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Multiphysics Depletion & Chemical Analysis of Molten Salt Reactors

Dr. Samuel Walker

INL, USA

17 April 2024

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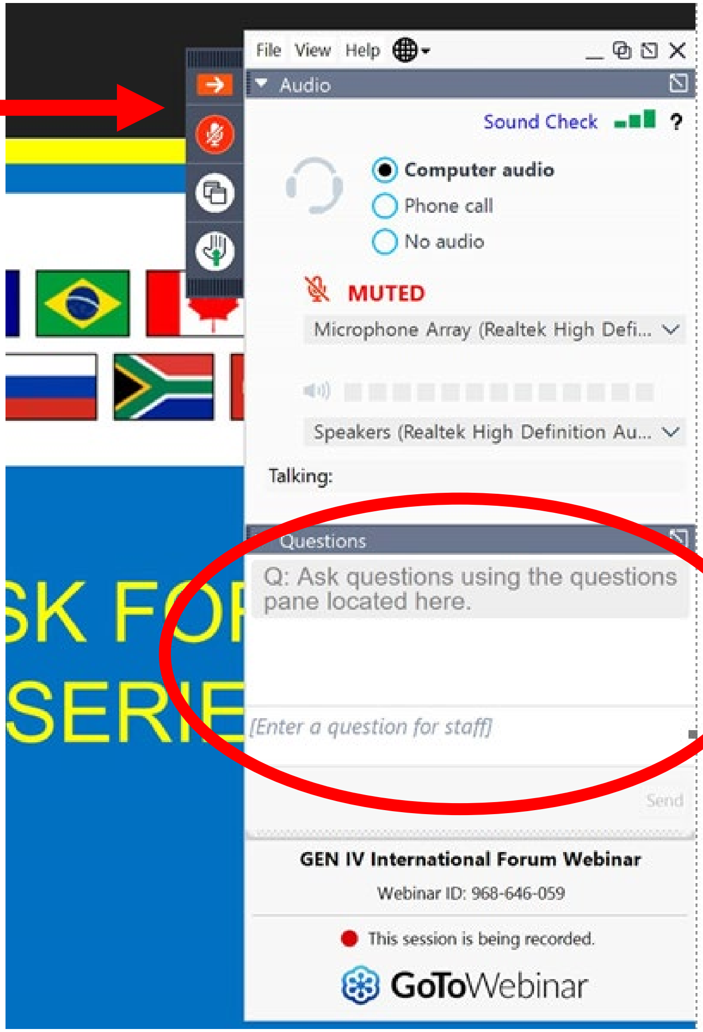

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Meet the Presenter

Dr. Samuel Walker is an R&D Staff Scientist in the Advanced Reactor Technology & Design Department of Idaho National Laboratory (INL).

He earned his Ph.D. in Nuclear Engineering from Rensselaer Polytechnic Institute in 2021 where he worked developing mass transfer modeling approaches for insoluble fission product transport in Molten Salt Reactor (MSR) systems. His graduate work was funded by a Department of Energy Nuclear Energy University Program (DOE NEUP) Fellowship that he was awarded in 2017.

His current work at INL focuses on coupling Nuclear Energy Advanced Modeling and Simulation (NEAMS) tools for multi-scale and multi-physics analysis of advanced reactors with a heavy focus on MSR multiphysics.

His expertise lies in modeling chemical species transport phenomena in molten salts used in fission and fusion systems. Applications of his work include source term and safety analyses, multiphysics core and system design, chemistry control system modeling, and novel MSR safeguard approaches.

Email: samuel.walker@inl.gov

Outline of Talk

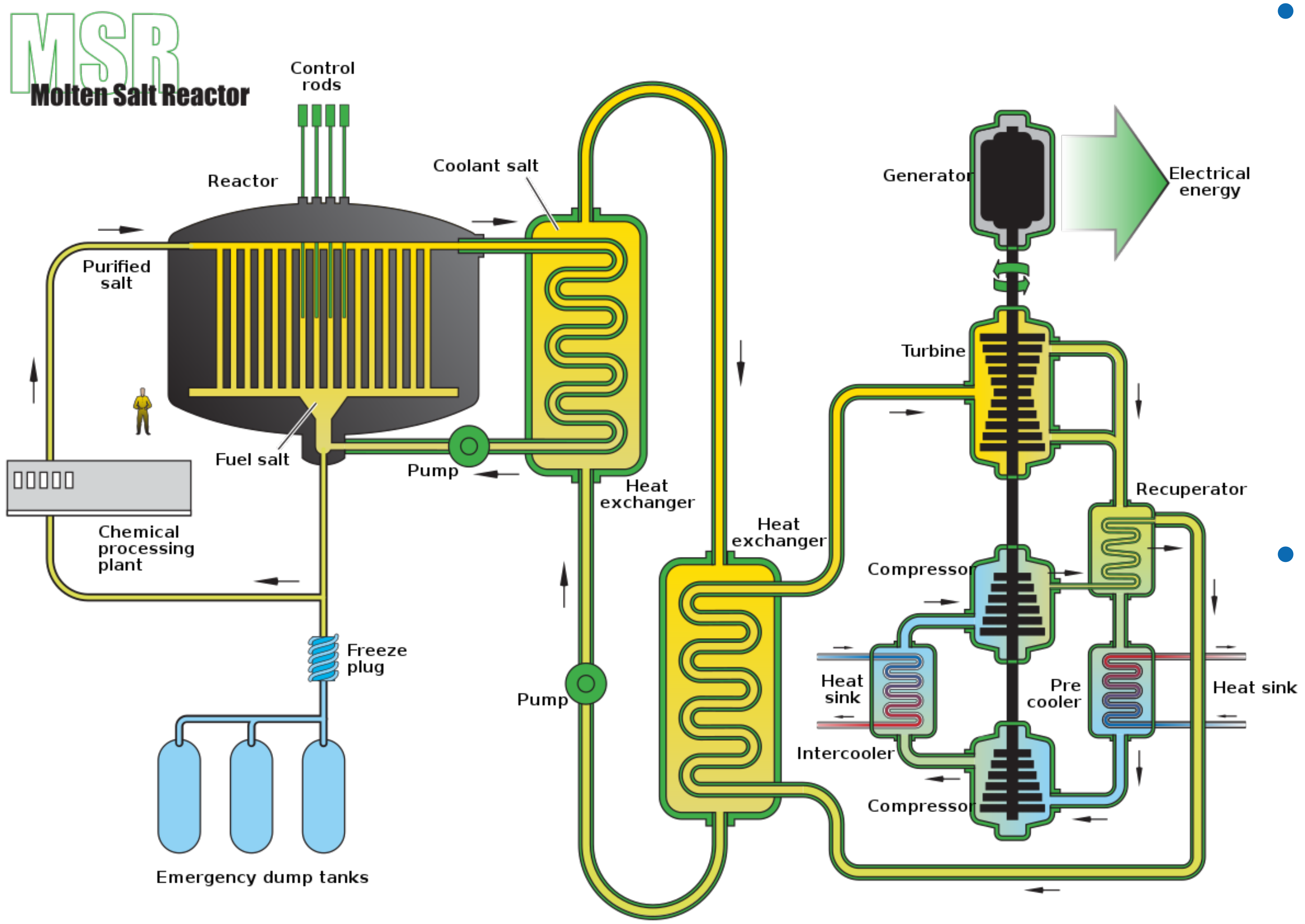
- Big Idea: Depletion-Driven Spatially-Resolved Thermochemistry & Chemical Species Transport in Molten Salt Reactors (MSRs)
- History Review: Molten Salt Reactor Experiment (MSRE) – Multiphysics Liquid Fuel Issues
- Nuclear Energy Advanced Modeling and Simulation (NEAMS) Framework for MSR Analysis
- Various Application Studies of Multiphysics Depletion & Chemical Species Transport Analysis
- Ongoing and Future Work

