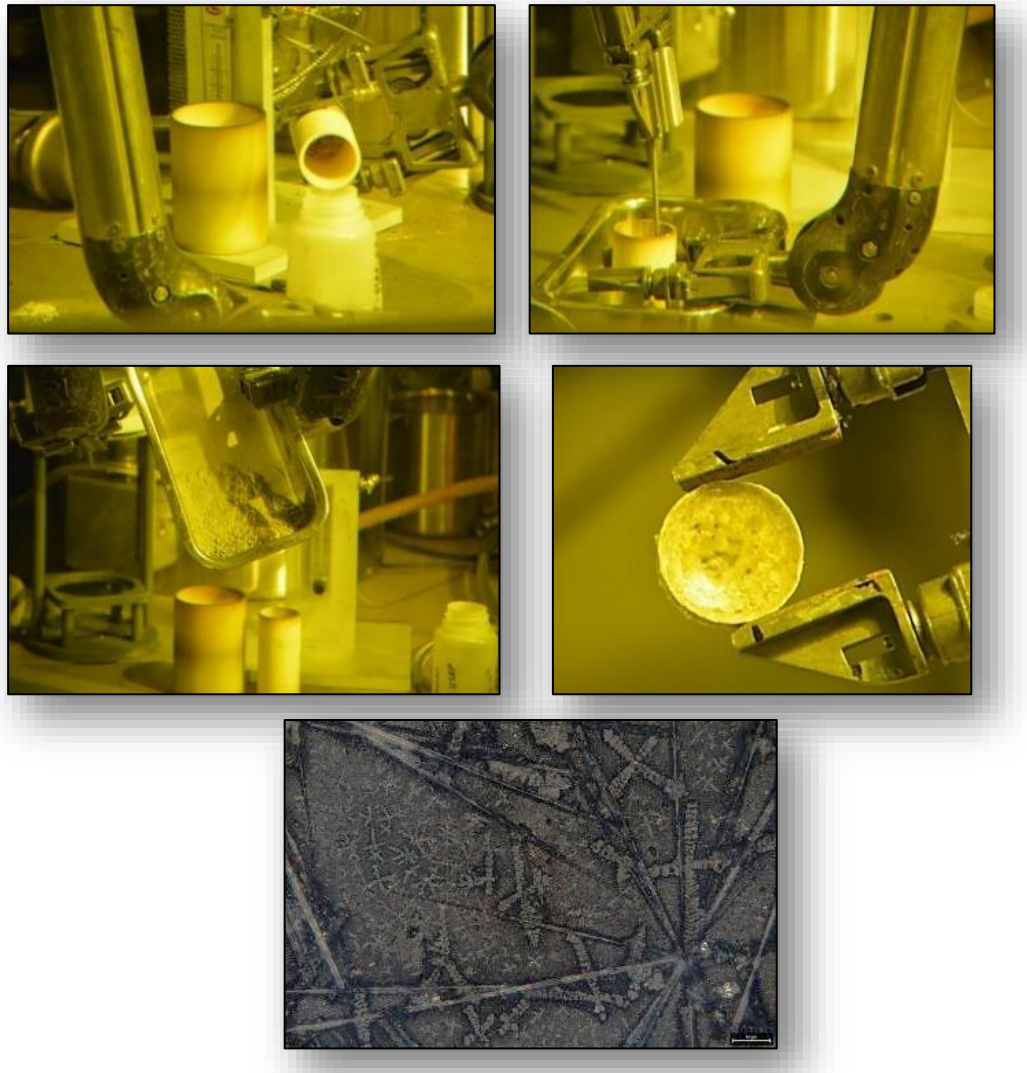
- Used to immobilize salt from
  - Oxide Reduction (OR), LiCl
  - Electrorefining (ER), LiCl-KCl eutectic
- Salt waste loading of ~10 – 14% by mass
  - Used NBS-4 glass binder for higher waste loading and increased durability over CWF
- Demonstration
  - HFEF (oxide fuel) ER salt combined with zeolite and NBS-4 glass frit
  - Heated to 500°C to occlude the salt in the zeolite
  - Cooled, mixed, heated to 500°C
  - Cooled, mixed, heated to 915°C
  - Cooled to form final ingot



