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Objectives

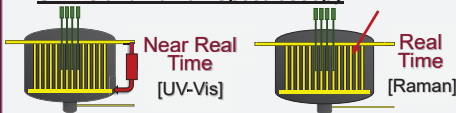
To assess the molecular structures and chemistry that govern **optical indicators** of molten salt reactors (MSRs) by establishing molten salt spectroscopic monitoring capabilities and leveraging them for Raman spectroscopic interpretation to develop online monitoring capabilities.

Introduction

Online Monitoring

- Provides **electronic** and **vibrational** signatures to characterize and monitor in real-time species in the core
- Key benefits are nondestructive data collection, radiation worker dose reduction, and proliferation risk minimization

UV-Vis and Raman Spectroscopy



Produces **elemental specific** information for actinides and other fission products such as U, Pu

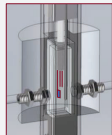
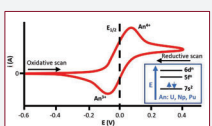
Probes the **chemical environment**, gives overall geometry and speciation information

Questions

- What chemistry is taking place that can contribute to **corrosion and precipitation effects**?
- How does the temperature and concentration of the analyte affect the **speciation** of the metal in the melt?

Plan

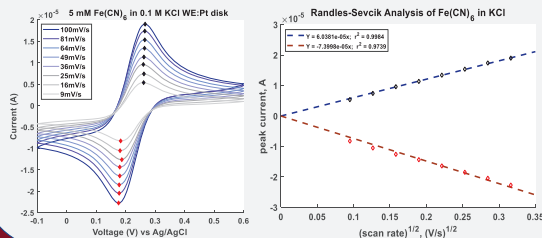
- Measure the **diffusion coefficient** of U through molten salts
 - Test method: $K_4Fe(CN)_6$
- Use diffusion coefficients to inform molecular dynamic simulations of molten salt U mixtures



Aqueous System

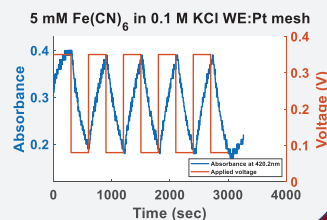
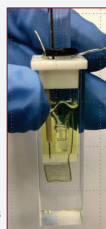
Goal

- Compare standard e-chem with spectroelectrochemistry (SEC), to show that SEC can perform more reliably on less reversible chemical systems
- Proof of concept of the SEC technique



Diffusion Coefficients

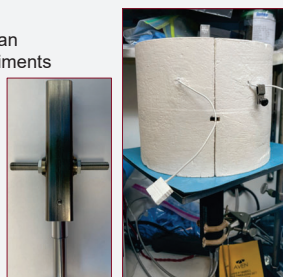
| | Randles-Sevcik (CV) | Cottrell (SEC) |
|------------------|------------------------------|------------------------------|
| Oxidation | 4.686e-06 cm ² /s | 3.905e-06 cm ² /s |
| Reduction | 7.038e-06 cm ² /s | 3.893e-06 cm ² /s |
| Literature Value | 7.6e-06 cm ² /s | |



Molten Salt System

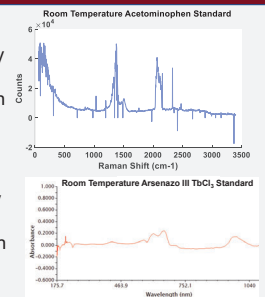
Experimental Design

- Equipped for UV-Vis, Raman and electrochemical experiments
- Furnace reaches temperatures of 700°C
- Experiments performed in an inert atmosphere
- Corrosion resistant design



Limitations

- Raman Spectroscopy
 - 532nm laser
 - 35 min integration time
 - Dark subtracted
- UV-Vis Spectroscopy
 - Quartz reference
 - 10mm path length



Future Work

Diffusion Coefficient

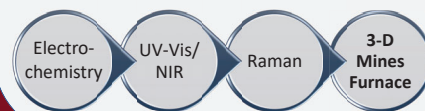
- Determine the diffusion coefficients in reversible, quasi-reversible, and irreversible systems with SEC and compare to standard electrochemical techniques
- Determine the diffusion coefficient for Ln(III) in the LiCl-KCl eutectic melt
 - Compare to literature
- Determine the diffusion coefficient for U(III) and U(IV) in molten salt mixtures
- Use results to inform molecular dynamics

Extended X-ray Absorption Fine Structure (EXAFS)

- Determine the local coordination environment of U in a chloride eutectic frozen salt
- Determine bond distance between bonded salt constituents relative to uranium

Raman Spectroscopy

- Define the bonding characteristics of U with the salt constituents



References

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