Credits – Collaborators

- Collaborators:
  - **Mauricio Tano** (mauricio.tanoretamales@inl.gov)
  - Abdalla Abou-Jaoude (abdalla.aboujaoude@inl.gov)
  - Parikshit Bajpai (parikshit.bajpai@inl.gov)
  - Rodrigo de Oliveira (rodrigo.deoliveira@inl.gov)
  - Ryan Stewart
  - Odera Dim (Brookhaven National Laboratory)
  - Thomas Fuerst
  - Olin Calvin

- Mentors
  - Wei Ji (Rensselaer Polytechnic Institute)
  - Gerhard Strydom

Why Molten Salts?



- Enhanced Passive Safety
  - Operate at low pressures (1-5 Atm)
  - Operate at low fuel temperature (~650° C)
  - Prompt negative temperature coefficient – liquid fuel expansion
  - Emergency drain tanks
- Enhanced Economic Opportunities
  - Process heat (high coolant temperatures)
  - Medical Isotope production (^{99}Mo)
  - Fuel cycle flexibility (Th – U, U – Pu)
  - Neutron spectrum flexibility (Breed & Burn, Experimental Reactor)

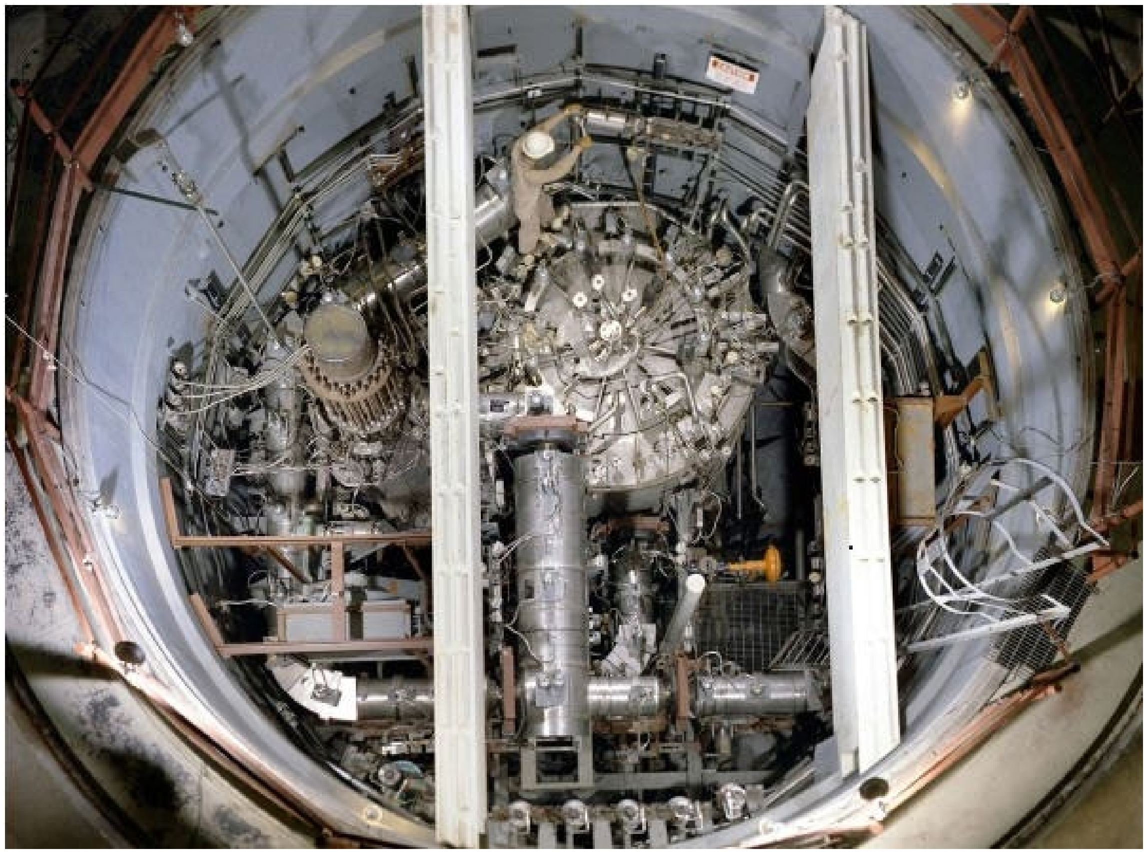
https://www.gen-4.org/gif/jcms/c_9359/msr

History Review: Molten Salt Reactor Experiment (MSRE)

- MSRE – Oak Ridge National Laboratory Design operated during the 1960's under Alvin Weinberg.

- UF_4 or ThF_4 Fuel in liquid phase is mixed with primary salt coolant.

- Proof of Concept - began a new way of doing nuclear energy.