HFEF = Hot Fuel Examination Facility

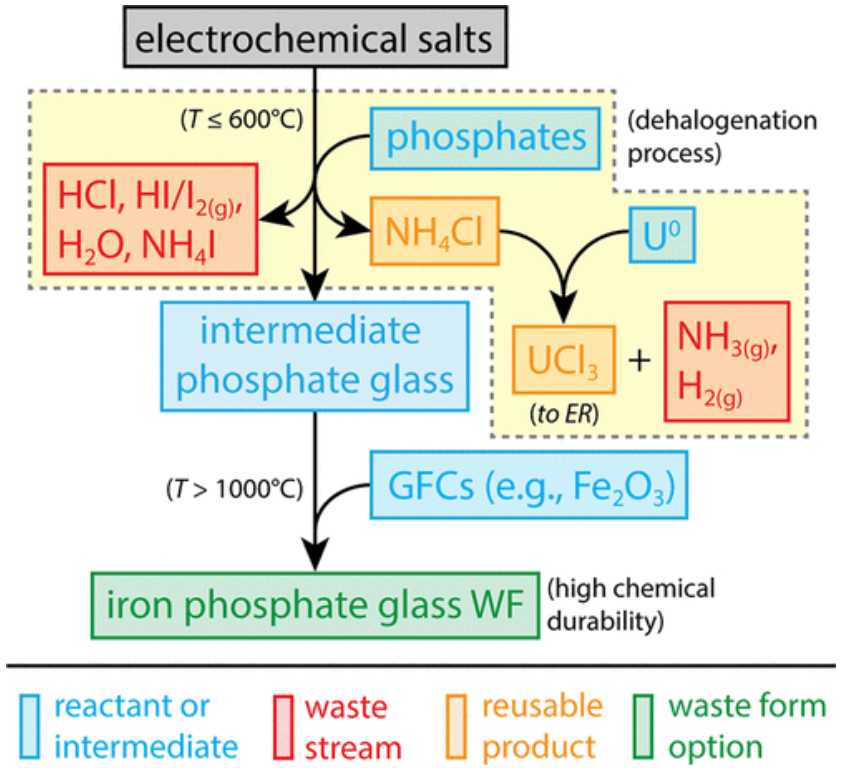
# Silica-Alumina-Phosphate (SAP)

- Used to immobilize salt from
  - Oxide Reduction (OR), LiCl
  - Electrorefining (ER), LiCl-KCl eutectic
  - Salt waste loading of ~14% by mass
- Demonstration
  - HFEF (oxide fuel) ER salt combined with SAP binder
  - Heated to 650°C to dehalogenate the salt
  - Cooled, mixed, and heated to 1200°C
  - Cooled to form final ingot



