INL/MIS-25-84440 Rev:000

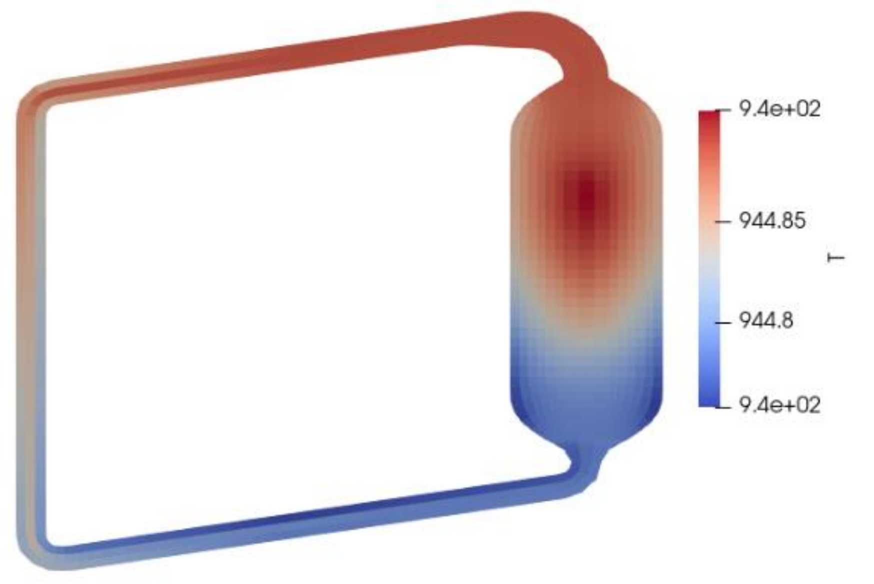
Molten Salt Reactor PROGRAM

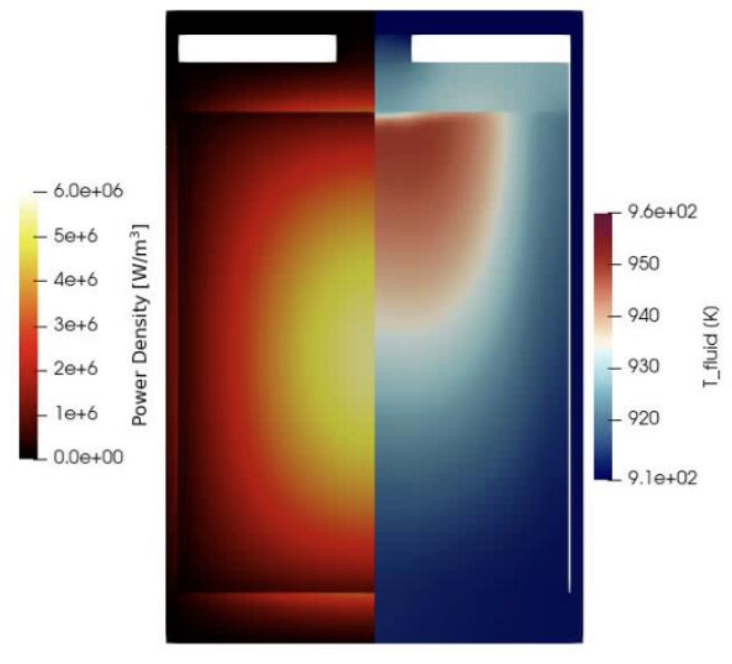
Species Tracking in Molten Salt Reactors: Leaching and Plating

Samuel Walker, Mauricio Tano

Argonne NATIONAL LABORATORY · INL Idaho National Laboratory · Los Alamos NATIONAL LABORATORY · OAK RIDGE National Laboratory · Pacific Northwest NATIONAL LABORATORY · Sandia National Laboratories · U.S. DEPARTMENT of ENERGY Office of Nuclear Energy

Content





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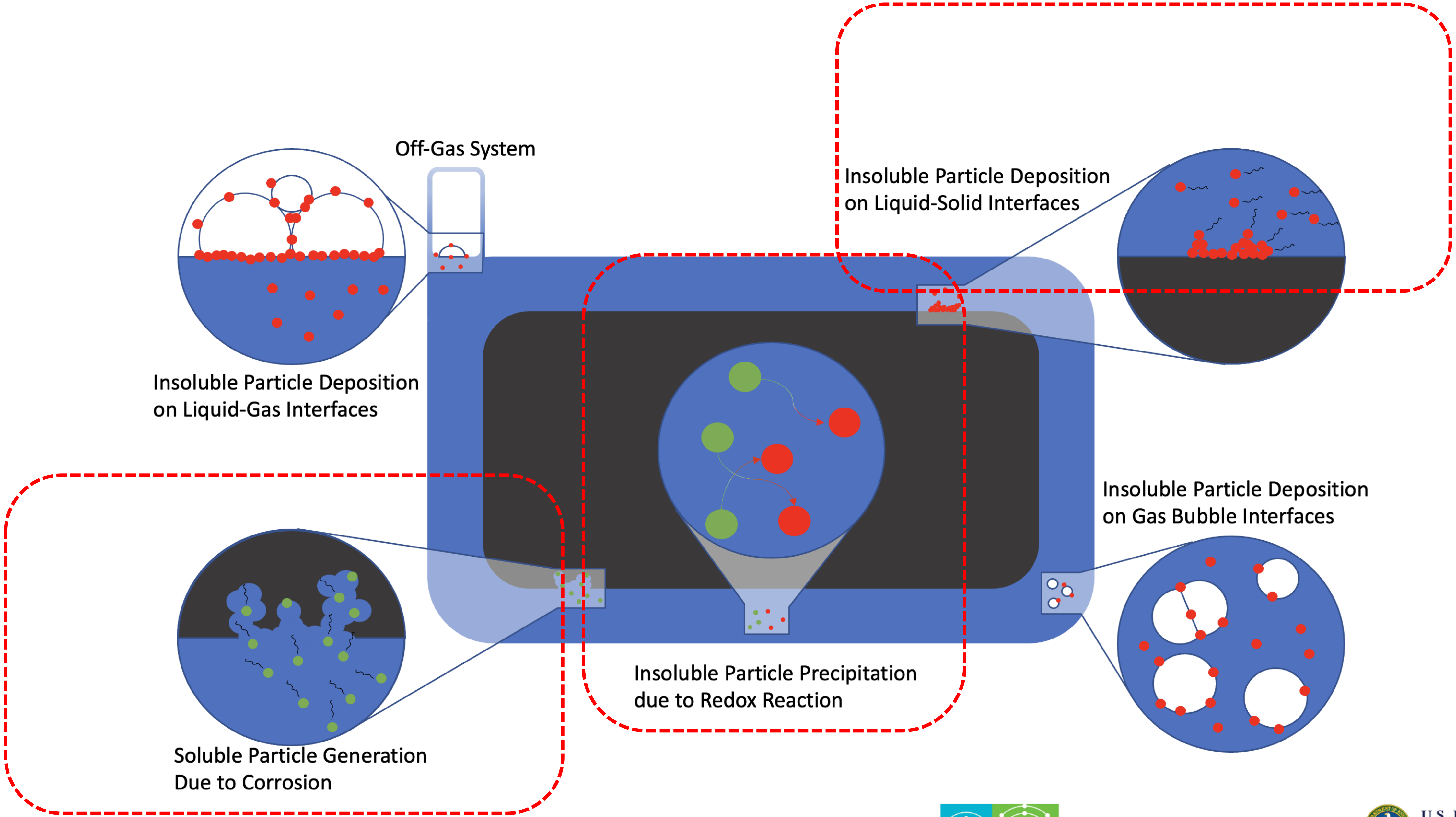
Motivation for Species Tracking in MSRs

Species Tacking Behavior in MSRs



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Why do we need to track species?

- Importance of Species Tracking
  - **Containment**: Where do the radionuclides go? What is the reactor source term?
  - **Heat removal**: Where do isotopes plate out? How do we cool the reactor?
  - **Reactivity**: Where do the neutron precursors go? What is the reactor β_{eff}?
  - **Corrosion**: How do fission products interact with the wall? How long will a barrier last?
  - **Safeguards**: Where do the fissile isotopes go? How do we monitor where they are?

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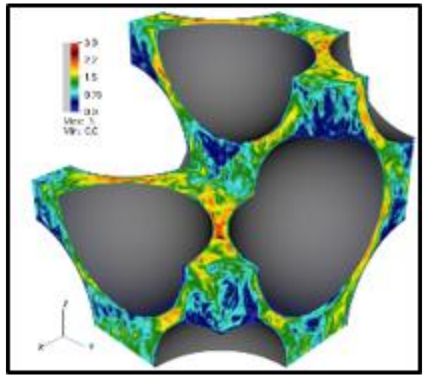
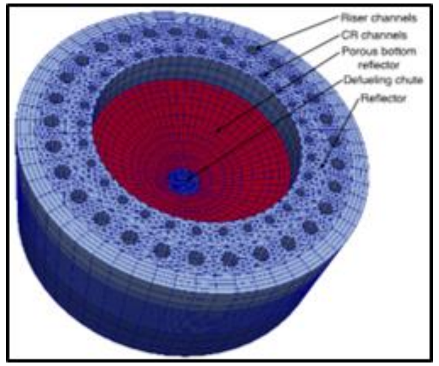
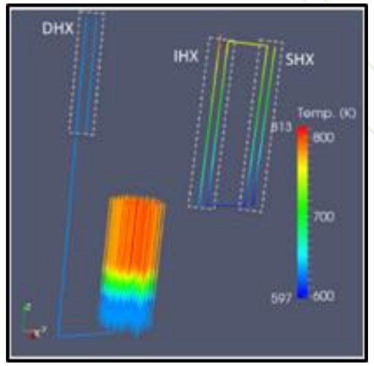
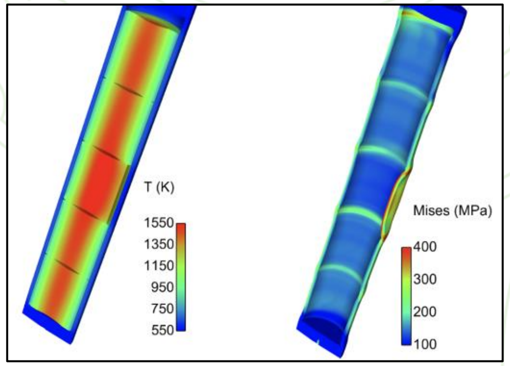
Development of Computational Framework for Species Tracking

Main NEAMS tools used for modeling and simulation of MSRs









Nek-5000 nekRS + **Griffin** + **Pronghorn** + SAM + BISON/ Thermochimica

Fuel Performance/ Thermochemistry

High Fidelity- Physics Resolved

Component Scale

Plant Scale

Operation Analyses

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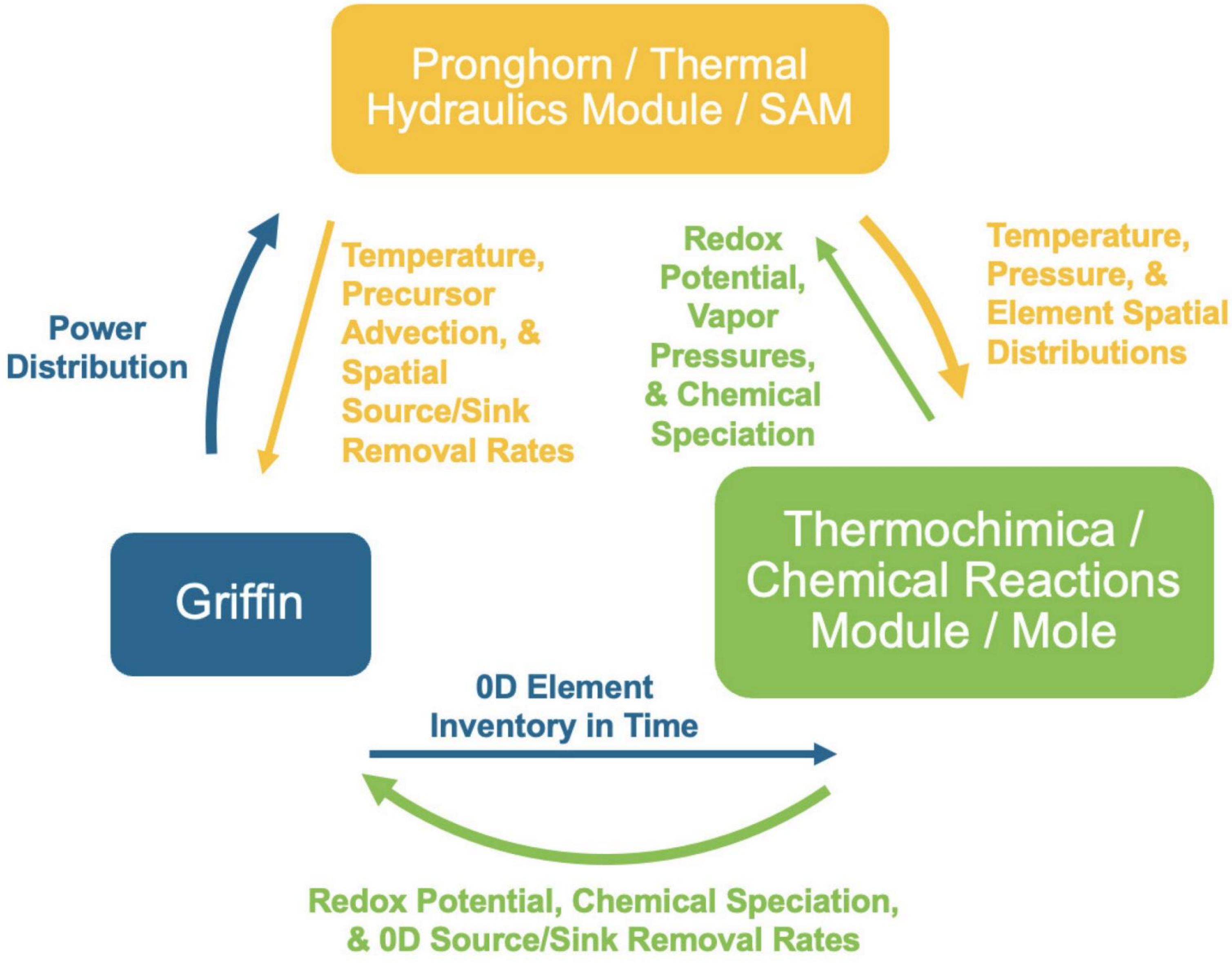
Coupling Framework for Species Tracking