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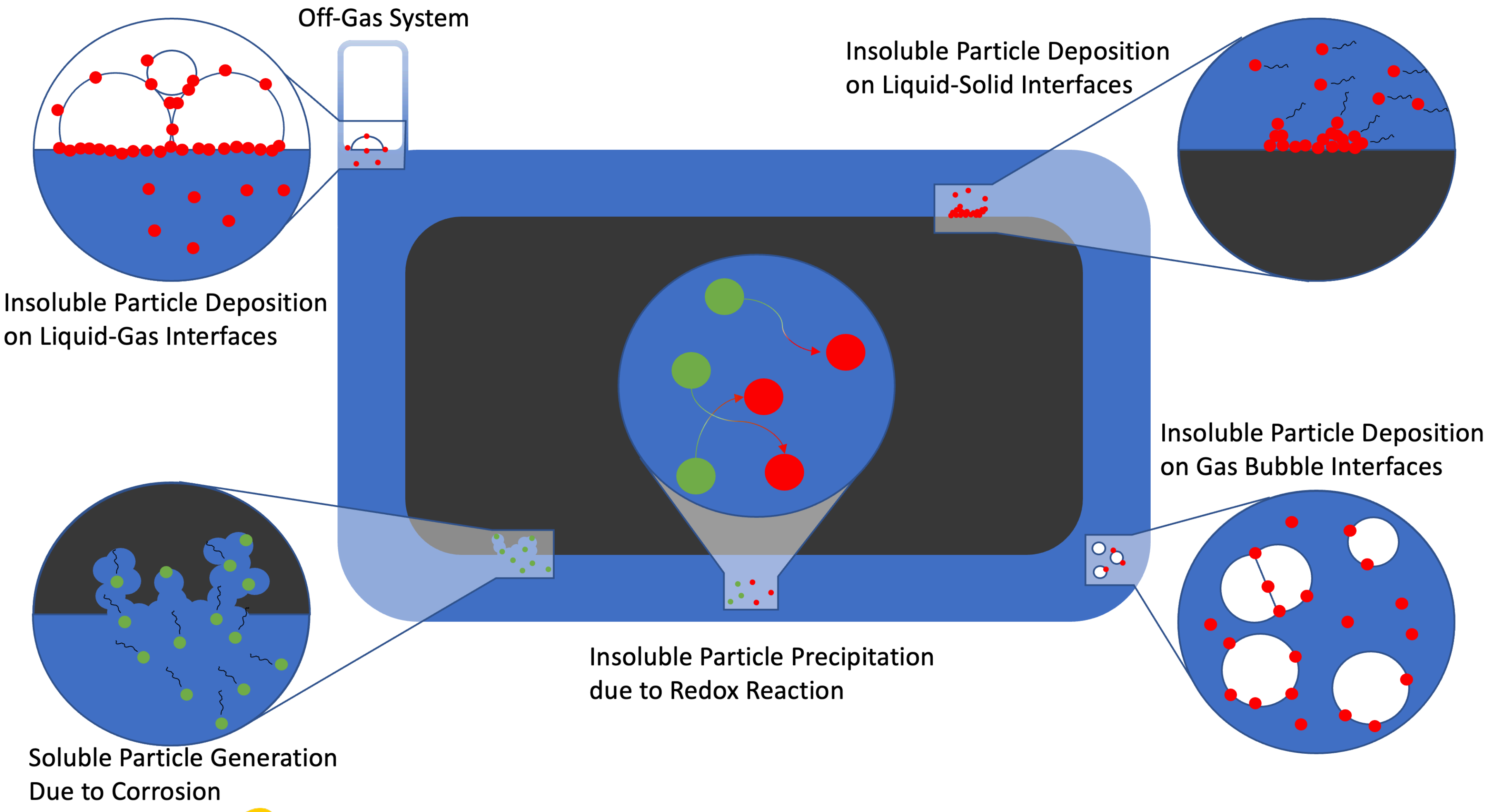
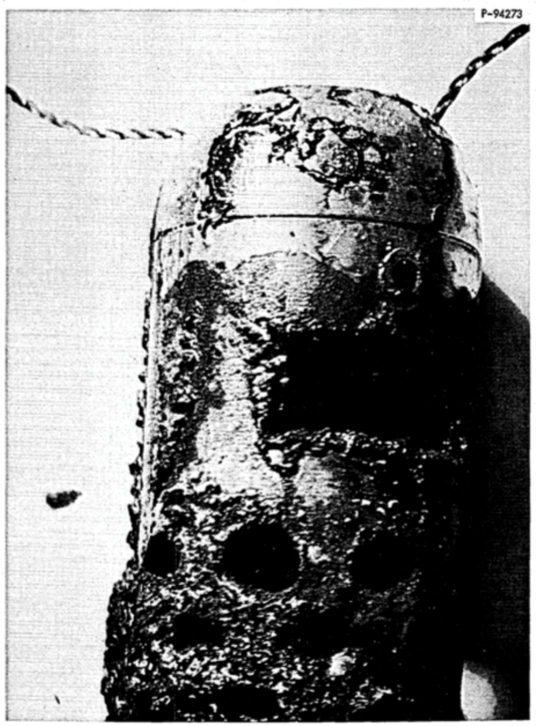
Depletion Driven Thermochemical Effects & Chemical Species Transport





Reformatted by Permission from Oak Ridge National Laboratory: J. R. Engel, P. N. Haubenreich and A. Houtzeel, "Spray, mist, bubbles, and foam in the Molten Salt Reactor Experiment," Oak Ridge Nat. Lab., Oak Ridge, TN, USA, ORNL-TM-3027, Jun. 1970

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1. Redox Potential Control → Corrosion Mitigation
2. Off-Gas System Control → Source Term Characterization
3. Material Accountancy → Safeguards & Decay Heat

Fluid Fuel Issues in the MSRE – Redox Potential

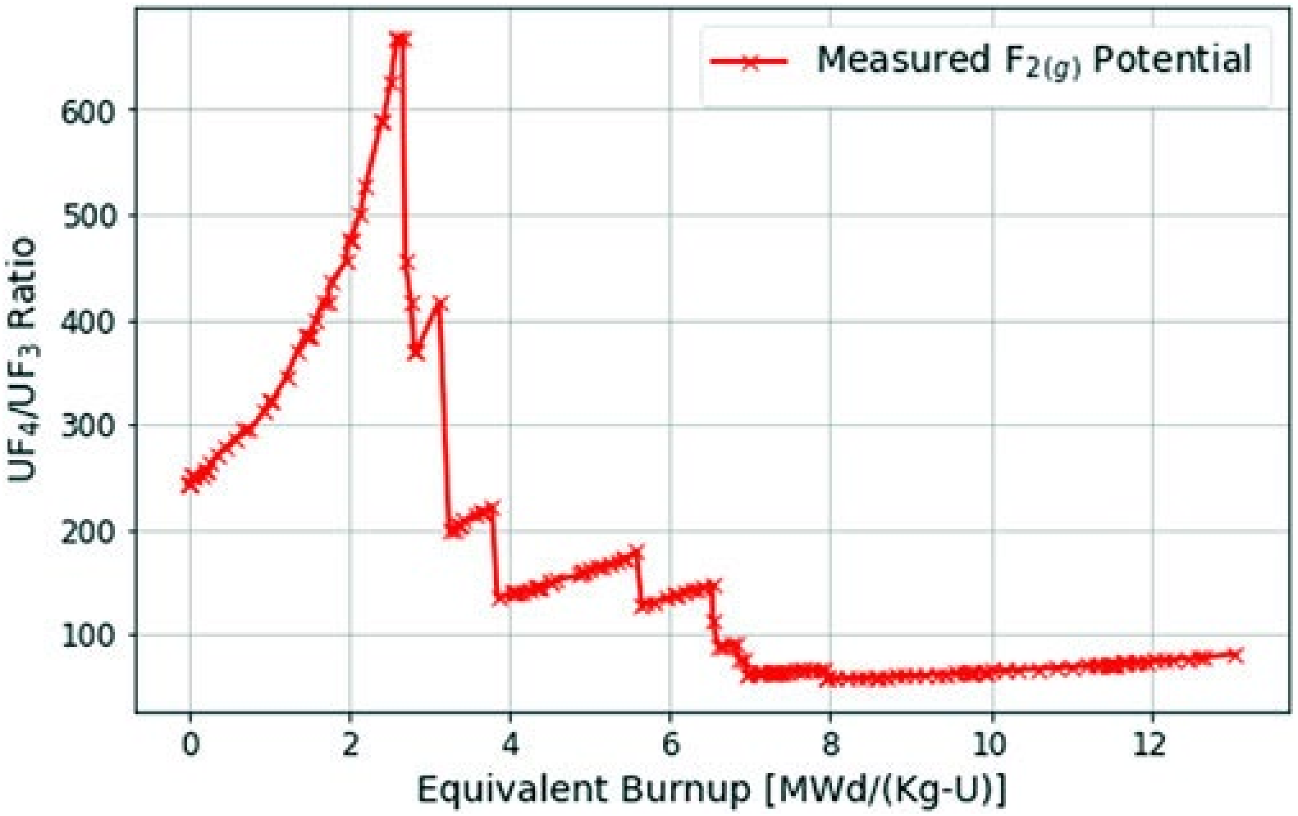

Figure: Experimental UF_4 to UF_3 ratio measurements during the ^{235}U MSRE runs – reformatted from (Thoma 1971 – ORNL-4658)