# Iron Phosphate Glass

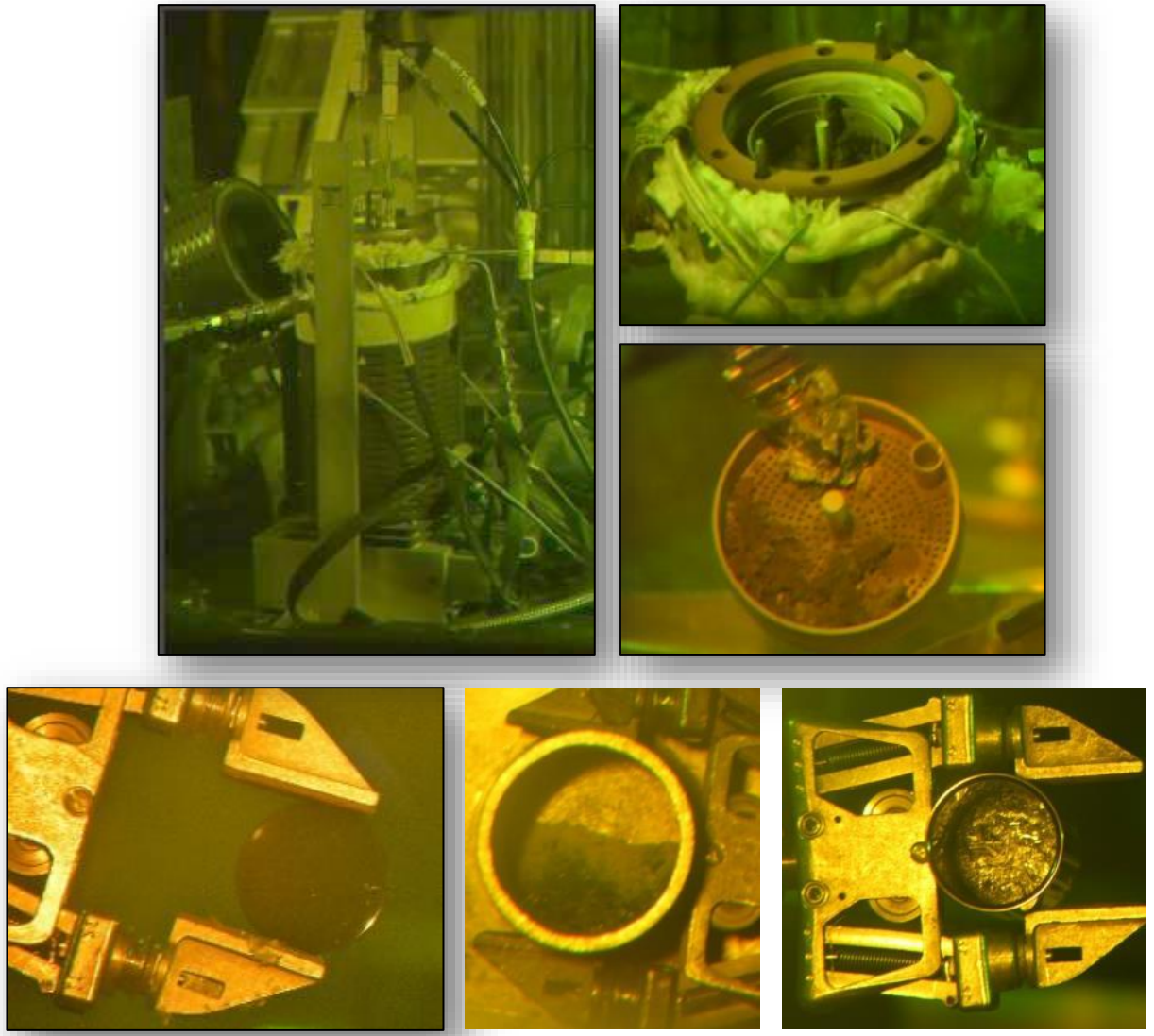
- Two-step process for dehalogenating the salt and immobilizing the remaining fission products
- More complex process, but results in a smaller overall waste volume for disposition
- Cl gas by-product is reacted with U dendrites to produce $UCl_3$ for recycle into the ER




Riley, Brian J. "Electrochemical Salt Wasteform Development: A Review of Salt Treatment and Immobilization Options". *Industrial & Engineering Chemistry Research* **2020** *59* (21), 9760-9774. DOI: 10.1021/acs.iecr.0c01357

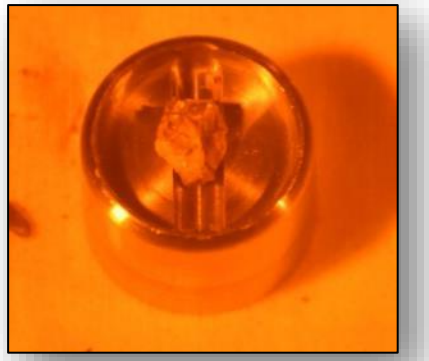
# Iron Phosphate Glass

- Demonstration
  - Ammonium phosphate combined with FCF ER salt,
  - Heated to 550°C, cooled, crushed
  - Combined with iron oxide
  - Heated to 1050°C
  - Cooled using a controlled profile in place of quenching to form final ingot