- The coupling framework for species tracking in MSRs involves:

1. **Griffin**: neutronics + depletion
2. **Pronghorn**: thermal-hydraulics + species transport
3. **Thermochimica**: chemical equilibrium calculations



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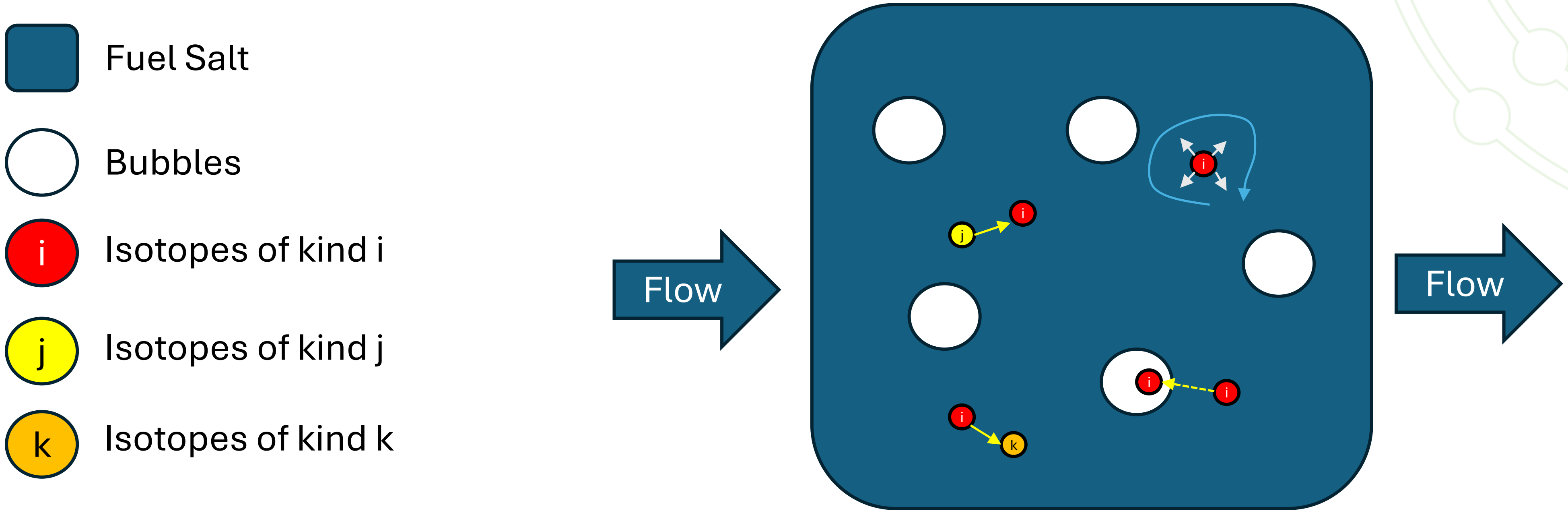
Review of FY24 Activities

Coupled Transport of Soluble and Gaseous Species

FY24 work: Coupled Transport of Soluble + Gaseous Species



$$\underbrace{\frac{\partial c_i^x}{\partial t}}_{\text{Time derivative}} + \underbrace{\nabla \cdot \left(\frac{\boldsymbol{u}}{\gamma} c_i^x\right)}_{\text{Convection in Porous Media}} - \underbrace{\nabla \cdot \left[\left(D_{C_i} + \frac{\nu_t}{Sc_t}\right) \nabla c_i^x\right]}_{\text{Molecular and Turbulent Diffusion}} = \underbrace{y_i f}_{\text{Fission Yield Source}} - \underbrace{\lambda_i^{eff} c_i^x}_{\text{Effective Decay Sink}} + \underbrace{\lambda_{j \to i}^{eff} c_j^x}_{\text{Effective Transmutation Rate}} - \underbrace{h^{xy}(c_i^x - c_i^y)}_{\text{Exchange of phases between states y and x}}$$

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MSRE – Reactivity Insertion Transient

- 100 pcms are injected in the reactor core at the initial time
- During a reactor transient, the distribution of void plays a fundamental role in power attenuation
- Additionally, the distribution of void is important in the reactor setpoint after the transient

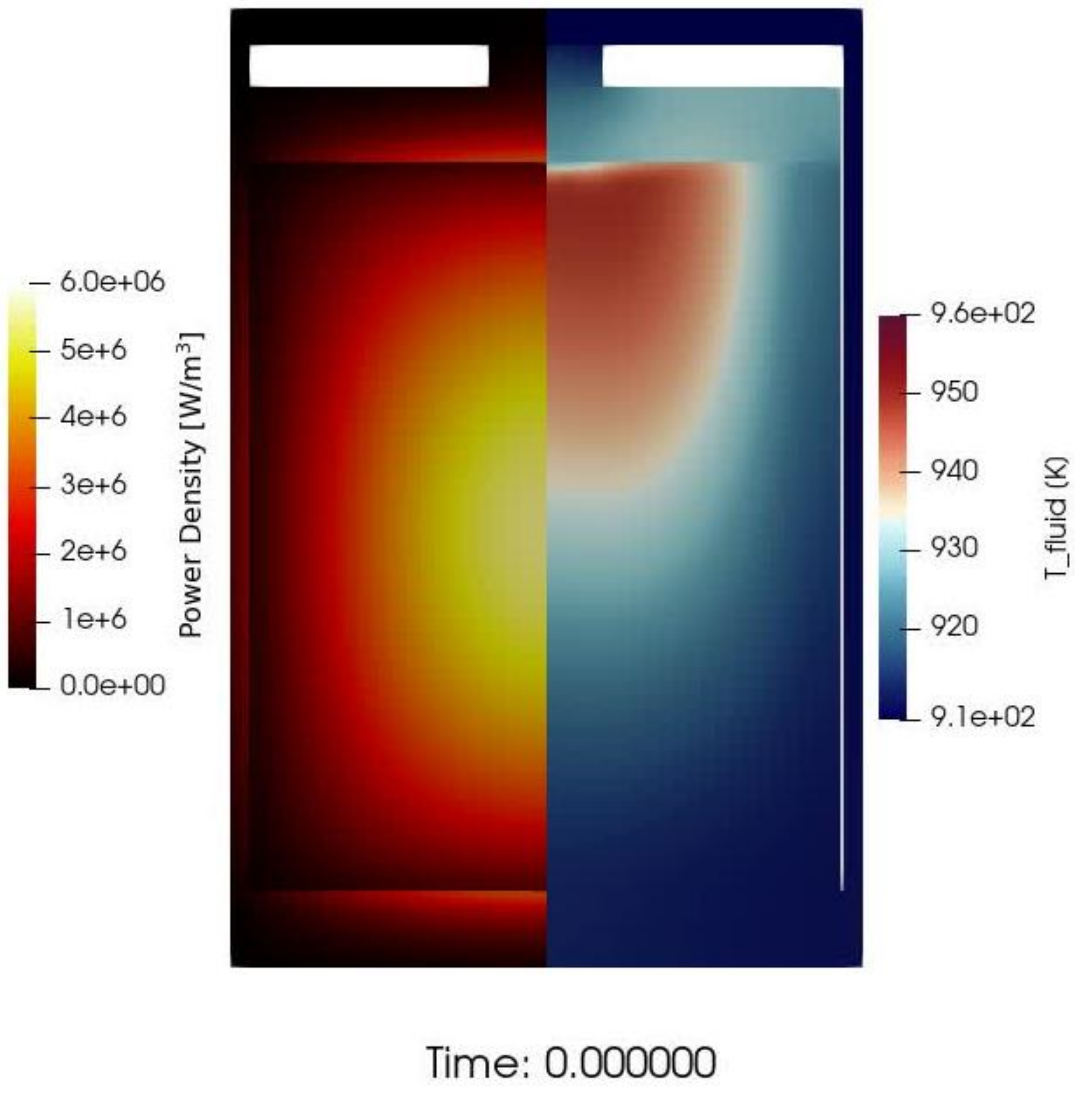

Figure: power density (left) and salt temperature (right)

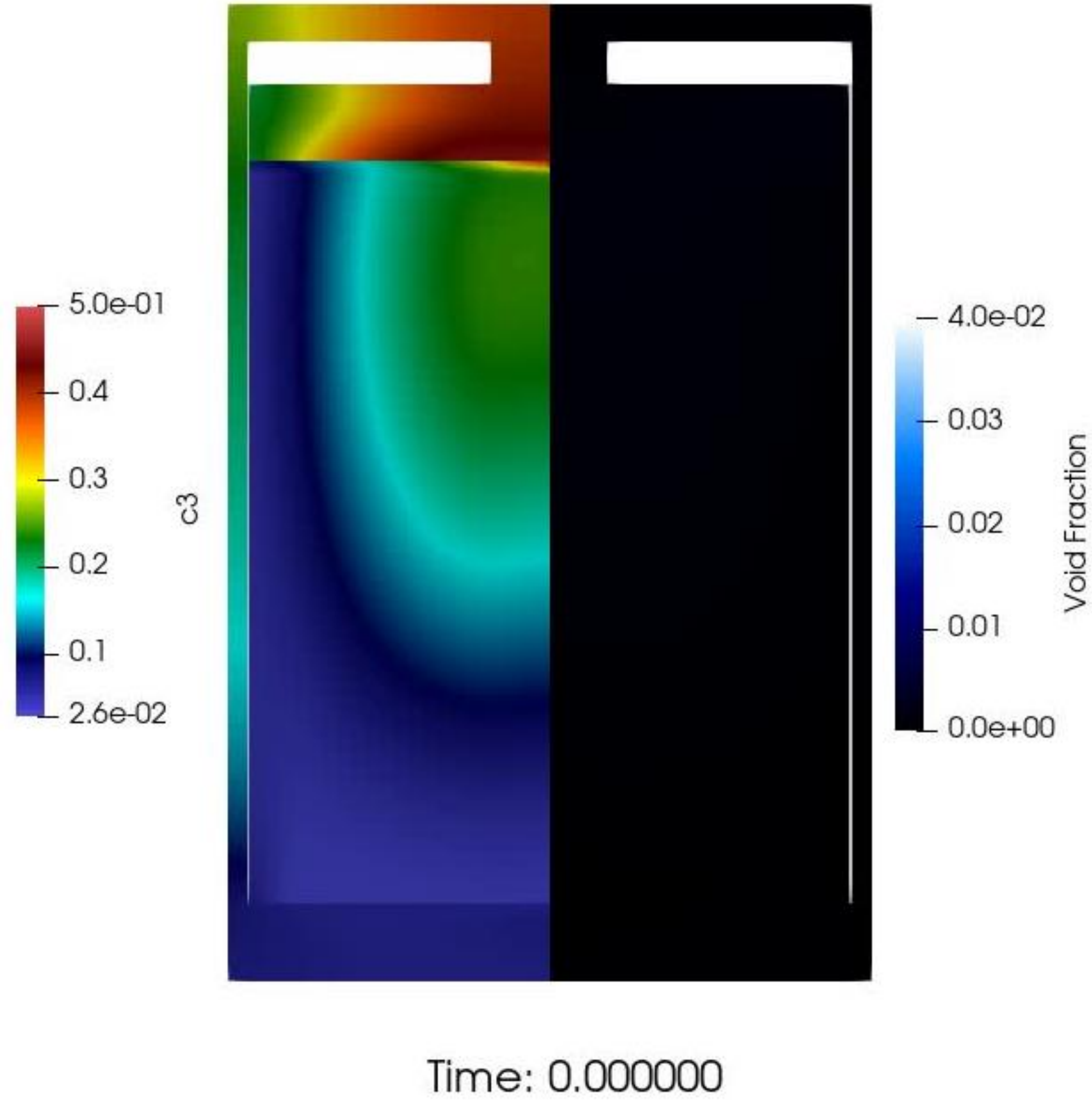

Figure: delay neutron precursors of group 3 (left) and void fraction (right)