Fission product	Assumed eq. ox. state (Z)	Yield Y	YZ
Br + I	−1	0.015	−0.015
Kr + Xe	0	0.606[a)]	0
Rb + Cs	+1	0.004	0.004
Sr + Ba	+2	0.072	0.144
Lanthanides + Y	+3	0.538	1.644
Zr	+4	0.318	1.272
Nb	0	0.014	0
Mo	0	0.201	0
Tc	0	0.059	0
Ru	0	0.126	0
		1.953	3.049

a) With rapid stripping from the system.

Figure: Chemical consequences of fission in an MSBR – from (Baes, C. F. (1974). *J. Nucl. Mat.* 51, 149–162)

i.e., $UF_4 + UF_3 \rightarrow YF_3 + Xe + UF_4$

$$Cr^0(s)+2UF_4 \leftrightarrow 2UF_3+CrF_2$$

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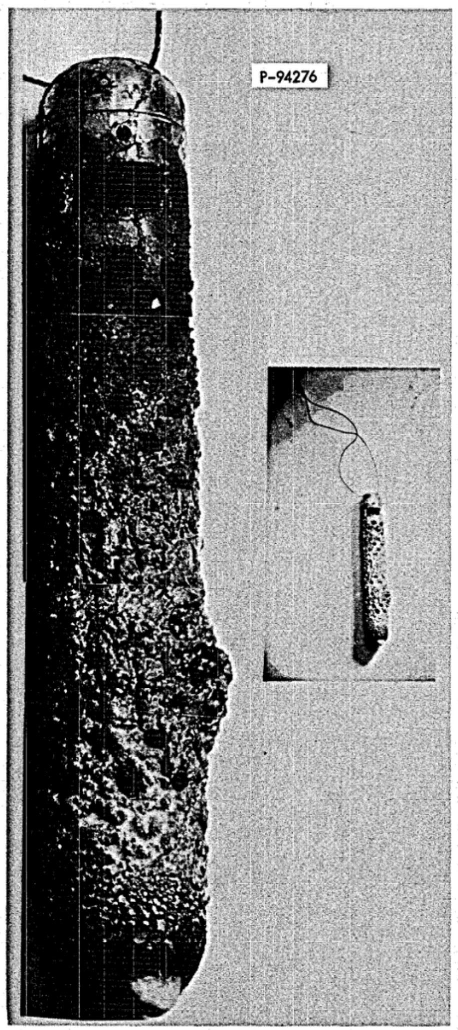
Controlling the Redox Potential

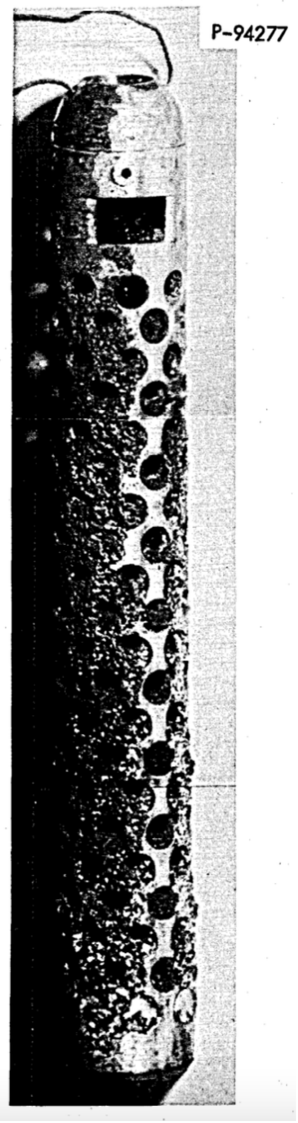
$$Cr^0(s)+2UF_4 \leftrightarrow 2UF_3+CrF_2$$

$$Fe^0(s)+2UF_4 \leftrightarrow 2UF_3+FeF_2$$

$$Be^0(s)+2UF_4 \leftrightarrow 2UF_3+BeF_2$$

Metallic deposits on nickel cage housing of beryllium rod during ^{233}U runs reformatted from (J. R. Engel, P. N. Haubenreich and A. Houtzeel, "Spray, mist, bubbles, and foam in the Molten Salt Reactor Experiment," Oak Ridge Nat. Lab., Oak Ridge, TN, USA, ORNL-TM-3027, Jun. 1970)





Calculated Electrode Potential in LiF-0.33BeF$_2$ (1,000 K) – Reformatted from (Baes, Jr. 1965 *J. Nucl. Mater.* 59, 149.)

Half Cell Reactions	E^O (1,000 [°K]), [V]
$Li^+ + e^- \Leftrightarrow Li_{(s)}$	−5.412
$La^{3+} + 3e^- \Leftrightarrow La_{(s)}$	−5.081
$Ce^{3+} + 3e^- \Leftrightarrow Ce_{(s)}$	−5.011
$Sm^{3+} + 3e^- \Leftrightarrow Sm_{(s)}$	−4.881
$Th^{4+} + 4e^- \Leftrightarrow Th_{(s)}$	−4.601
$Be^{2+} + 2e^- \Leftrightarrow Be_{(s)}$	−4.592
$U^{3+} + 3e^- \Leftrightarrow U_{(s)}$	−4.281
$U^{4+} + 4e^- \Leftrightarrow U_{(s)}$	−4.190
$Zr^{4+} + e^- \Leftrightarrow Zr_{(s)}$	−4.187
$U^{4+} + e^- \Leftrightarrow U^{3+}$	−3.915
$Cr^{2+} + 2e^- \Leftrightarrow Cr_{(s)}$	−3.261
$Fe^{2+} + 2e^- \Leftrightarrow Fe_{(s)}$	−2.882
$HF_{(g)} + e^- \Leftrightarrow F^- + \frac{1}{2}H_{2\ (g)}$	−2.871
$Ni^{2+} + 2e^- \Leftrightarrow Ni_{(s)}$	−2.398
$\frac{1}{2}F_2(g) + e \Leftrightarrow F^-$	0.0