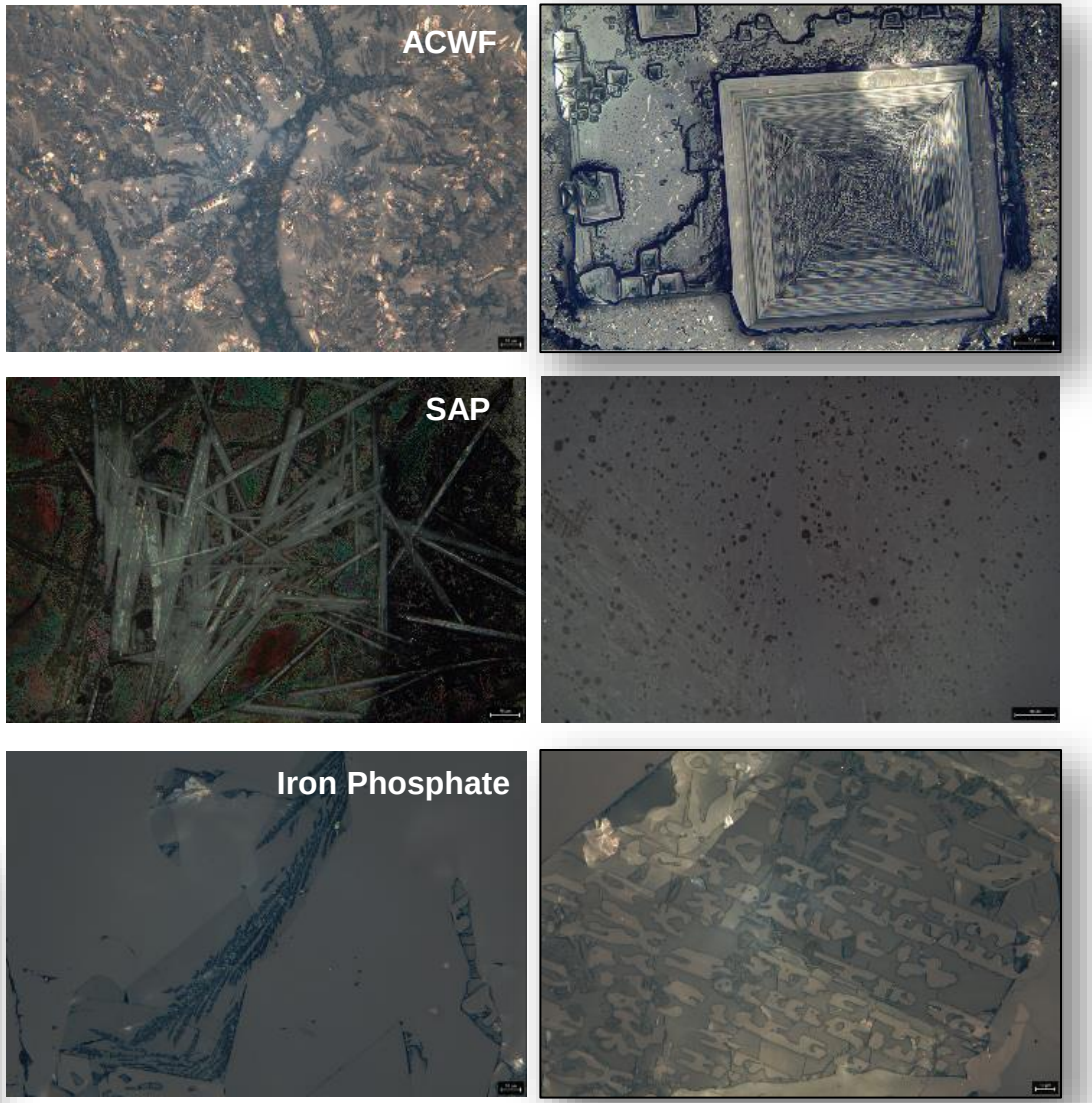


# Benchmark Characterization

- Examining three waste forms
  - ACWF
  - SAP
  - Iron Phosphate
- Waste forms were prepared by incorporating ER salt from HFEF and FCF
- Benchmarking characterization is ongoing but includes:
  - **Optical microscopy**
  - Leaching
  - Electron microscopy
  - Elemental/isotopic analysis