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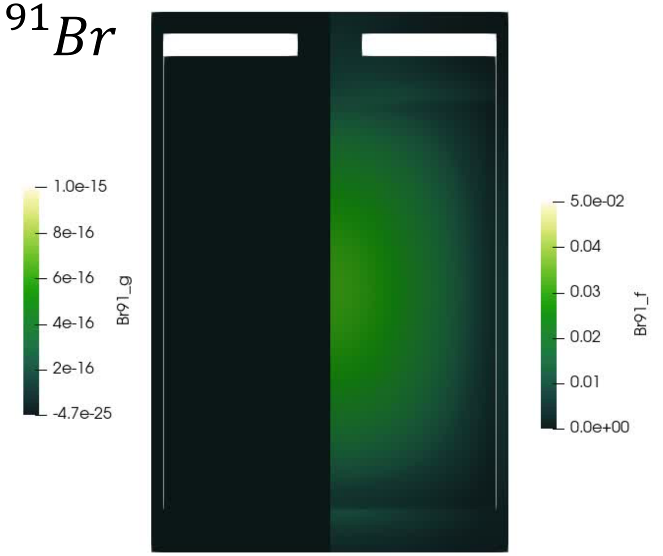
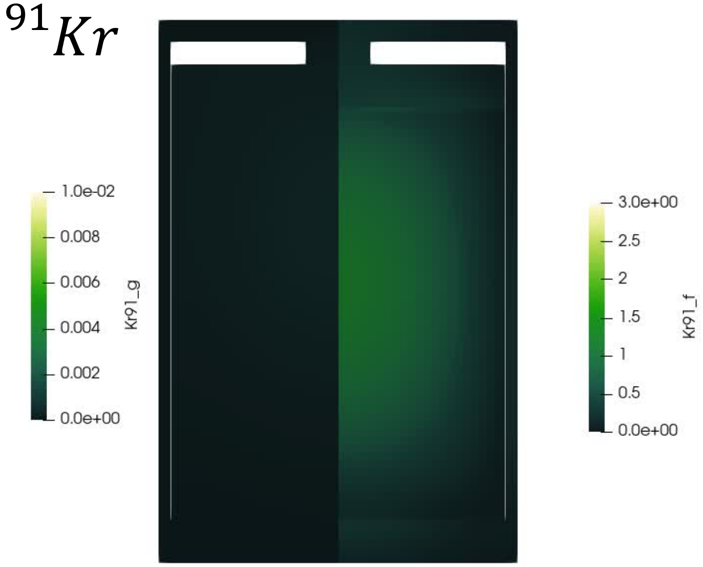
Evolution of gaseous species during reactivity insertion transient

^{91}Br



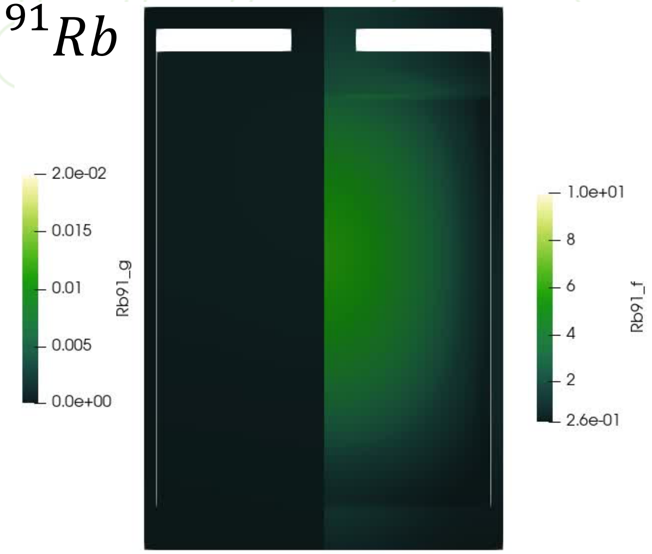
Time: 0.000000

^{91}Kr



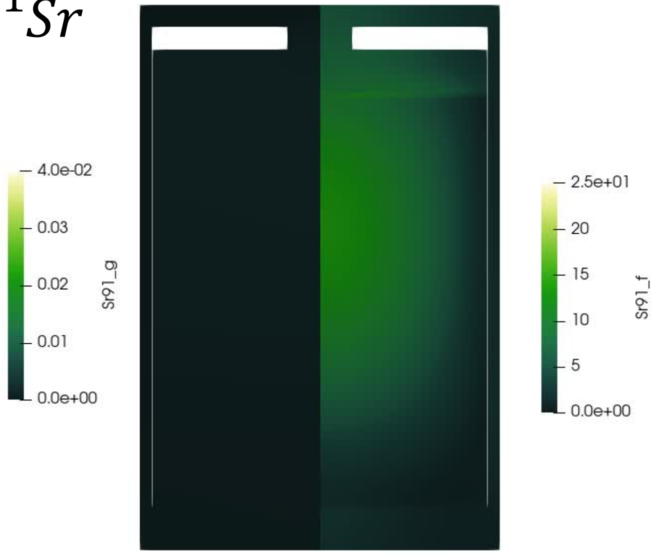
Time: 0.000000

^{91}Rb



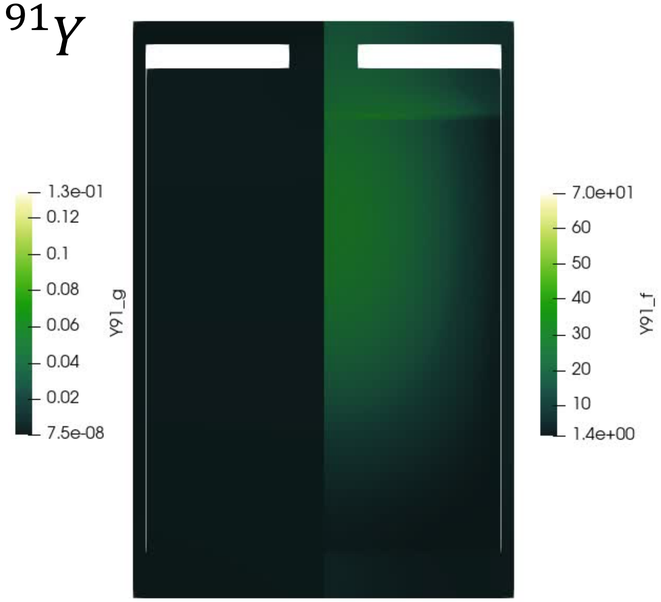
Time: 0.000000

^{91}Sr



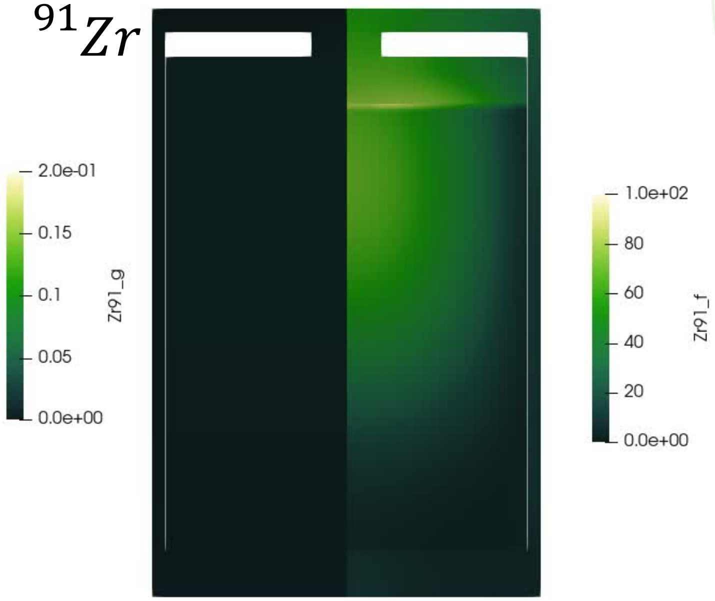
Time: 0.000000

^{91}Y



Time: 0.000000

^{91}Zr



Time: 0.000000

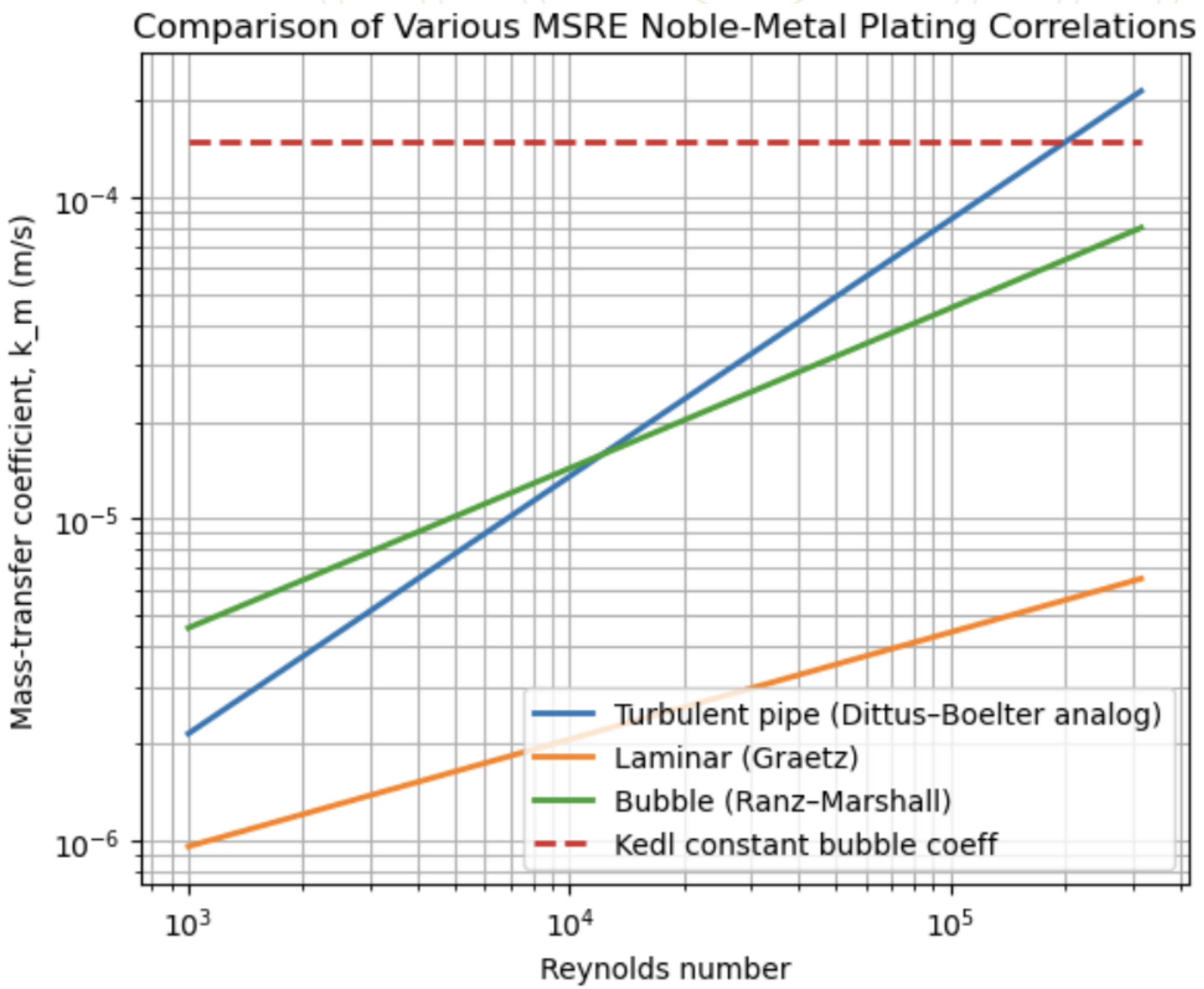
FY25Activities

Coupled Transport of Soluble and Insoluble Metallic Species

Motivation: Dispersion in Mass Transfer Correlations

- A lot of dispersion is observed in the mass transfer coefficients used for MSRE
- A more refined mechanistic approach is needed for mass transfer

Correlation	Applicability	$k_m\ [ms^{-1}]$ range
Dittus-Boelter Analog	Turbulent pipes	[2.14e-06, 2.14e-04]
Graetz	Laminar / Laminarization	[9.55e-07, 6.50e-06]
Ranz–Marshall	Bubbly Flow	[4.57e-06, 8.04e-05]
Kedl's constant	Bubble Film	1.50e-04



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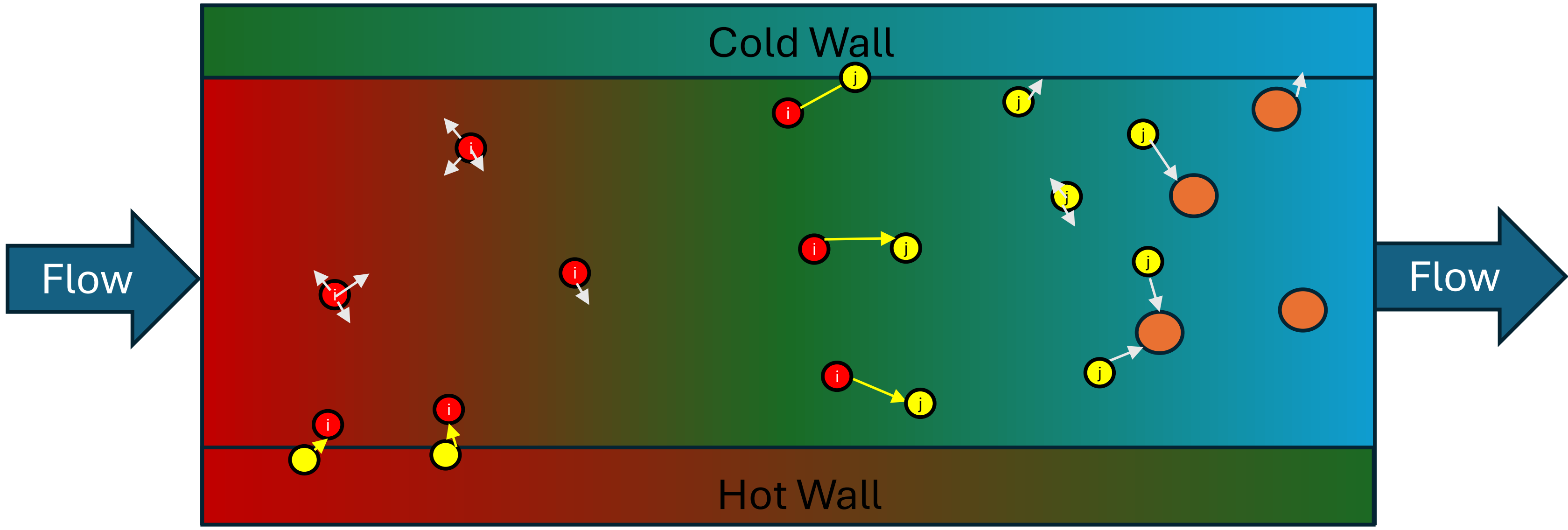
FY25 work: Coupled Transport of Soluble + Insoluble Metallic Particulate Species

- Fuel Salt
- j — Summed nuclides (elements) of insoluble behavior (i.e., Fe(s))
- i — Summed nuclides (elements) of soluble behavior (i.e. Fe^{2+})
- Colloids



$$\underbrace{\frac{\partial c_i^x}{\partial t}}_{\text{Time derivative}} + \underbrace{\nabla \cdot \left(\frac{\boldsymbol{u}}{\gamma} c_i^x\right)}_{\text{Convection in Porous Media}} - \underbrace{\nabla \cdot \left[\left(D_{C_i} + \frac{\nu_t}{Sc_t}\right) \nabla c_i^x\right]}_{\text{Molecular and Turbulent Diffusion}} = \underbrace{y_i f}_{\text{Fission Yield Source}} - \underbrace{\lambda_i^{eff} c_i^x}_{\text{Effective Decay Sink}} + \underbrace{\lambda_{j\to i}^{eff} c_j^x}_{\text{Effective Transmutation Rate}} - \underbrace{h^{xy}(c_i^x - c_i^y)}_{\text{Exchange of phases between states y and x}}$$

NOTE: c_i^y term comes directly from Thermochimica equilibrium calculation to determine phase change (melting/precipitation) due to redox and temperature changes.