Fluid Fuel Issues in the MSRE – Liquid-gas Interface Phenomena

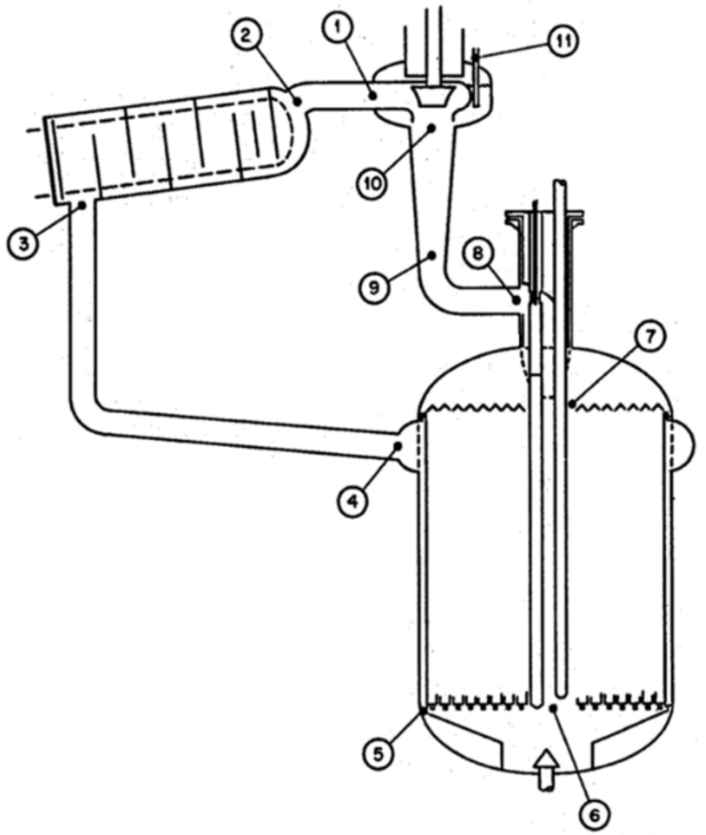


Figure: MSRE Primary Loop - reformatted from (Engel et al. (1970), ORNL-TM-3027)

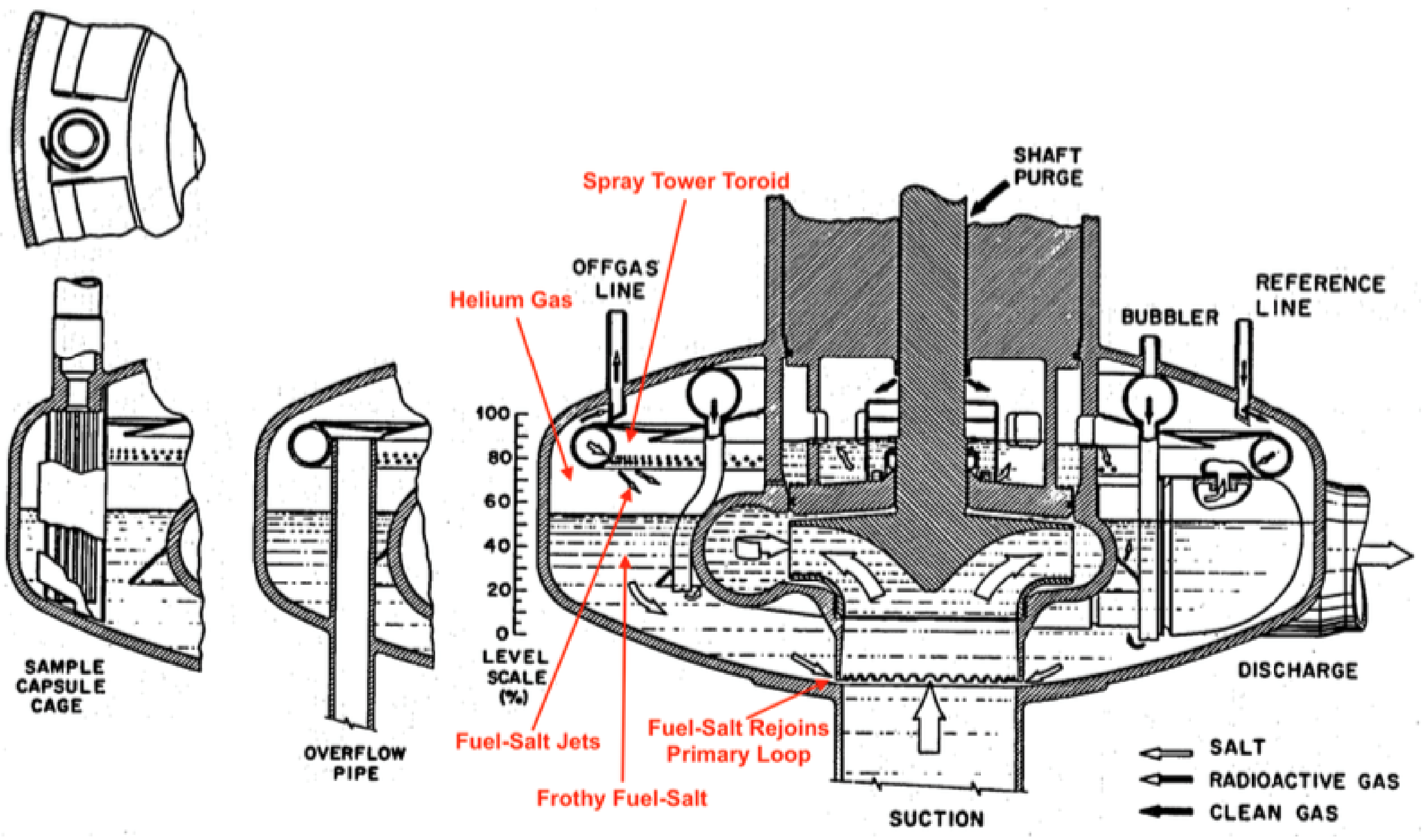


Figure: MSRE Pump Bowl – reformatted from (Engel et al. (1970), ORNL-TM-3027)