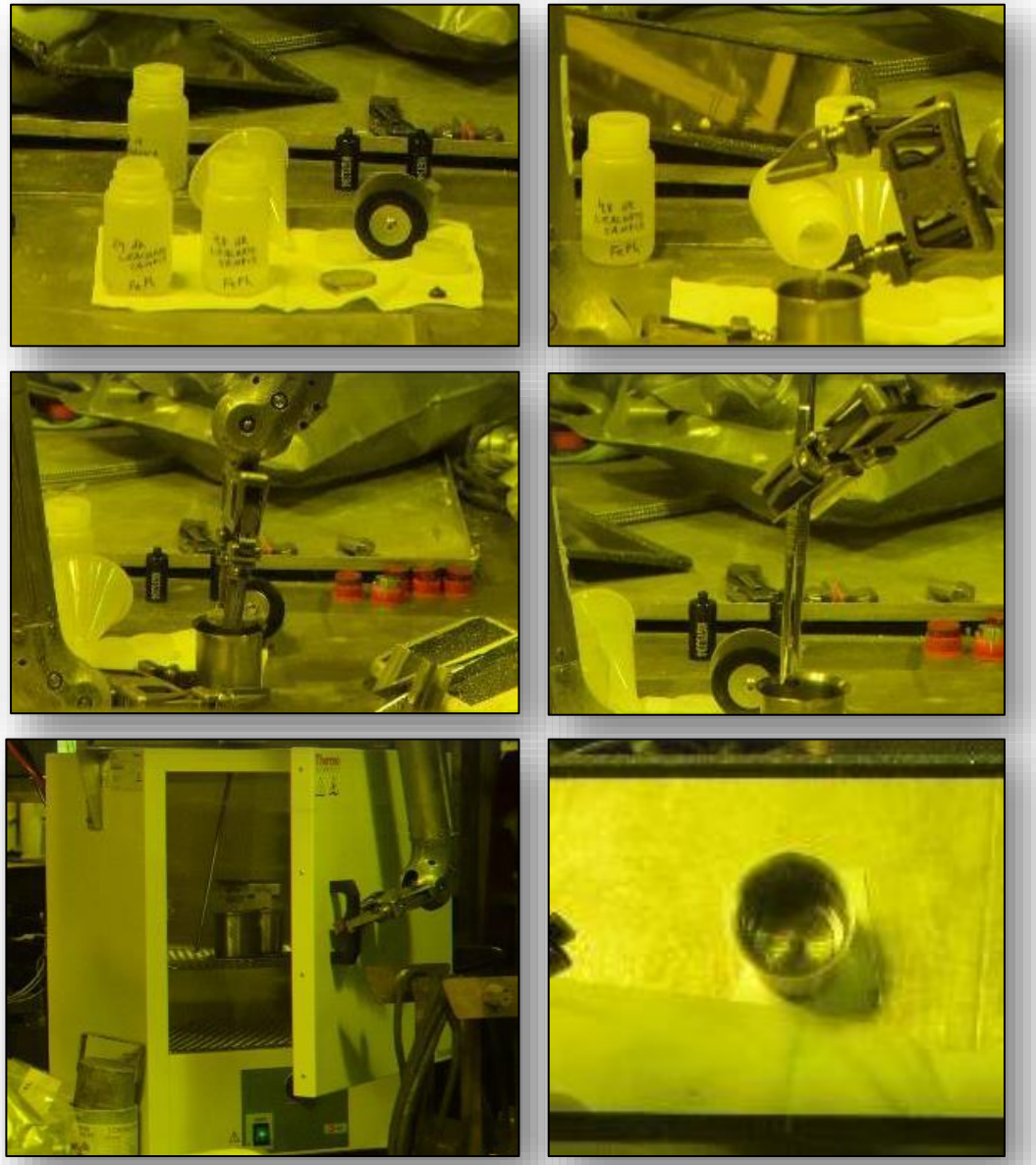


Waste form sample preparation



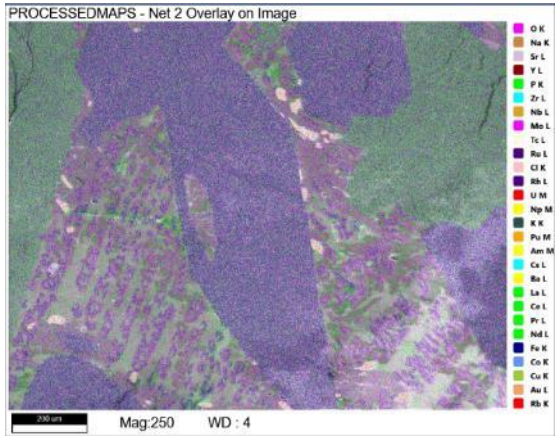
# Benchmark Characterization

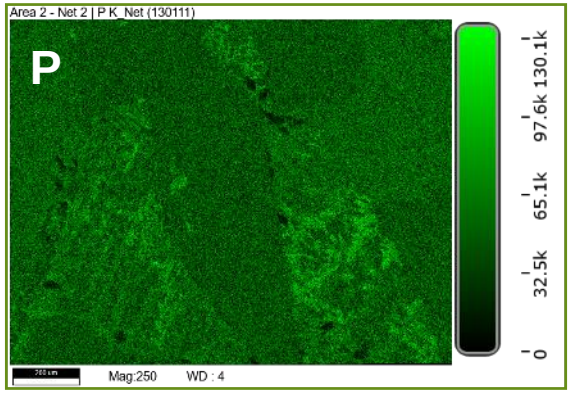
- Benchmarking characterization is ongoing but includes:
  - Optical microscopy
  - **Leaching**
  - Electron microscopy
  - Elemental/isotopic analysis
- Leachate testing
  - Established capability within HFEF hot cell
  - ASTM-C1308 compliant and analogous to surrogate testing capabilities
  - Basis of comparison is Savannah River high-level “EA glass”