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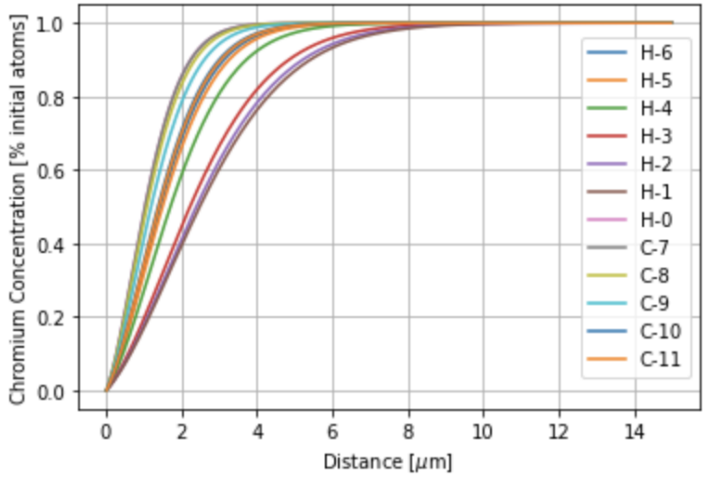
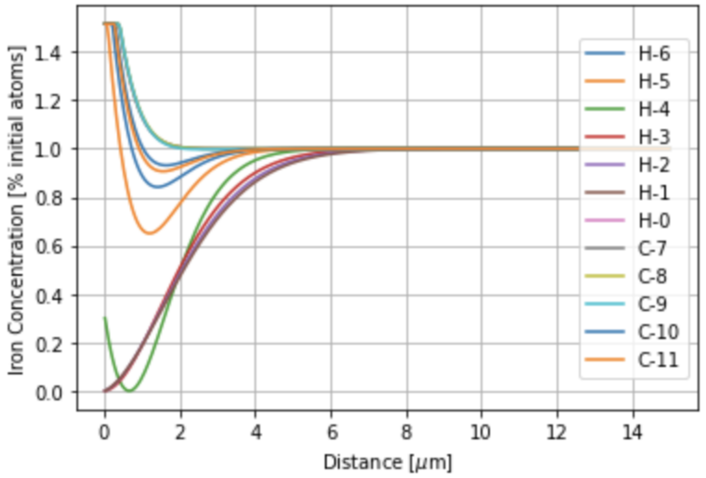
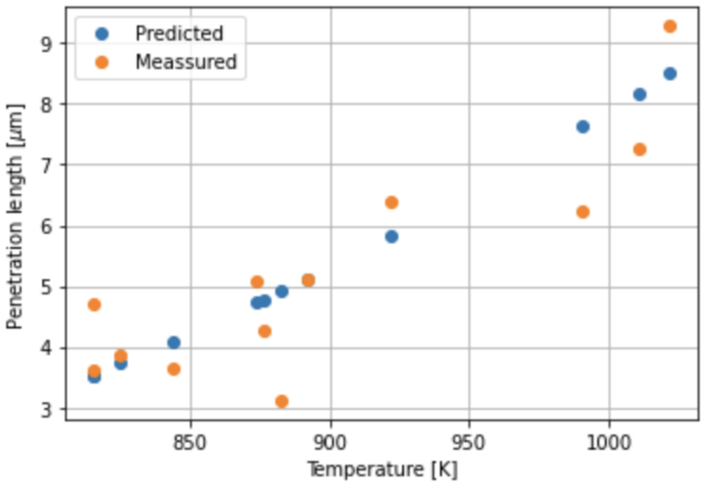
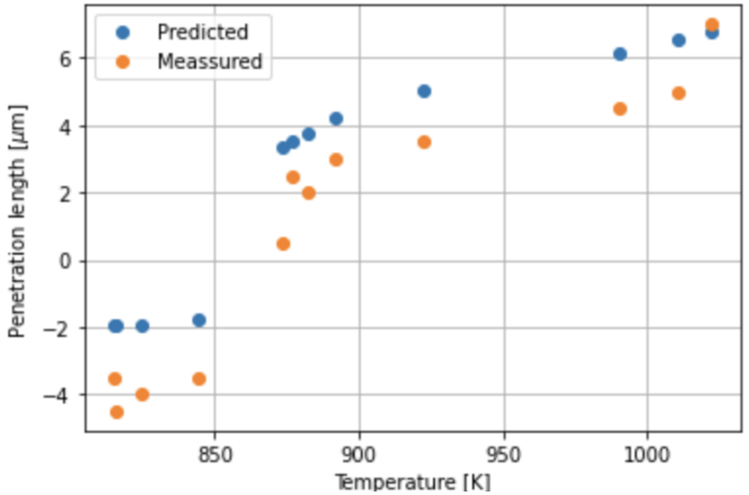
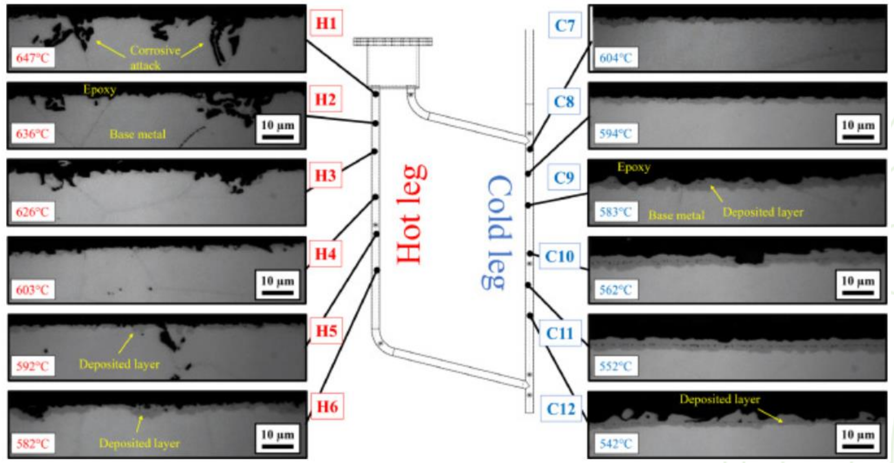
Verification against existing Poisson-Nernst-Planck Model and Molten Salt Loop Experiment









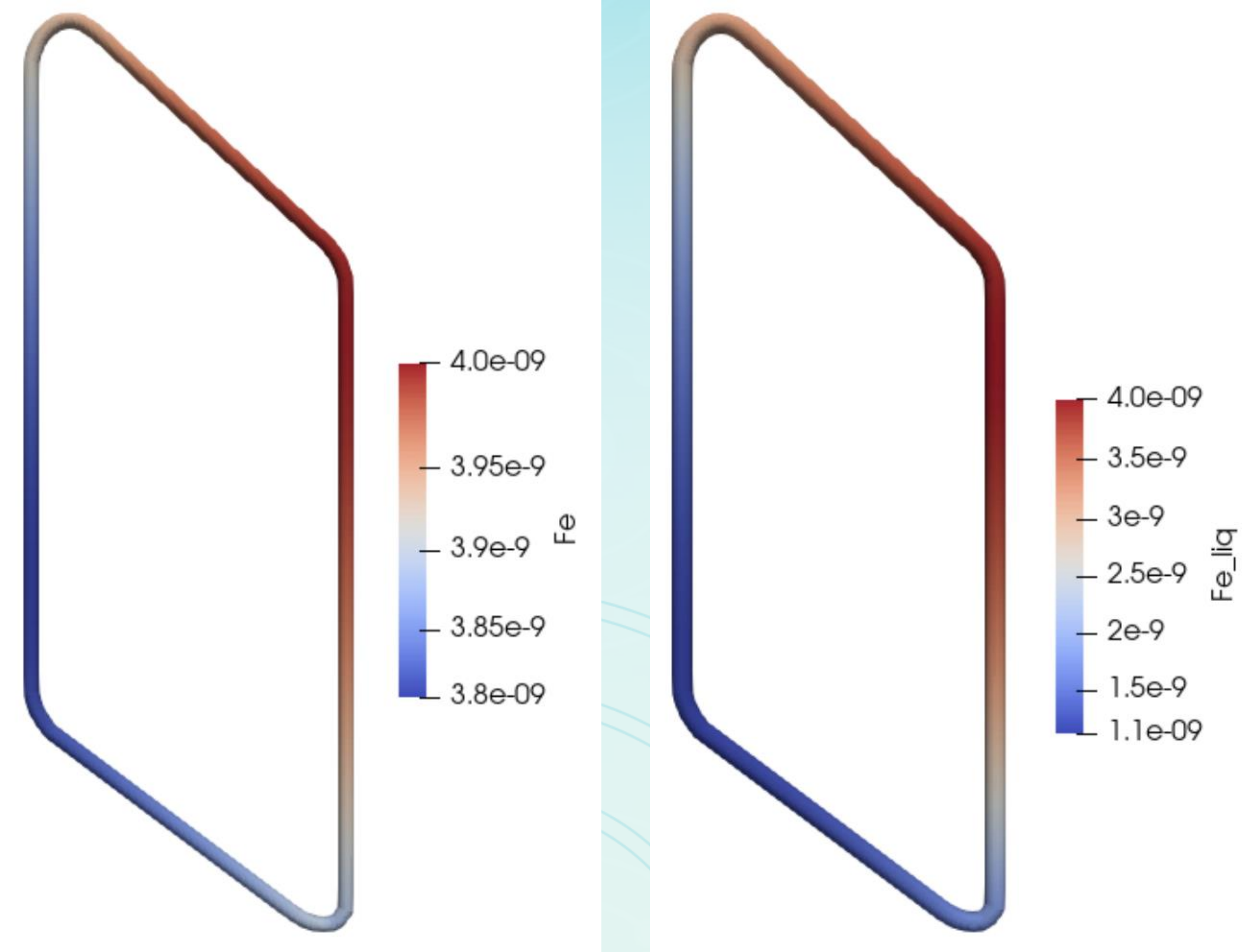


Raiman, S. S., Kurley, J. M., Sulejmanovic, D., Willoughby, A., Nelson, S., Mao, K., ... & Pint, B. A. (2022). Corrosion of 316H stainless steel in flowing FLiNaK salt. *Journal of Nuclear Materials*, *561*, 153551.