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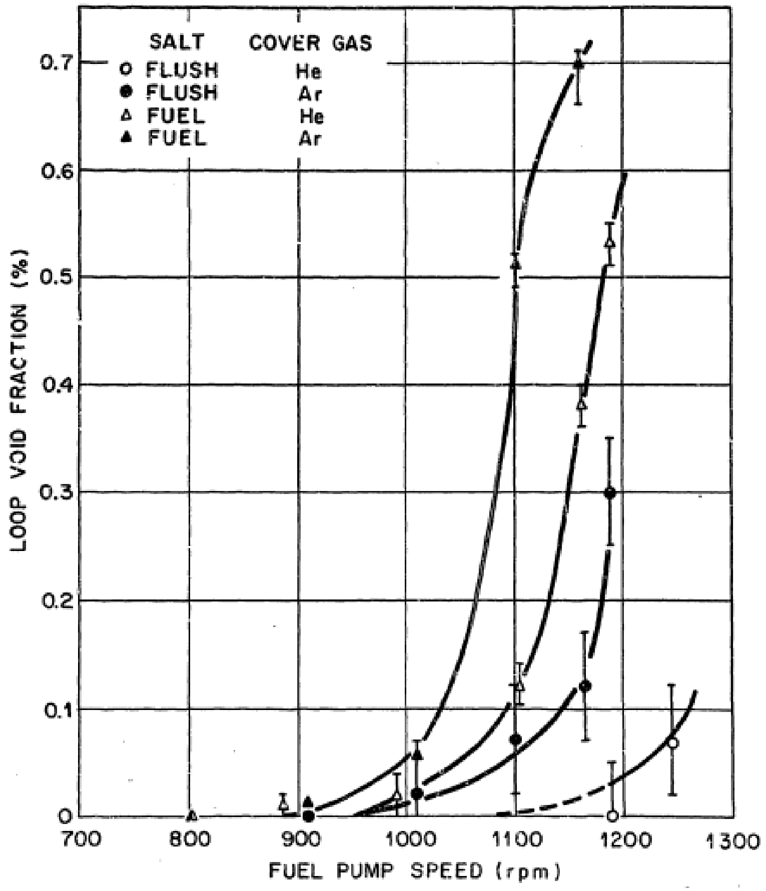
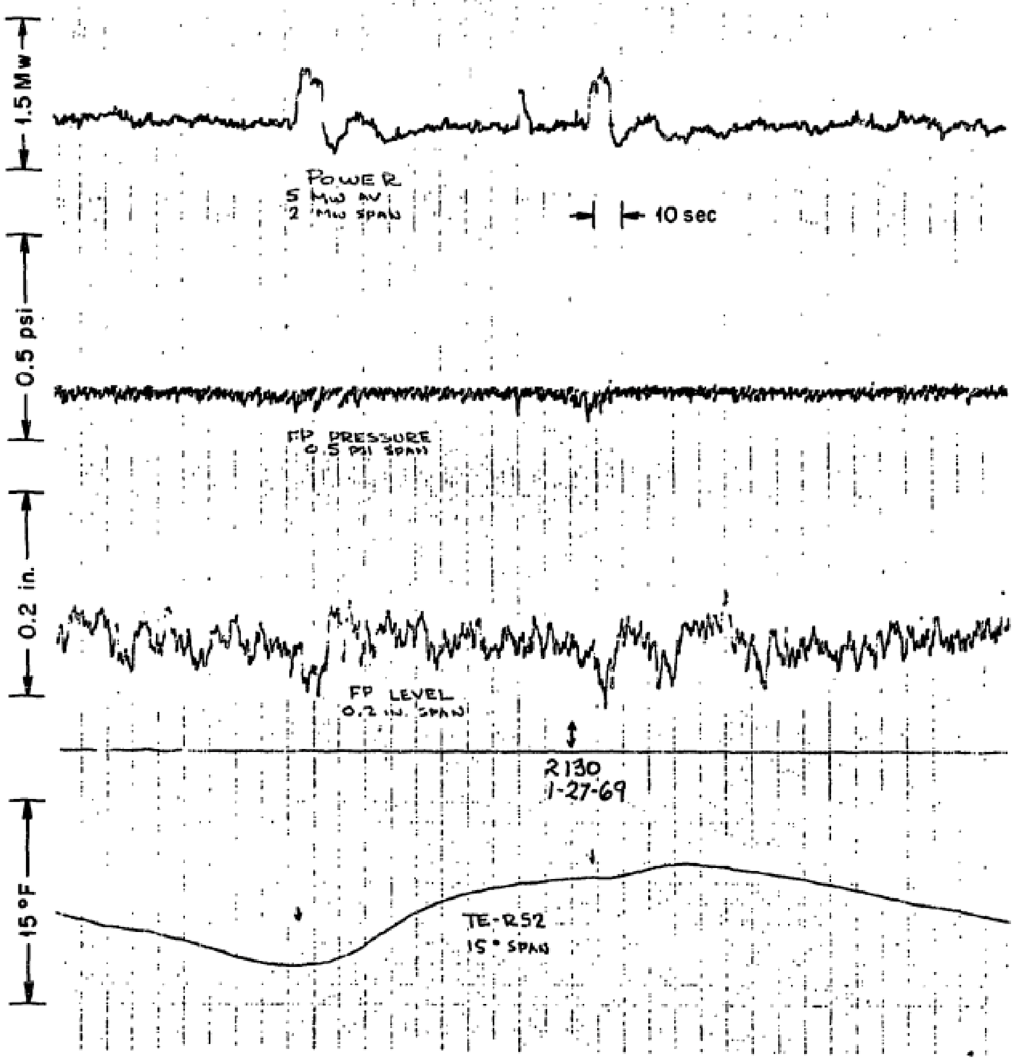
Liquid-gas Interface Phenomena Causing Mist, Bubbles, and Circulating Voids

Figure left: Effect of Fuel Pump Speed on Void Fraction in Fuel Loop - reformatted from (Engel et al. (1970), ORNL-TM-3027)

Figure right: Nuclear Power, Fuel-Pump Pressure, Indicated Fuel-Pump Level, and Thermocouple reading during blips - reformatted from (Engel et al. (1970), ORNL-TM-3027)





Mist and Foam – Surfactant Effects?

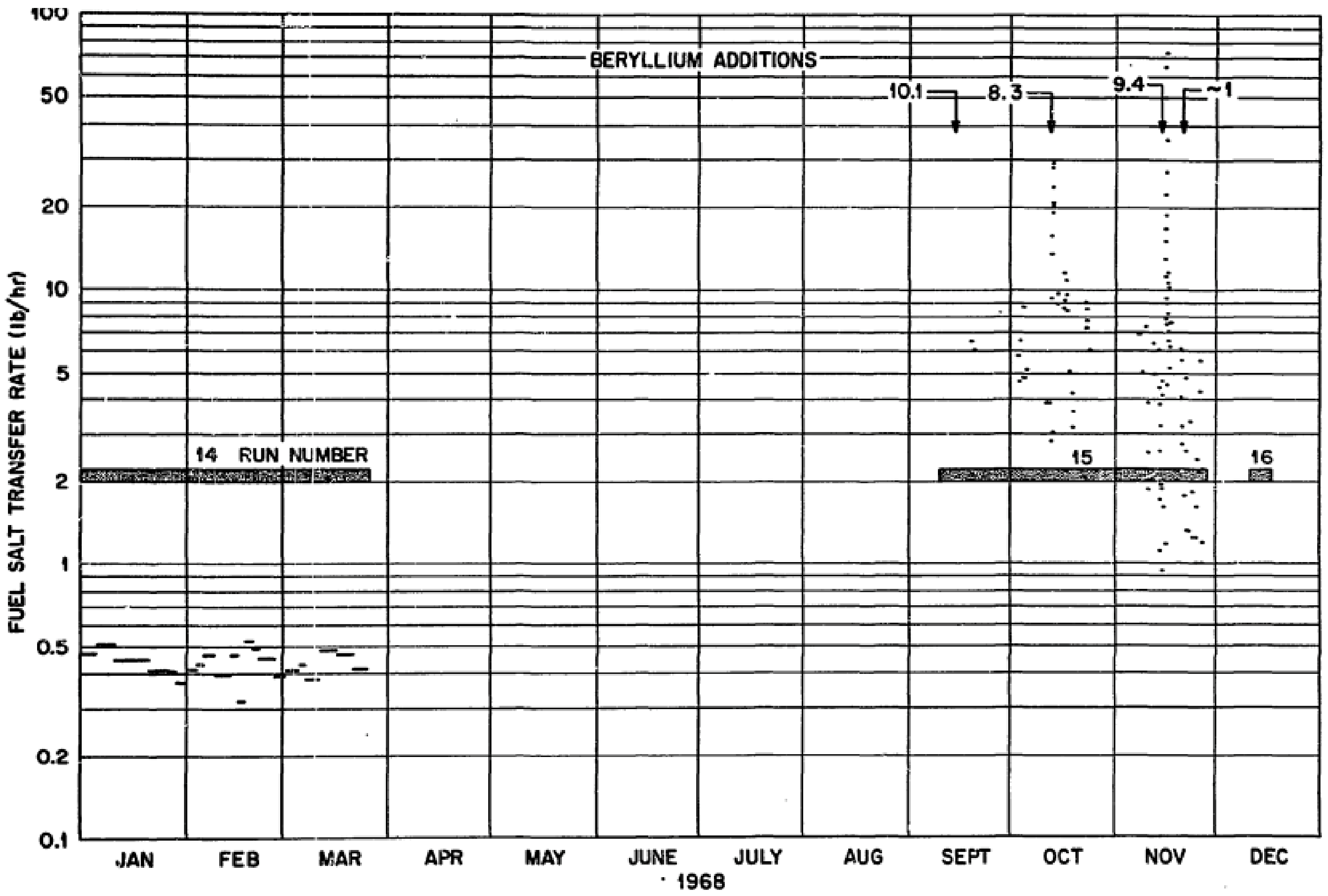
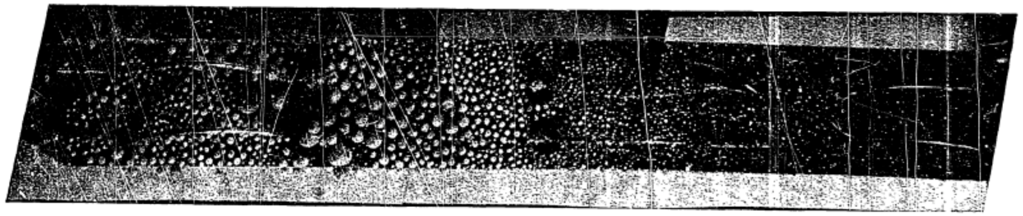