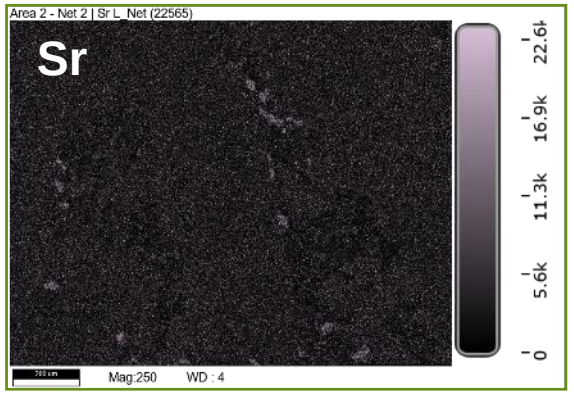


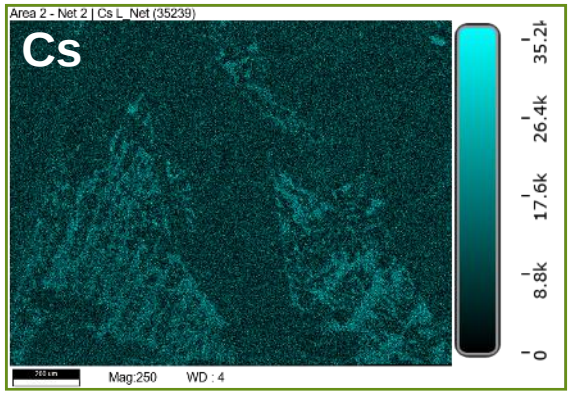
# Benchmark Characterization

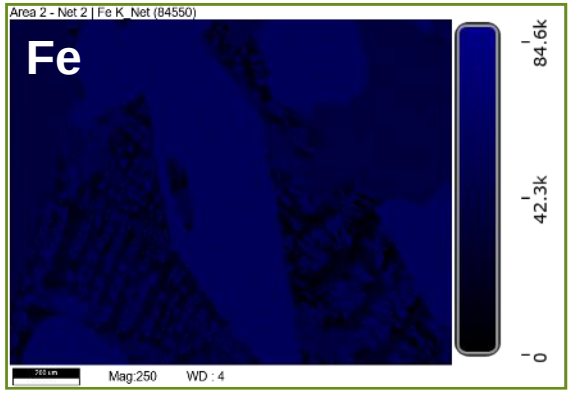
- Benchmarking characterization is ongoing but includes:
  - Optical microscopy
  - Leaching
  - **Electron microscopy**
  - Elemental/isotopic analysis
- Iron Phosphate (preliminary data)
  - Several phases present
  - Concentrations of Sr
  - Cs found with P
  - Some U-enrichment along phase boundary
  - Several other elements not shown