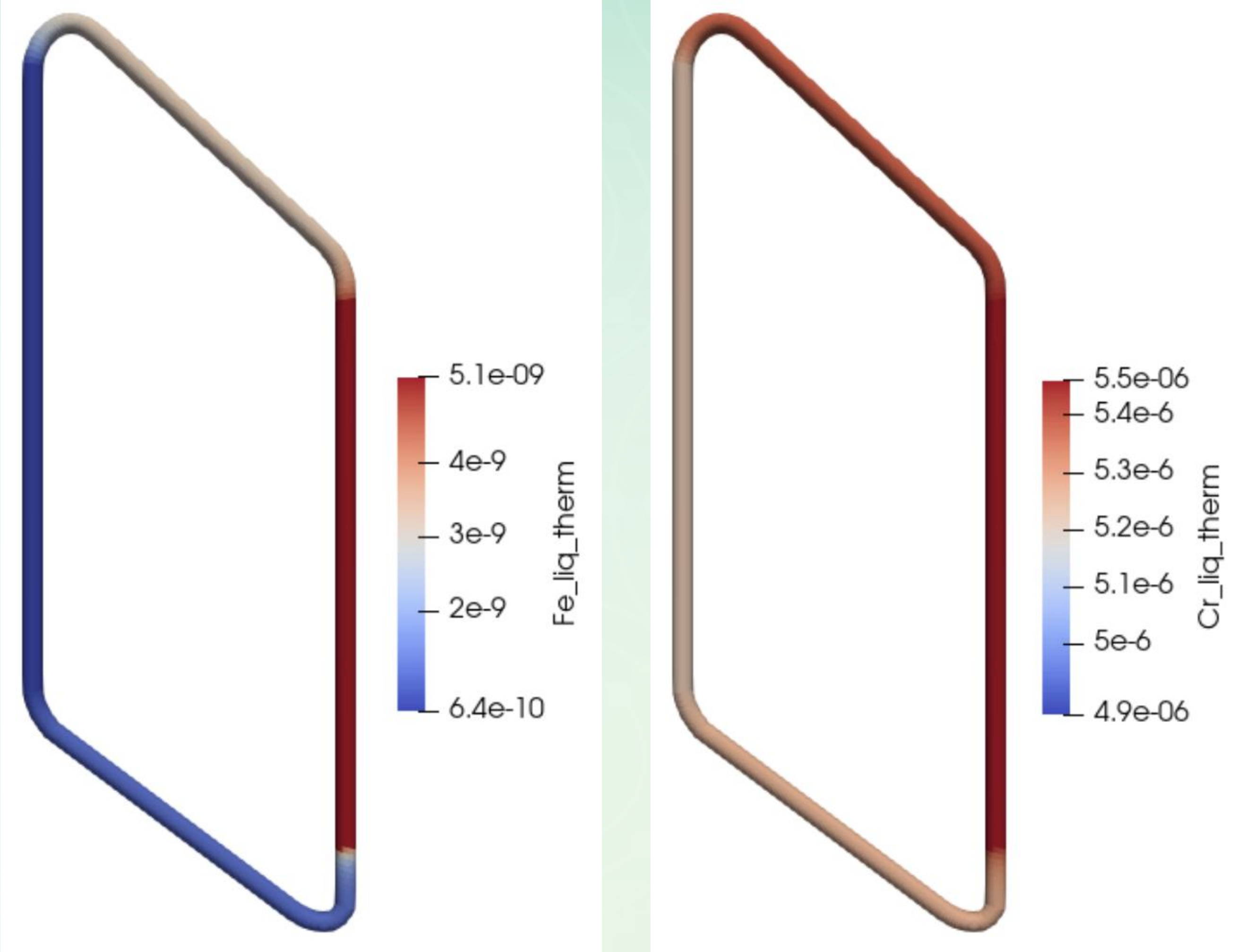
Predicted Concentration [wppm]	Cr	Fe
Experiment	255 ± 28	60 ± 8
Mechanistic model	235	75
PNP model	251	72
Thermochimica model	861	69

Remaining amounts in the salt

Pronghorn + Thermochimica Model Results

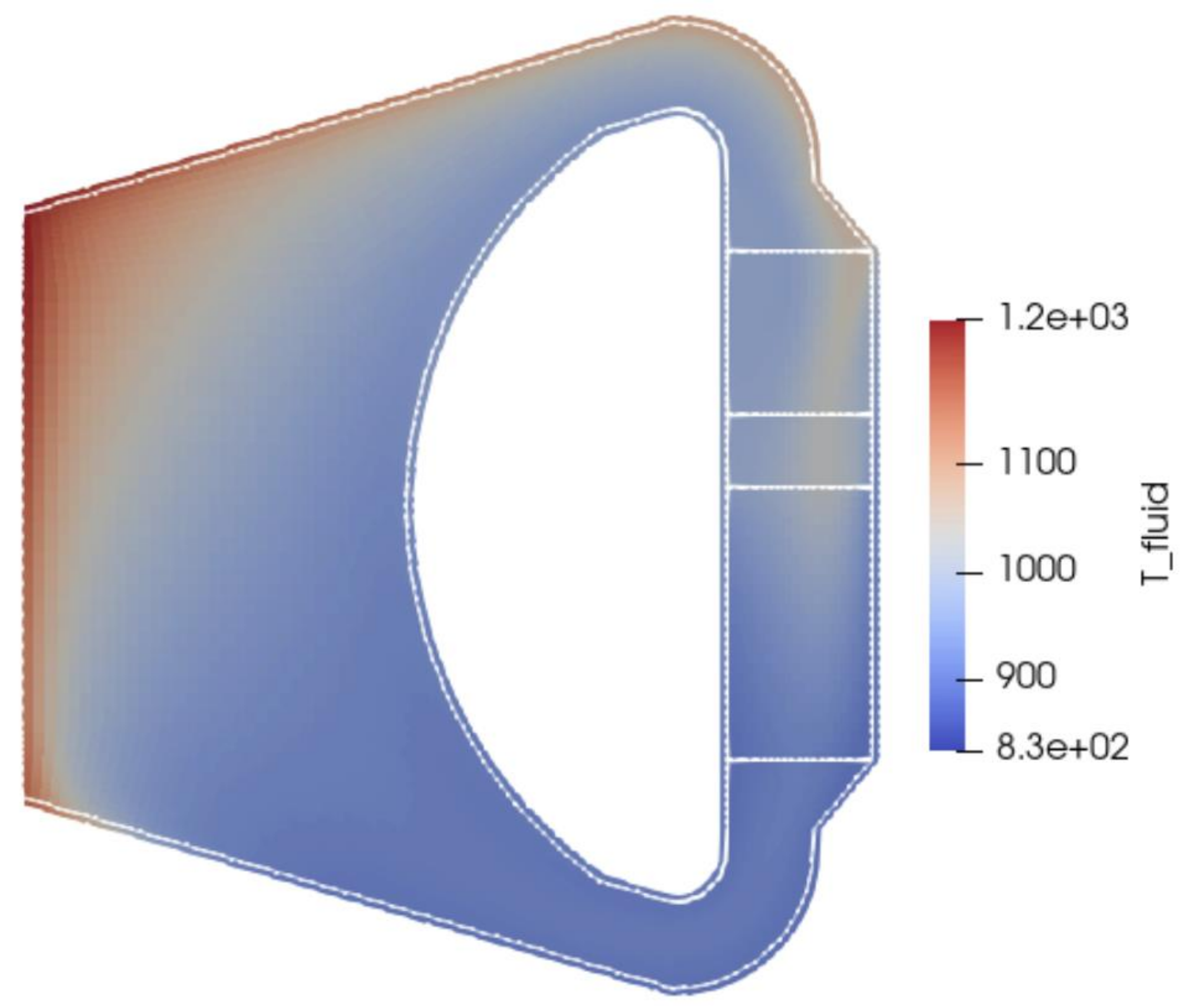


Equilibrium Concentration of FeF_2 in FLiNaK loop: (left) PNP Results; (right) Pronghorn + Thermochimica Results

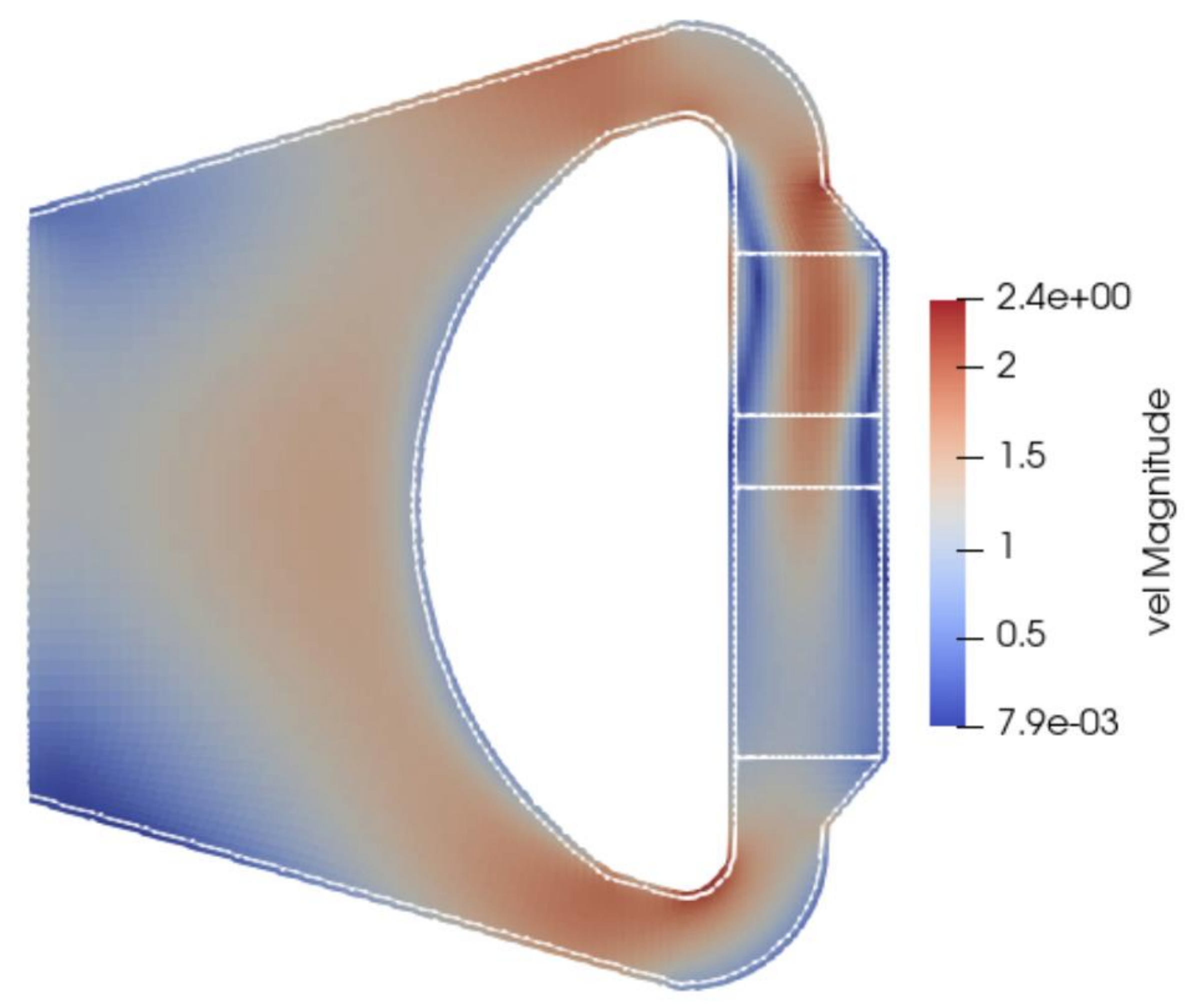


Equilibrium Source Terms from Thermochimica: (Left) FeF_2; (right) Sum of CrF_2 and CrF_3

Application to Molten Salt Fast Reactor (MSFR)



Temperature Profile in MSFR at Steady State



Velocity Profile in MSFR at Steady State

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Demonstration Problem

Tracking Hot-Leg Corrosion – Cold-Leg Deposition with Colloid precipitation via Pronghorn + Thermochimica model

Liquid Cr Equation

$$\frac{\partial Cr_l^x}{\partial t} + \nabla \cdot \left(\frac{\boldsymbol{u}}{\gamma} Cr_l^x\right) - \nabla \cdot \left[\left(D_{Cr_l^x} + \frac{\nu_t}{Sc_t}\right) \nabla Cr_l^x\right] = \underbrace{-h^{l-s}(Cr_l^x - Cr_l^*)}_{\text{Exchange of phases between liquid and solid wall interface}} + \underbrace{h^{l-c}(Cr_c^x - Cr_s^*)}_{\text{Exchange of phases between liquid and colloids in bulk}}$$

Metallic Cr Colloids Equation

$$\frac{\partial Cr_c^x}{\partial t} + \nabla \cdot \left(\frac{\boldsymbol{u}}{\gamma} Cr_c^x\right) - \nabla \cdot \left[\left(D_{Cr_c^x} + \frac{\nu_t}{Sc_t}\right) \nabla Cr_c^x\right] = \underbrace{-h^{l-c}(Cr_c^x - Cr_s^*))}_{\text{Exchange of phases between liquid and colloids in bulk}} - \underbrace{h^{c-s}(Cr_c^x - 0)}_{\text{Mass Transfer of colloids adhering to wall interface}}$$

Solid Cr Wall Equation

$$\frac{\partial Cr_s^x}{\partial t} = \underbrace{+h^{l-s}(Cr_l^x - Cr_l^*)}_{\text{Exchange of phases between liquid and solid wall interface}} + \underbrace{h^{c-s}(Cr_c^x - 0)}_{\text{Mass Transfer of colloids adhering to wall interface}}$$

NOTE: Cr_l^* and Cr_s^* terms comes directly from Thermochimica equilibrium calculation to determine phase change (melting/precipitation) due to redox and temperature changes.