Figure: Measured Rates of salt overflow from Fuel Pump (Engel et al. (1970), ORNL-TM-3027)



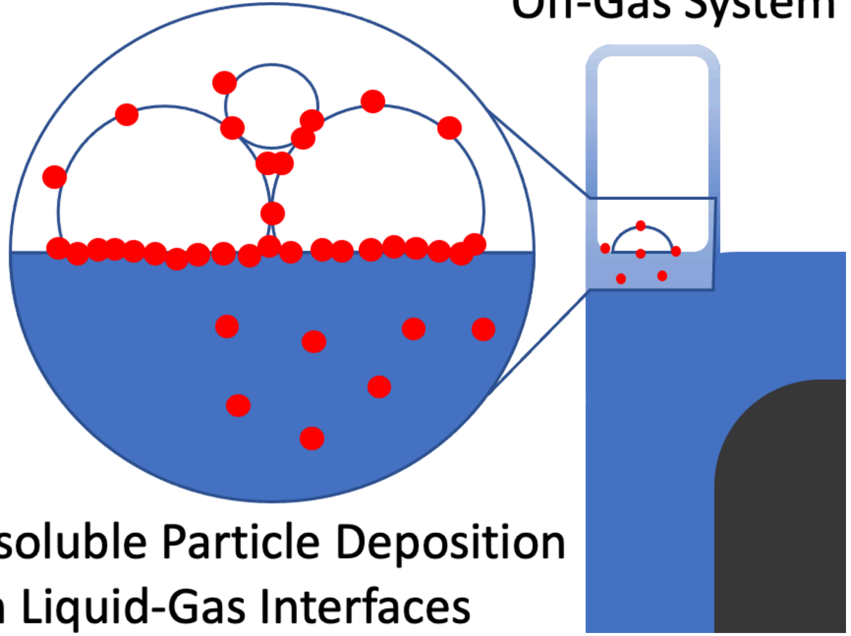


Figure top: Salt Droplets on a Metal Strip Exposed in MSRE Pump Bowl Gas Space for 10 Hours (Engel et al. (1970), ORNL-TM-3027)

Multiphysics Liquid Fuel Issues – A chemical species tracking problem

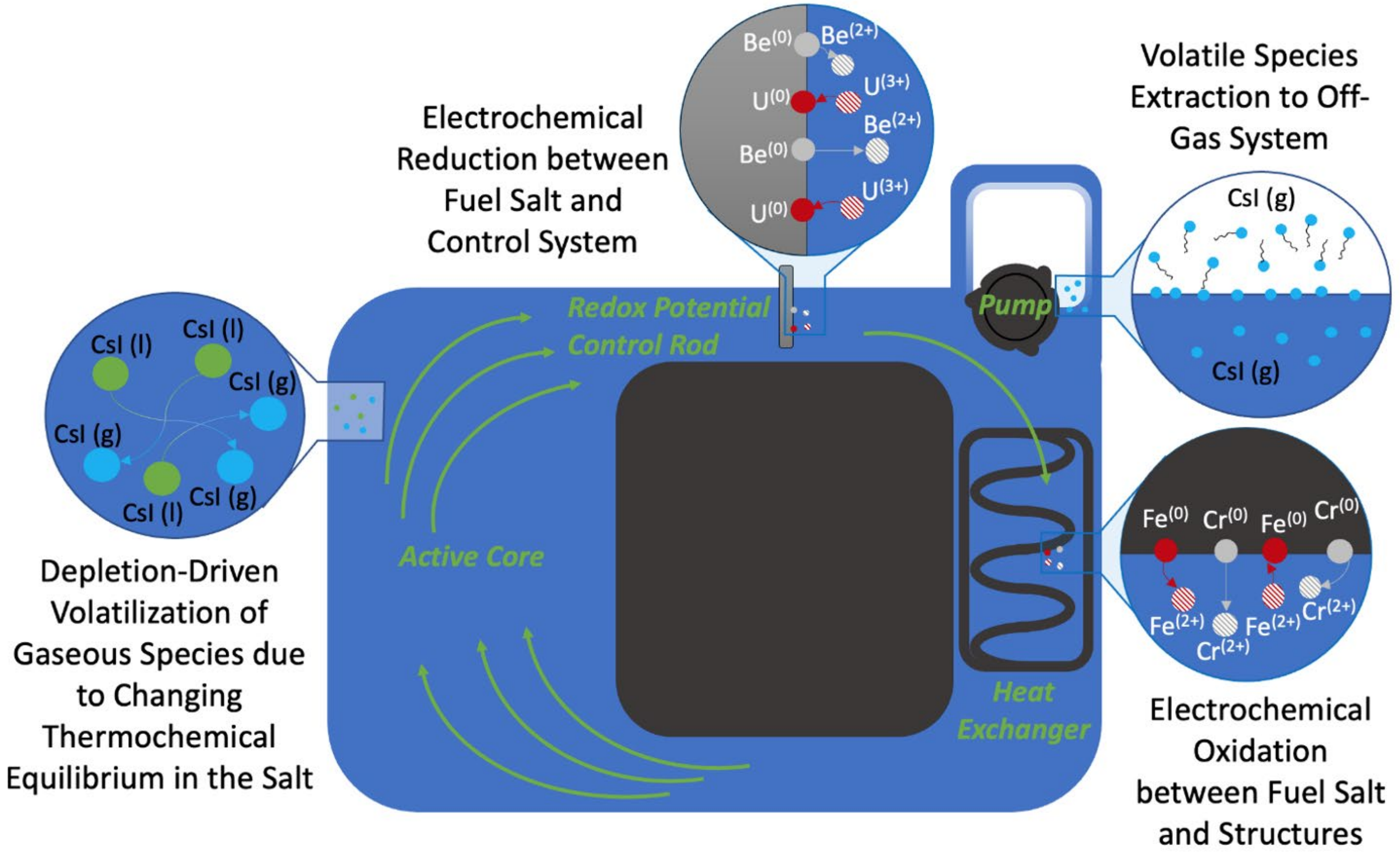


Figure: Chemical species transport examples in MSR system.

