Optimization of Factors Governing the Partitioning of Radionuclides during Melt Decontamination of Stainless Steels

Sandia National Laboratories

- ESR furnace experiments
  - Surrogate oxides packed into tube melted with electrode
  - Various slag compositions
  - Utilize split mold to study surrogate partitioning to slag skin and cap
  - Melted stainless steel pipe electrodes to evaluate process stability for controls development

- Laboratory thermochemical measurements
  - At BU, construct small cell to measure thermodynamics and kinetics of slag/surrogate reactions
  - Characterize slag physical properties (electrical conductivity and viscosity)

- Thermochemical modeling
  - Free Energy Minimization modeling of each metal/slag system
    - Surrogates
    - Uranium

- Modeling predicts that
  - Cerium and Lanthanum will be removed from the steel by the formation of fluor[illegible] oxides, and oxy-aluminates
  - Uranium oxides react with slags to form fluorides

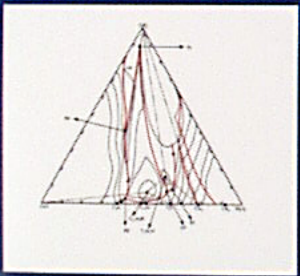
- Partnerships for ESR Melt Decontamination
  - Install and operate ESR furnace at K-26 in Krasnoyarsk, Russia
    - Funded by International Proliferation Prevention Program to recycle Pu-contaminated stainless steel piping
  - Develop advanced ESR controls for melt decontamination
    - Funded by the Amarillo National Resource Center for Plutonium
    - Control optimization by Prof. Joe Beaman, UT Austin

Section of slag skin shows variations in phase structure against mold (left) and ingot (right).

Detailed analysis of slag skin phases performed using new EBSP technique.

ESR controls experiment used electrode fabricated of concentric stainless steel piping.

- **Authors and Participants:**
  - Sandia National Laboratories
    James Van Den Avyle, (505)845-3105, javande@sandia.gov
    Eric Schlienger
    Dave Melgaard
    James Stepanek
    Greg Shelmidine
  - Boston University
    Prof. Uday Pal, (617)353-7708, upal@bu.edu
    William Chernicof



Phase equilibria for the slag system CaF, CaO, Al2O3

Split ESR mold allows slag skin sampling; mold wall thermocouples measure the melt thermal history.

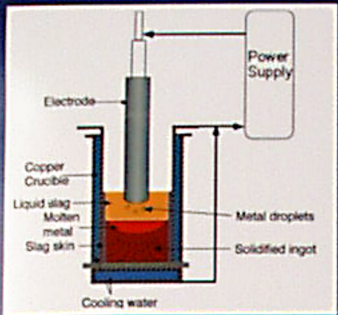
- **Advantages of Electroslag Remelting for Melt Decontamination:**
  - Maintains the pedigree of the high value metals
  - Produces an ingot suitable for direct forming
    - Reduced contamination
    - No shrinkage porosity
  - Captures radionuclides in a stable, solid slag mass
  - Furnaces are easily enclosed and use no refractories

Surrogate Selection

- Surrogates were selected to mimic the behavior of radionuclides

- Criteria
  - Thermochemical properties
  - Physical properties
  - Availability

- Choices
  - Ce for U and Pu
  - La and Nd for Pu
  - Nd for Am, Cm, and Nd (fission product in spent fuel)
  - Ce, La, and Nd for all rare earth fission products



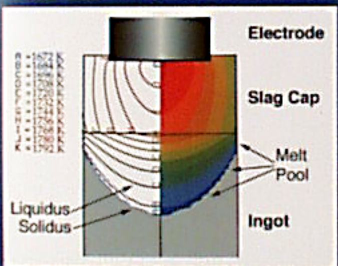
In Electroslag Remelting, a metal electrode is melted through a resistively heated bath of molten slag. During the passage of metal drops through the slag, thermochemical and electro- chemical reactions occur.

- **Goals**
  - Characterize the thermodynamics and kinetics of slag/surrogate reactions in the Electroslag Remelting (ESR) process
  - Determine capacity of slags for radionuclide containment
  - Minimize levels of contamination in processed metal

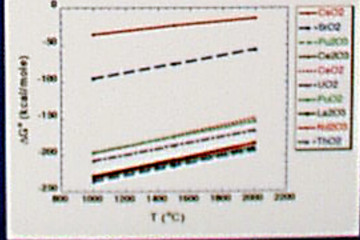
- **Approach**
  - Apply ESR process models to characterize slag thermal environment
  - Experimentally determine thermodynamics and kinetics of slag/surrogate reactions
  - Characterize surrogate removal in ESR furnace via slag reactions
  - Optimize ESR process controls for solid and pipe electrode geometries
  - Evaluate slag additives to insure waste form stability
  - Perform off-gas sampling and analysis

- In Electroslag Remelting (ESR), the slag, composed of molten oxides and fluorides, acts as a refining medium. The chemistry of the slag controls radionuclide separation.

- Mechanisms involved in refining are:
  - Thermochemical reaction
  - Electrochemical reaction
  - Mechanical entrapment of inclusions



ESR process model predicts temperatures and fluid flow patterns present during melting and solidification.



Relative stability of metal oxides are predicted by free energy modeling; lower curves are more stable.