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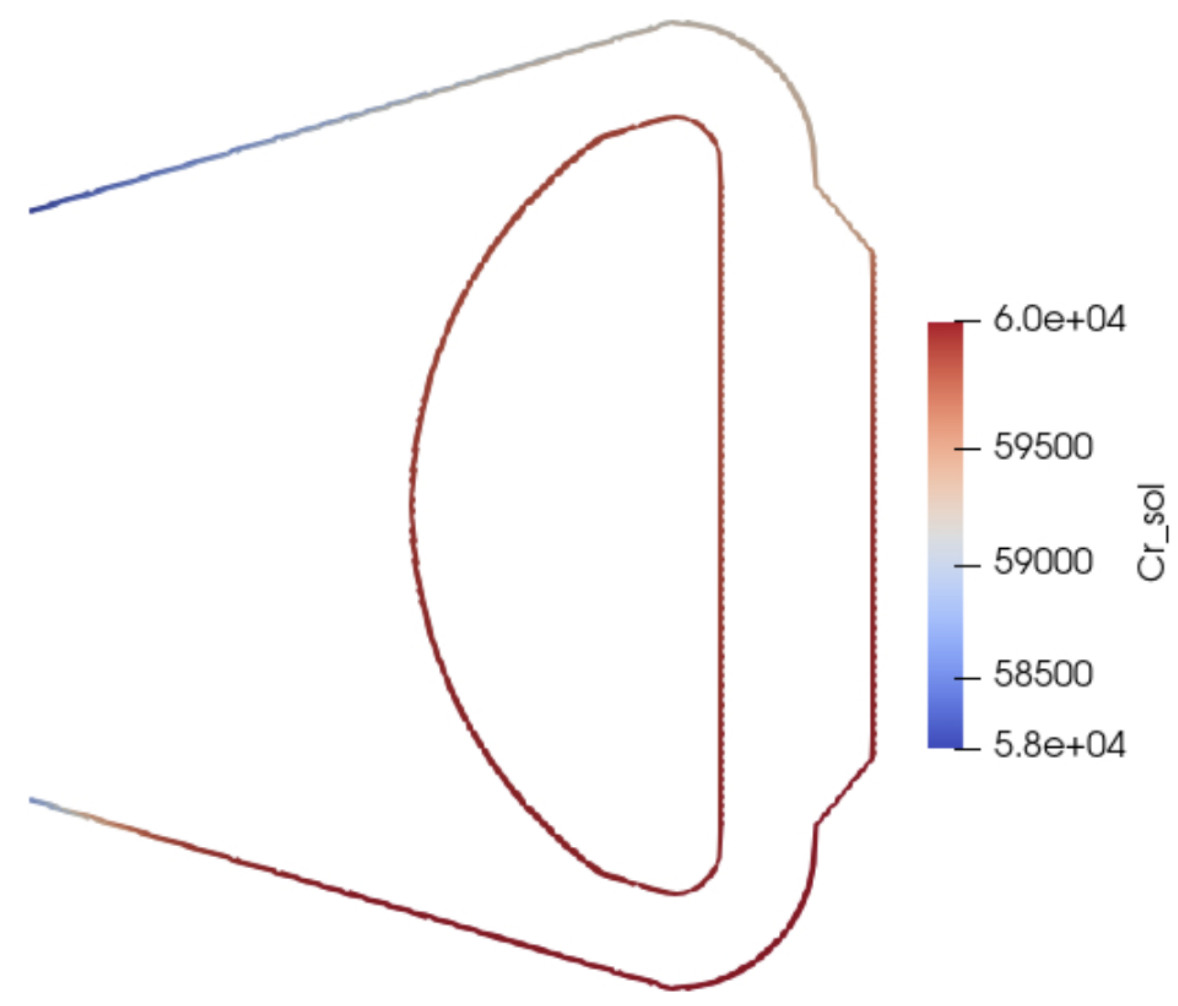
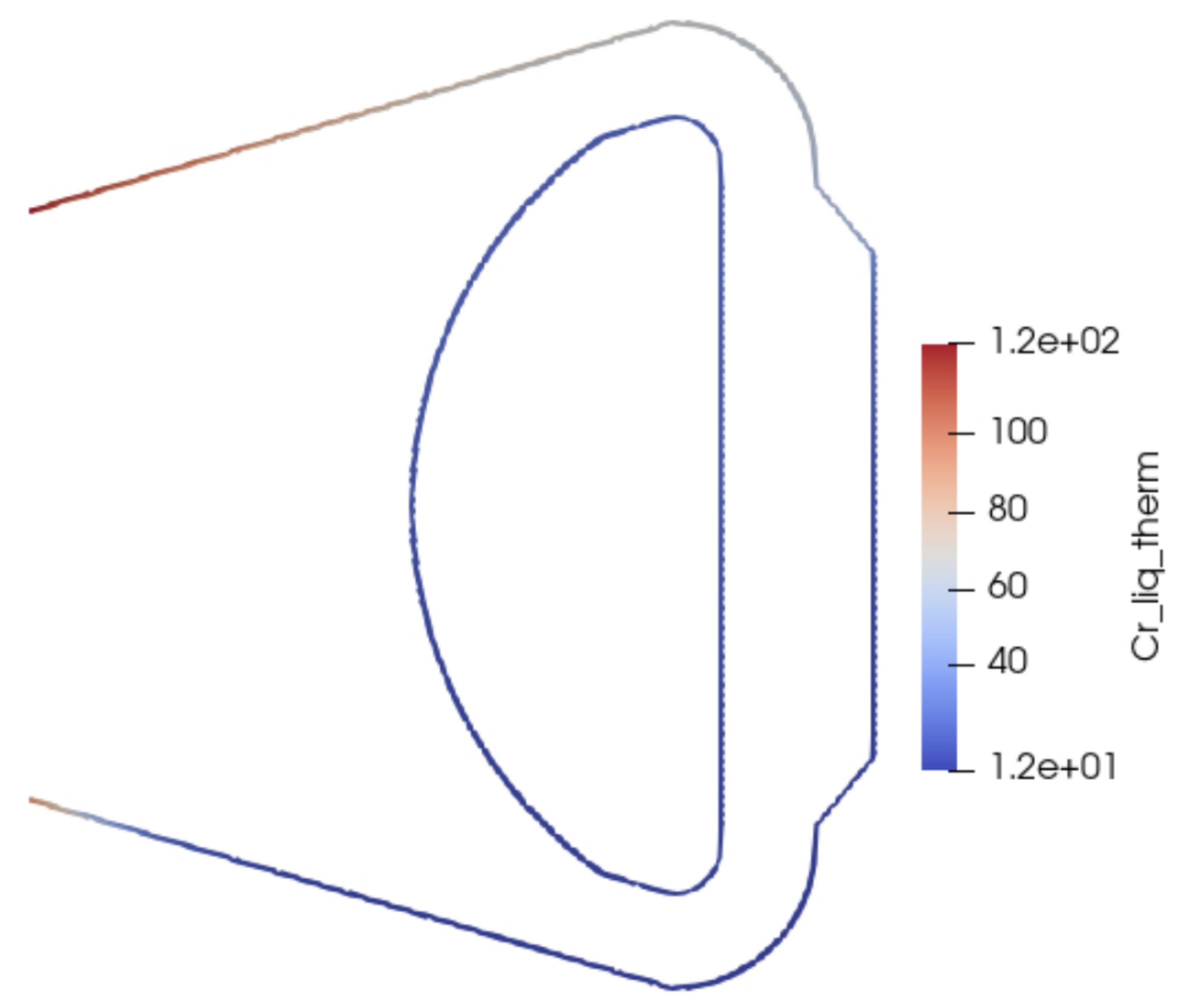
MSFR: Chromium Leaching



Significant chromium loss at top of hot reactor core and minimal deposition in cold plenum entrance after heat exchange



Corresponding Cr_l^* source term from Thermochimica driving chromium corrosion

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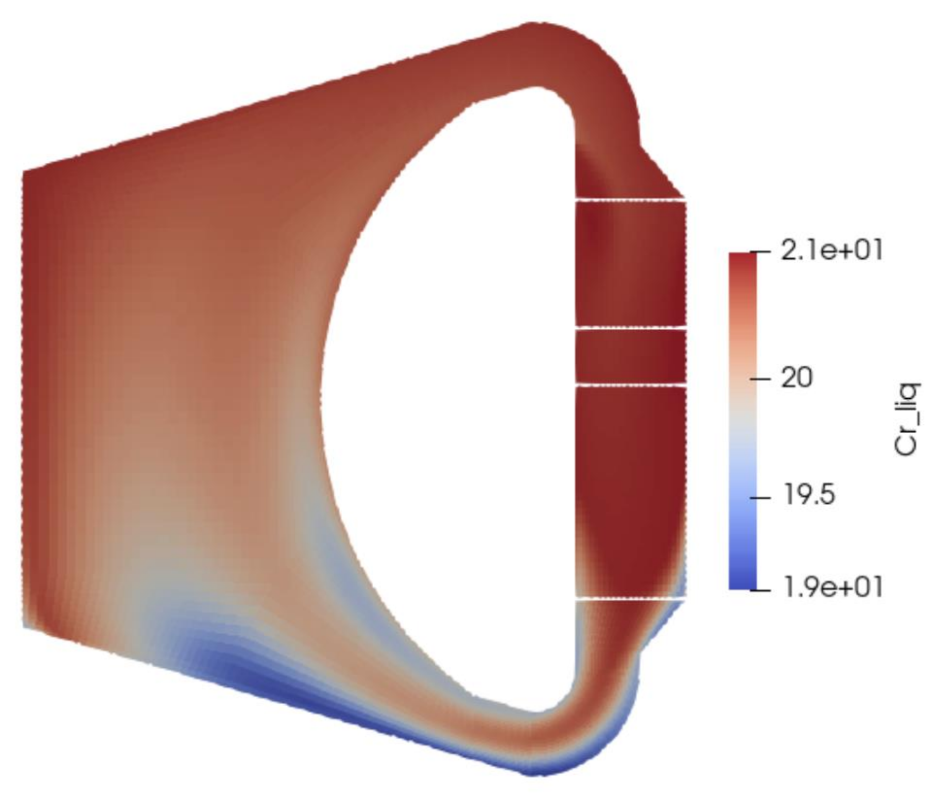
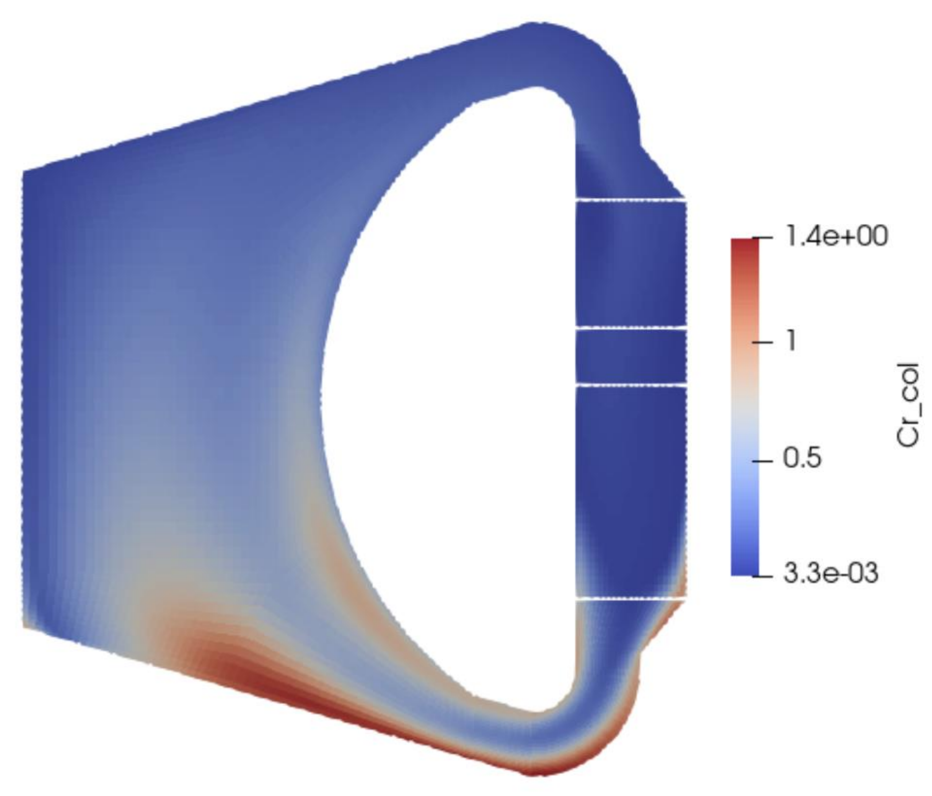
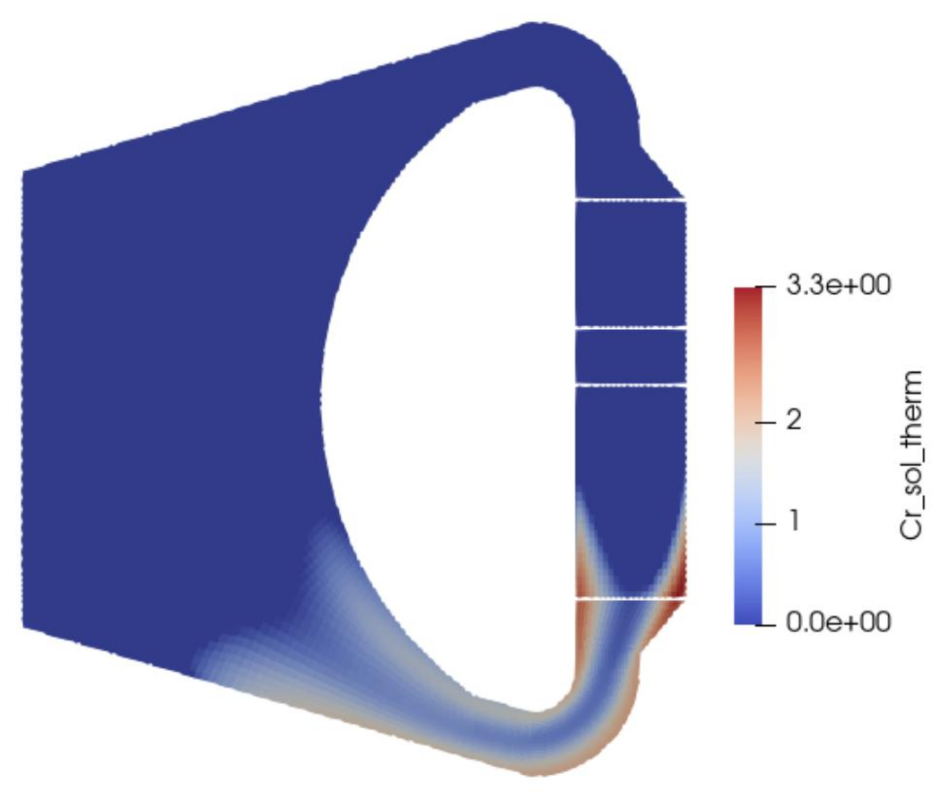
MSFR: Chromium Leaching – impact of temperature



Equilibrium liquid chromium concentration in MSFR



Corresponding equilibrium colloid chromium concentration in MSFR



Corresponding Cr_s^* source term from Thermochimica driving metallic chromium precipitation in cold leg of MSFR