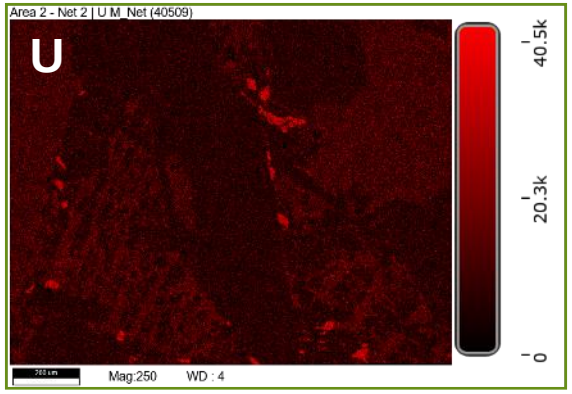


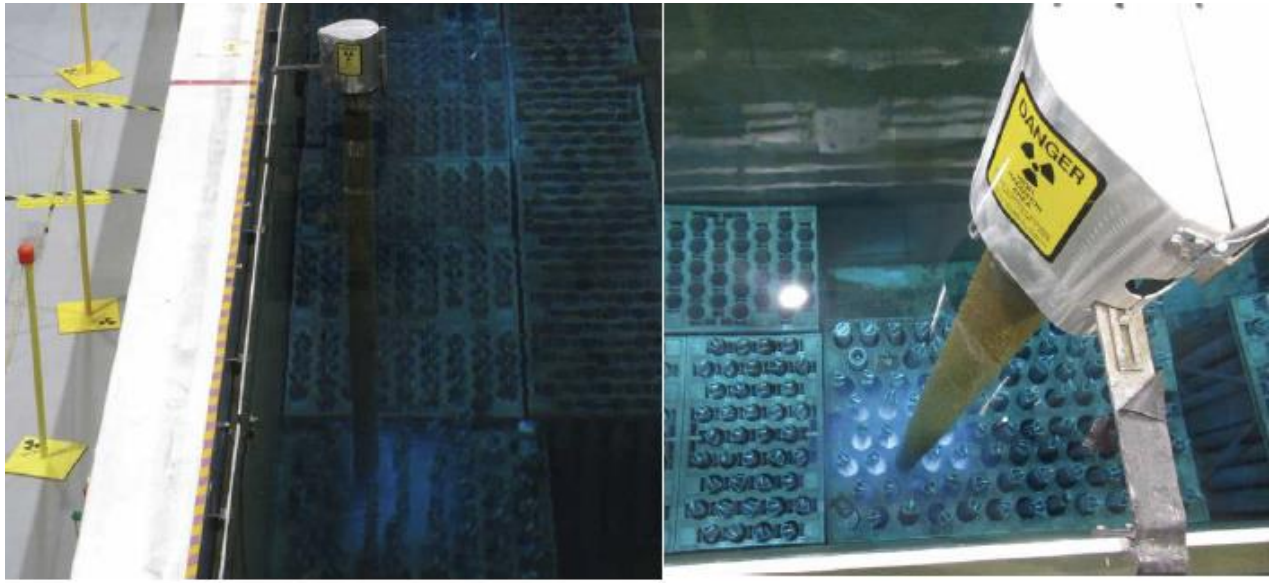



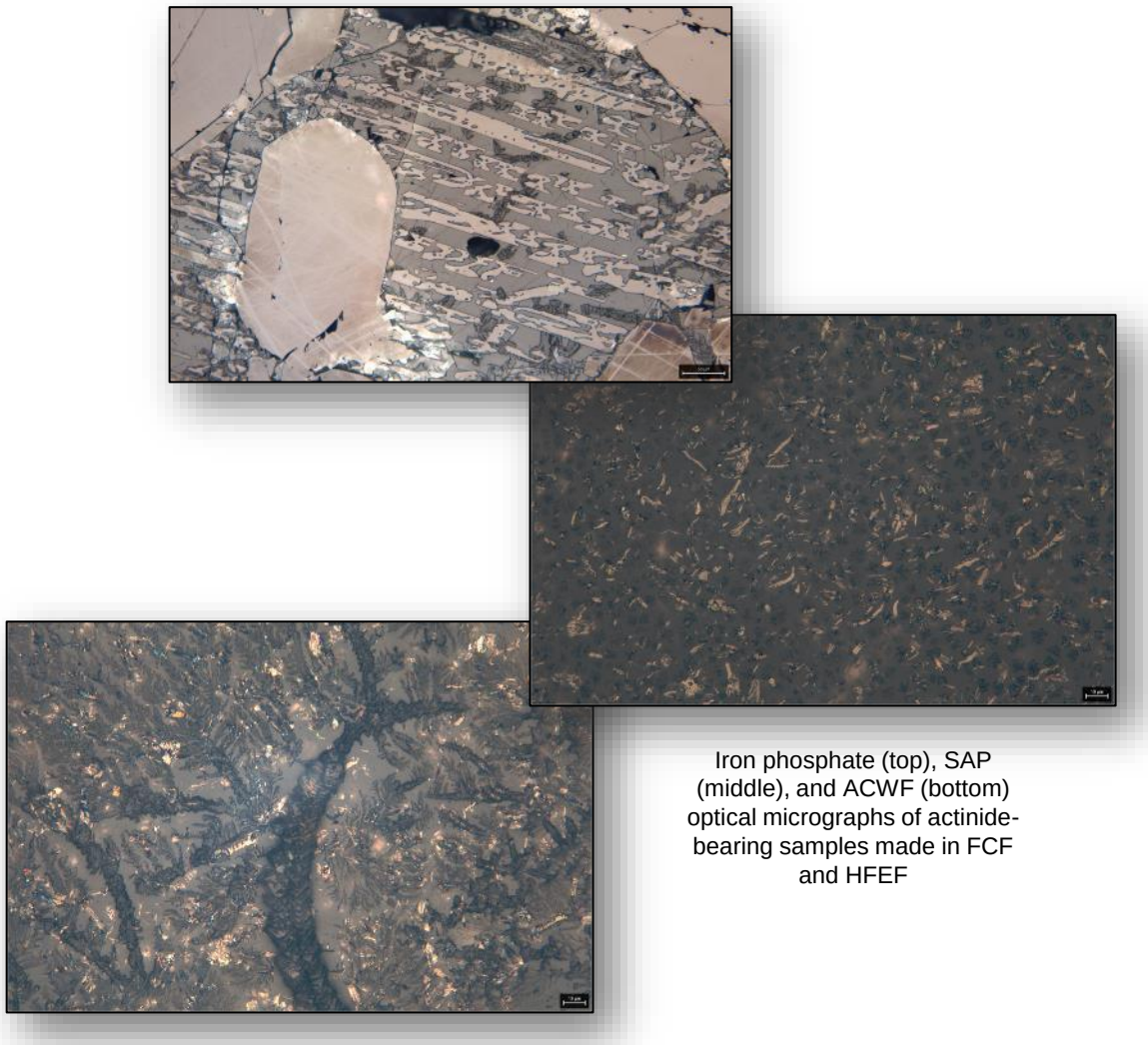
# Durability Testing (Irradiation)

- Waste forms samples will be placed in the Advanced Test Reactor (ATR) spent fuel pool gamma tube
  - Gamma field is $5 \times 10^6$ rad/hr
  - Testing of waste form durability in a high radiation environment and at elevated temperatures (ambient, 100°C, 500°C)
- Will perform characterization of irradiated waste form samples
  - Optical microscopy
  - Elemental/isotopic analysis
  - Leaching
  - Electron microscopy
- Compare characterization results from benchmark and irradiated samples




# Conclusions

- Processing spent fuel in an ER results in accumulation of fission products and actinides in the salt phase
- The salt must be periodically purified or disposed and replaced with clean salt
- Several waste form options are being evaluated for development and deployment
- Extensive characterization techniques are in use at INL to fully describe the waste form products
- Waste form durability testing (i.e. gamma irradiation, leaching) is ongoing



Iron phosphate (top), SAP (middle), and ACWF (bottom) optical micrographs of actinide-bearing samples made in FCF and HFEF

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