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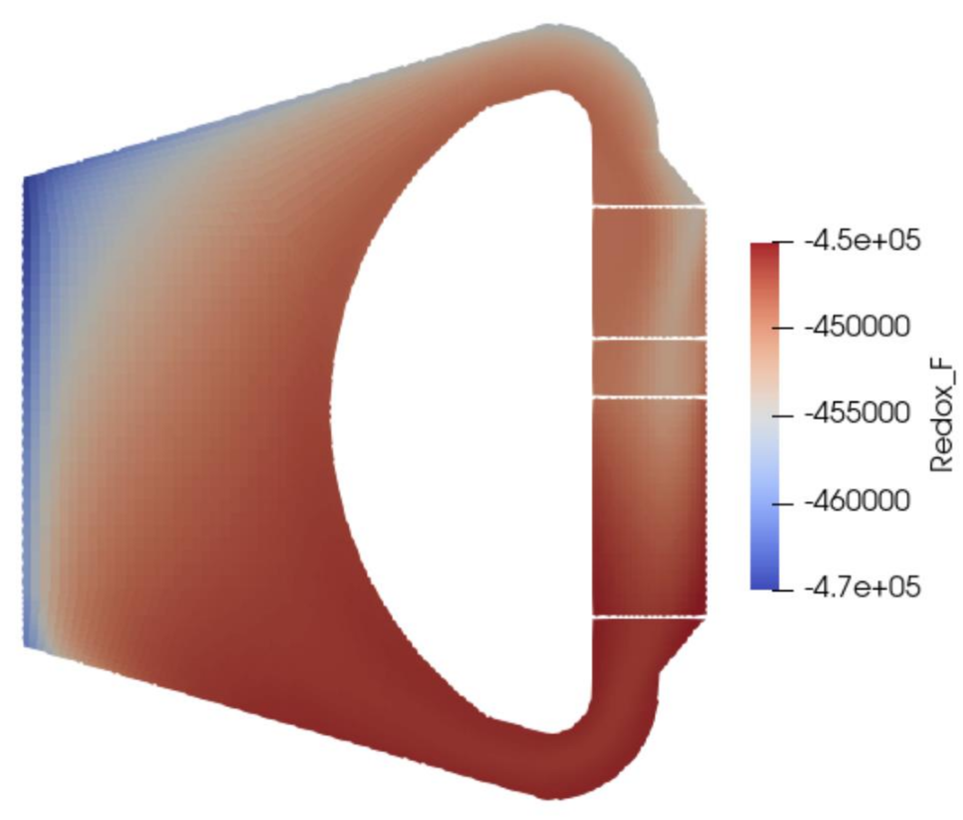
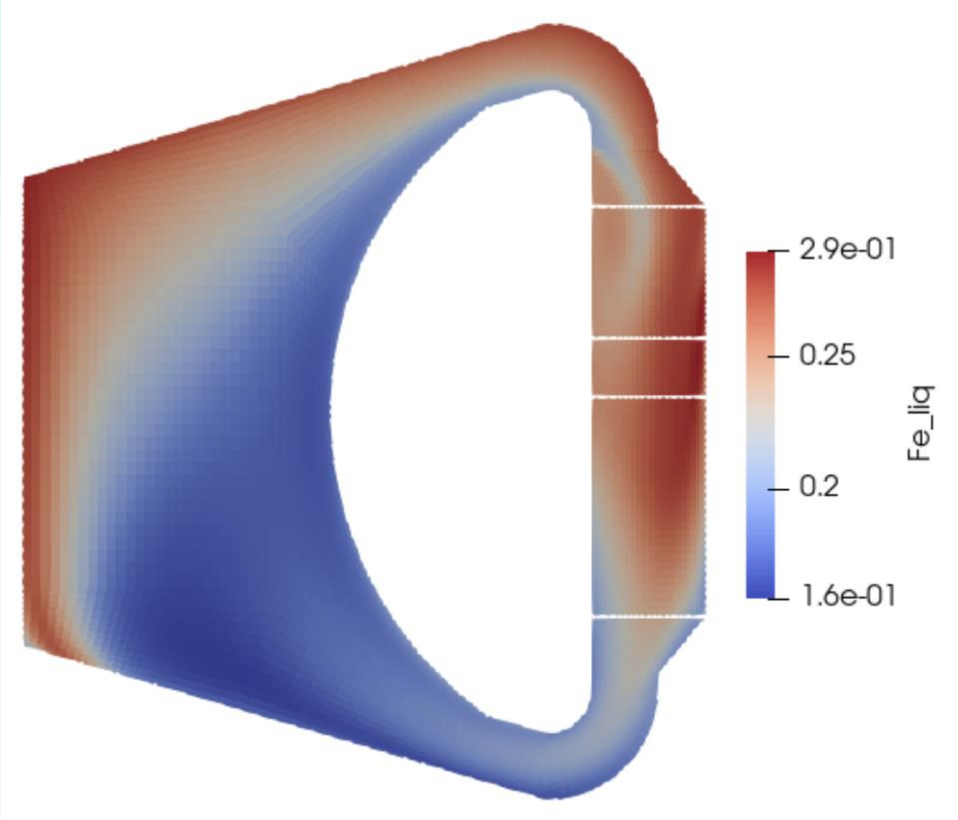
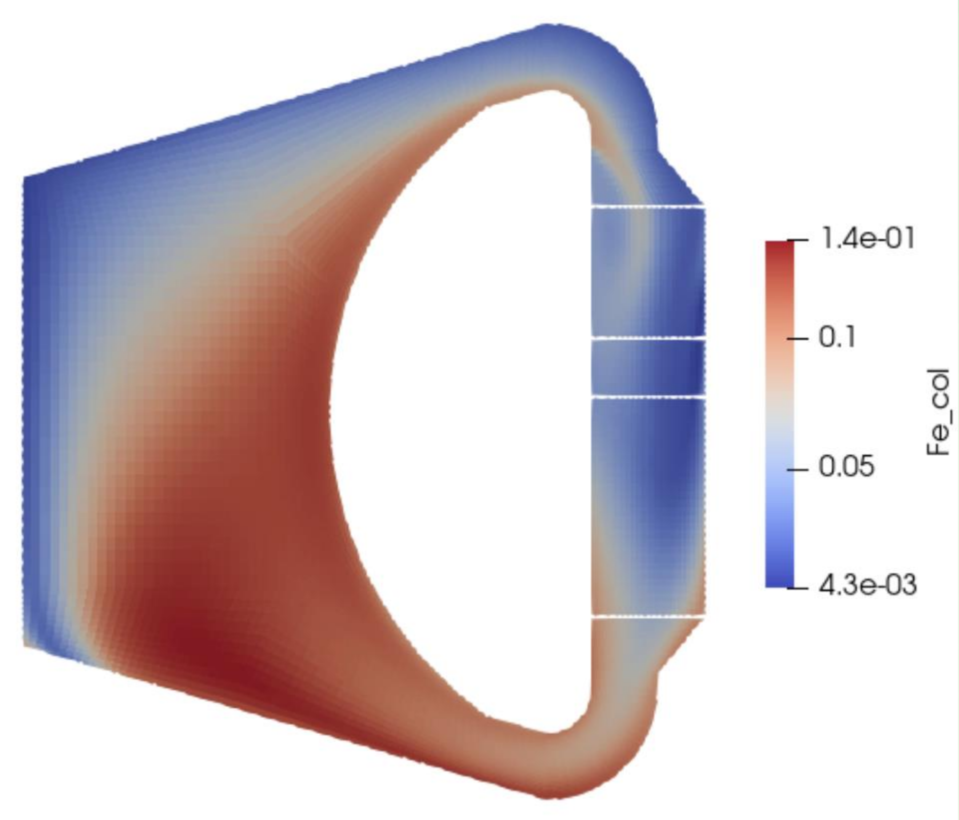
MSFR: Iron Leaching – impact of redox potential



Equilibrium Redox Potential
(F^- Chemical Activity)



Equilibrium liquid iron
concentration in MSFR



Corresponding metallic iron colloid
concentration in MSFR

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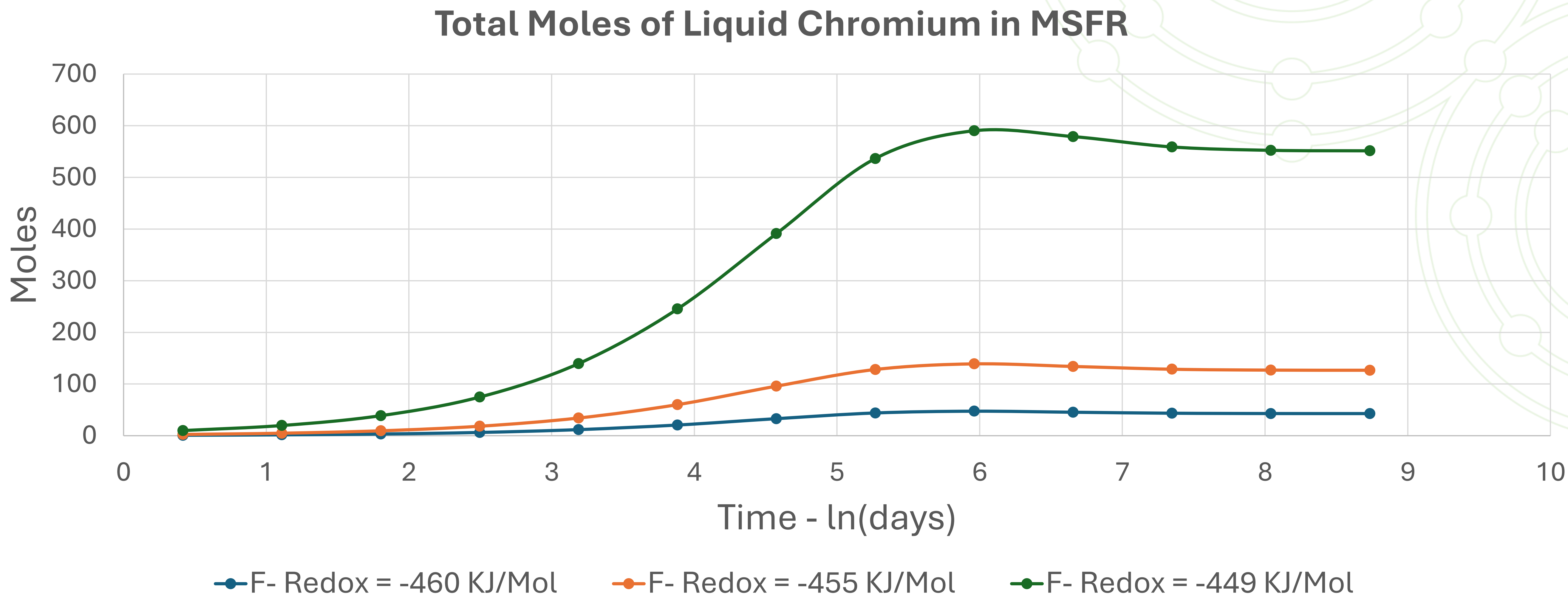
Effect of Redox Potential on Leaching



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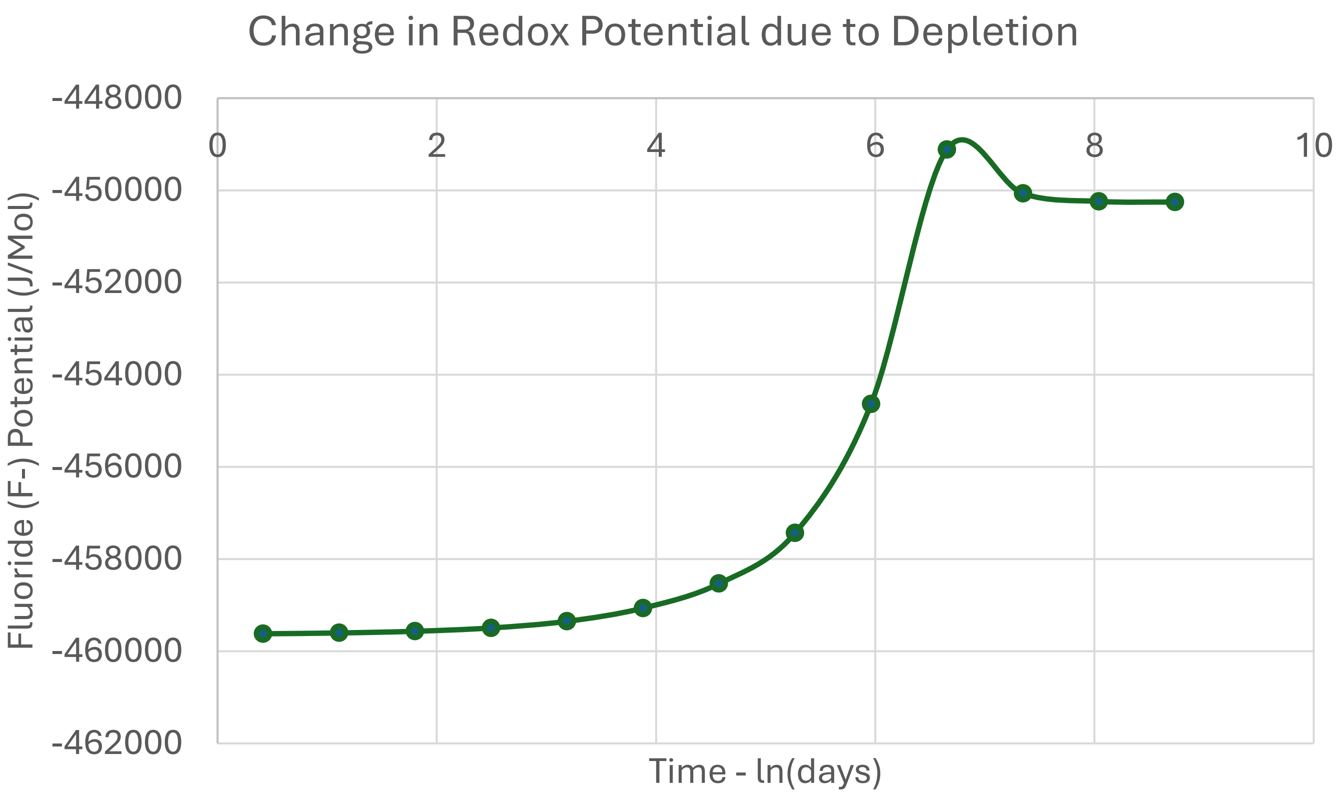
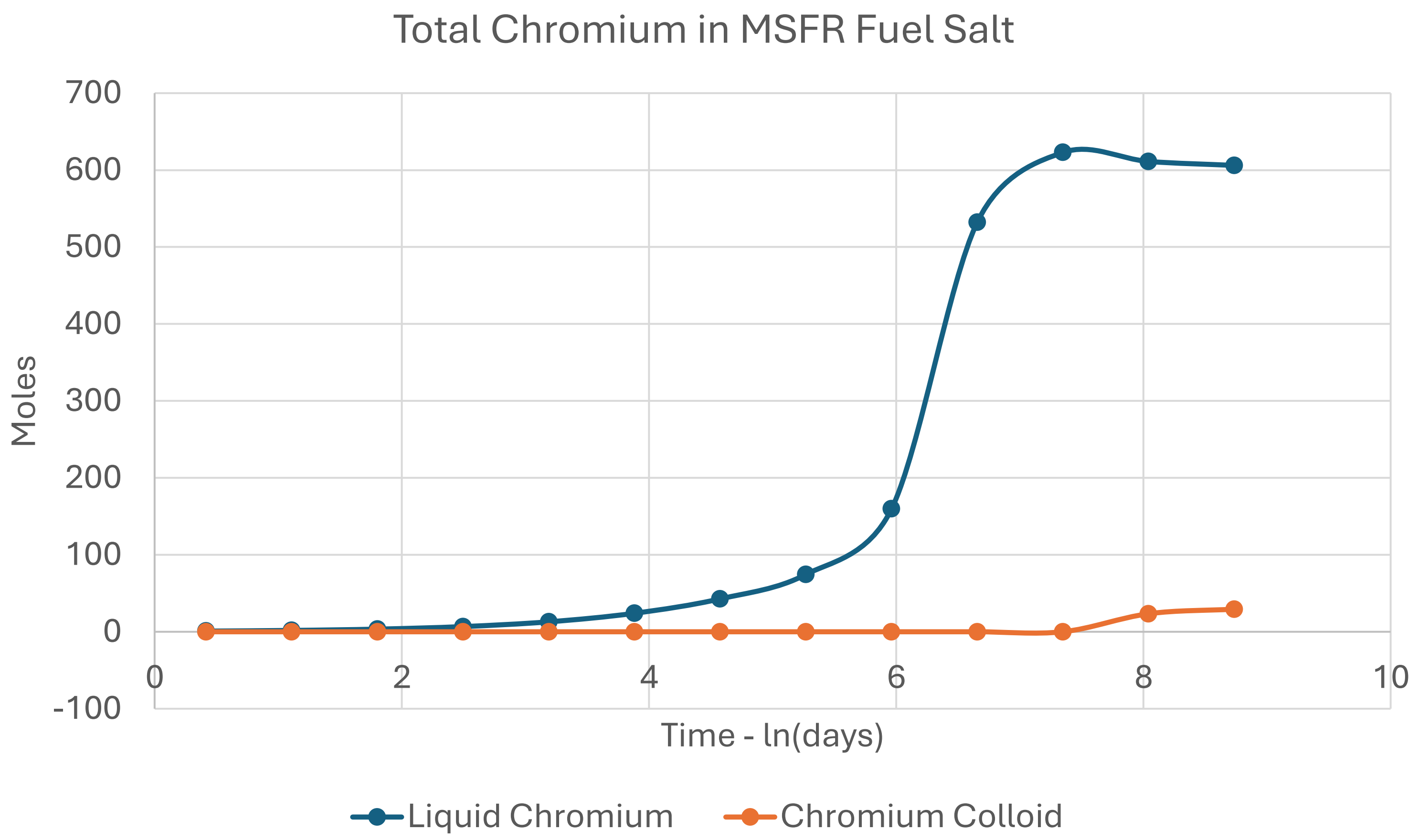
Effect of Redox Potential on Leaching



Increasing Redox Potential due to Depletion – driving higher solubility of Chromium in Fuel Salt



Decrease of redox potential at end of life due to end of depletion and buffer by chromium equilibrium in the fuel salt (illustrated by colloid formation in cold leg)

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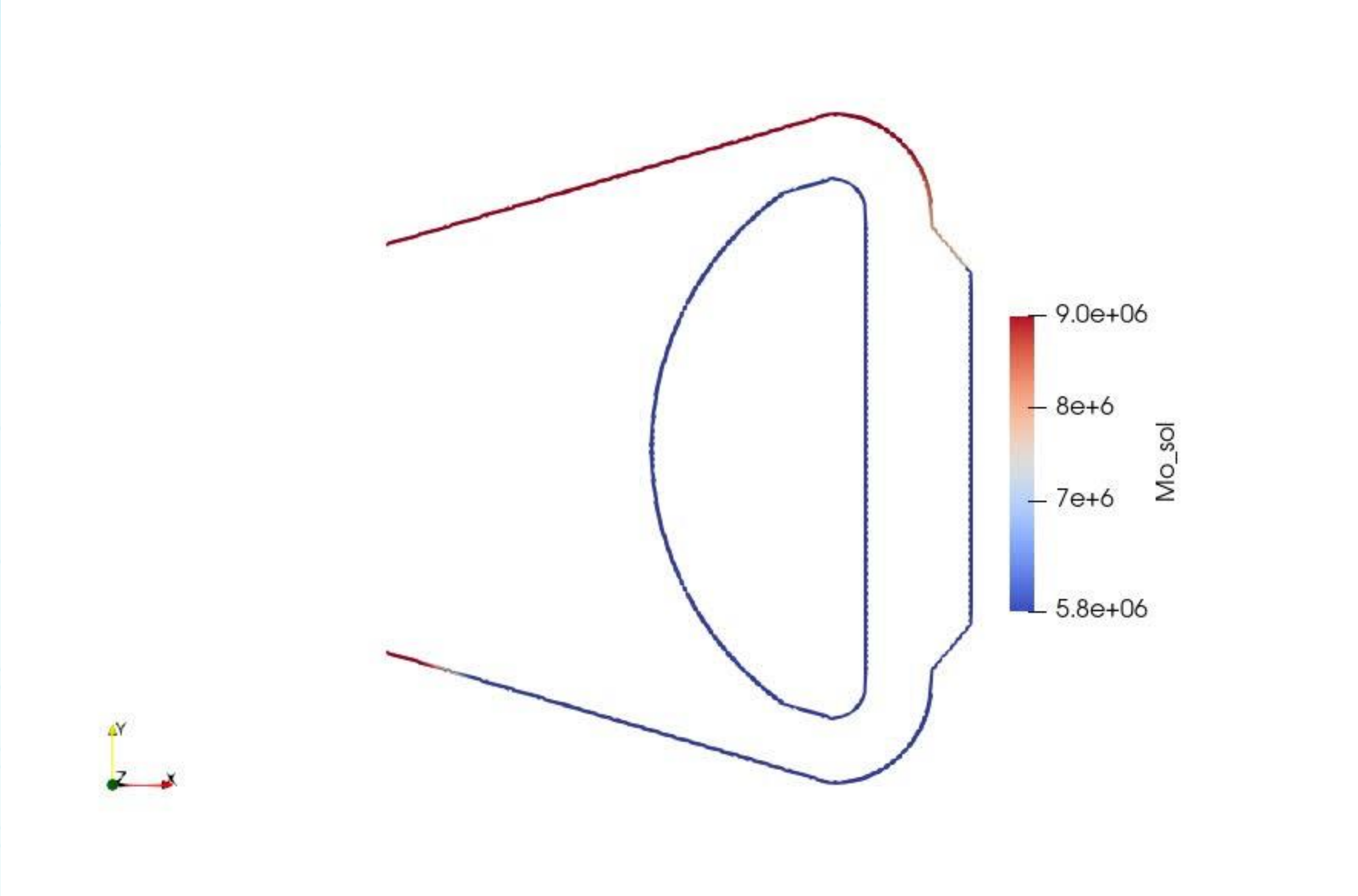
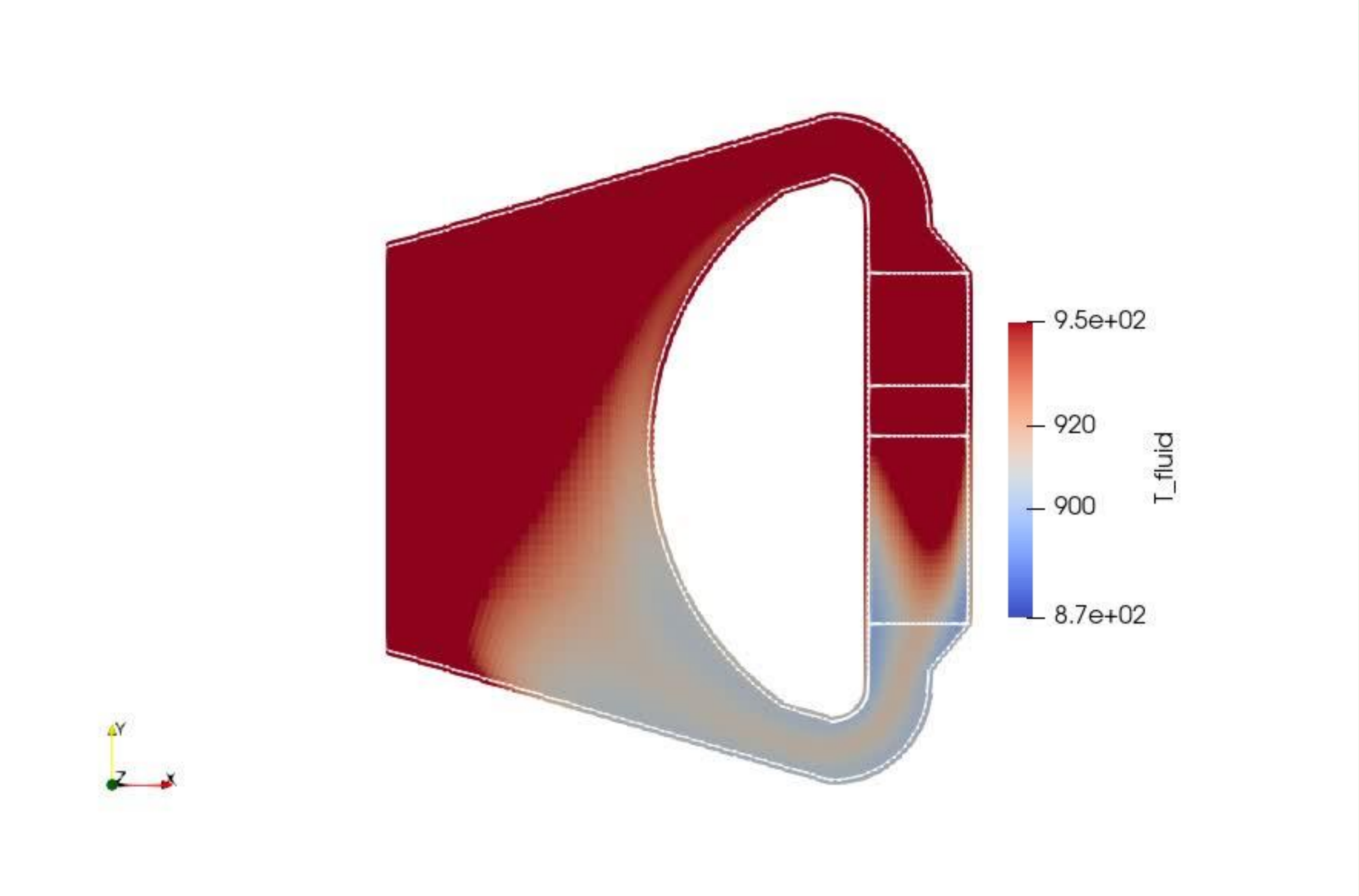
Effect of Noble Metal Decay Heat



Initial increased deposition of solid Molybdenum on wall and gradual decay after reactor scram (Pump and Power shut down)



Temperature decrease and re-heating from walls due to noble metal decay after reactor scram (Pump and Power shut down)

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Conclusions and Future Work

- **Key conclusions:**
  - New framework for coupled soluble + metallic species tracking has been implemented using NEAMS tools
  - The model has been implemented for a Molten Salt Loop Experiment and compared against higher fidelity PNP model and applied to MSFR and shows importance of species tracking for corrosion and noble metal decay heat modeling for reactor design and safety analyses.
- **Ongoing work:**
  - Further refined validation of model results against Molten Salt Loop Experient utilizing correct alloy database in Thermochimica for molten salt with metallic alloy thermochemical equilibrium calculations.
  - Validation of complete species transport (soluble, gaseous, metallic) results against MSRE composition measurements in salt samples, off-gas measurements, and heat exchanger.
  - Application of plating/leaching model to model electrochemistry in various applications (i.e., fuel-salt production, electrorefining, fluorination etc.)

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Thank you!

Any Questions?

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