Walker et. al (2023) INL/RPT-23-74376

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

How do we tackle chemical species tracking challenges?

- Our Approach:

  1. Develop “approximate” or “wrong” albeit useful models to capture MSR multiphysics effects.

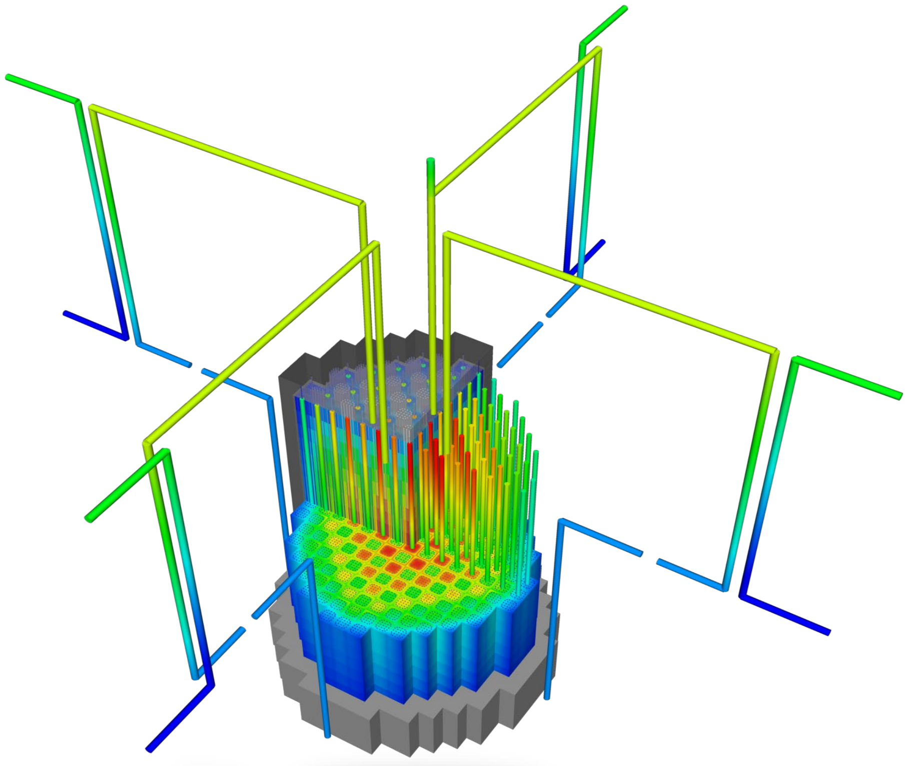
  2. Verify numerically that the “approximate” models are implemented correctly.

  3. Test and validate them against separate physics experiments.

  4. Use them to inform integral effects test designs and validate multiphysics models.

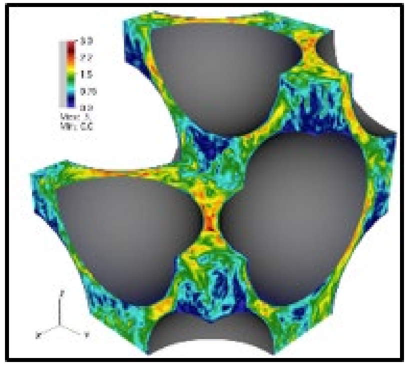
- Enter: Department of Energy’s Nuclear Energy Advanced Modeling and Simulation (NEAMS) suite of tools for Generation IV reactor design.

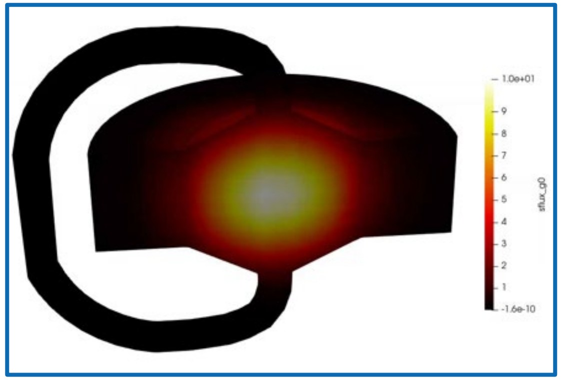
MOOSE

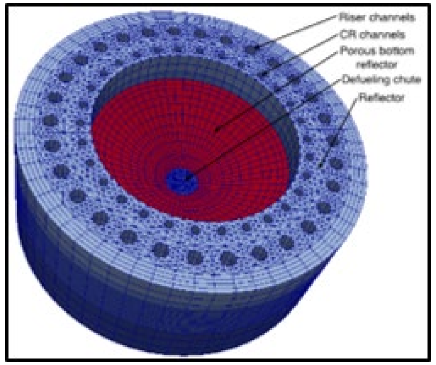


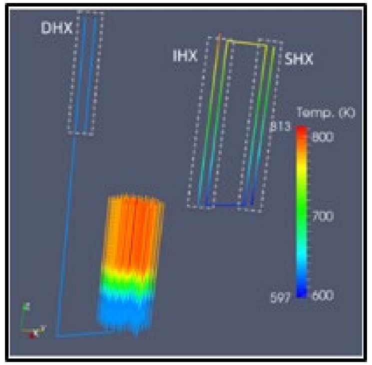
https://mooseframework.inl.gov/

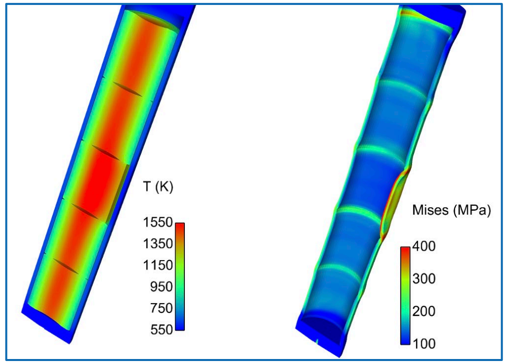
Main NEAMS tools used for modeling and simulation of MSRs











NEK-5000 + GRIFFIN + PRONGHORN + SAM + Bison/ Thermochimica

Fuel Performance/ Thermochemistry

High Fidelity

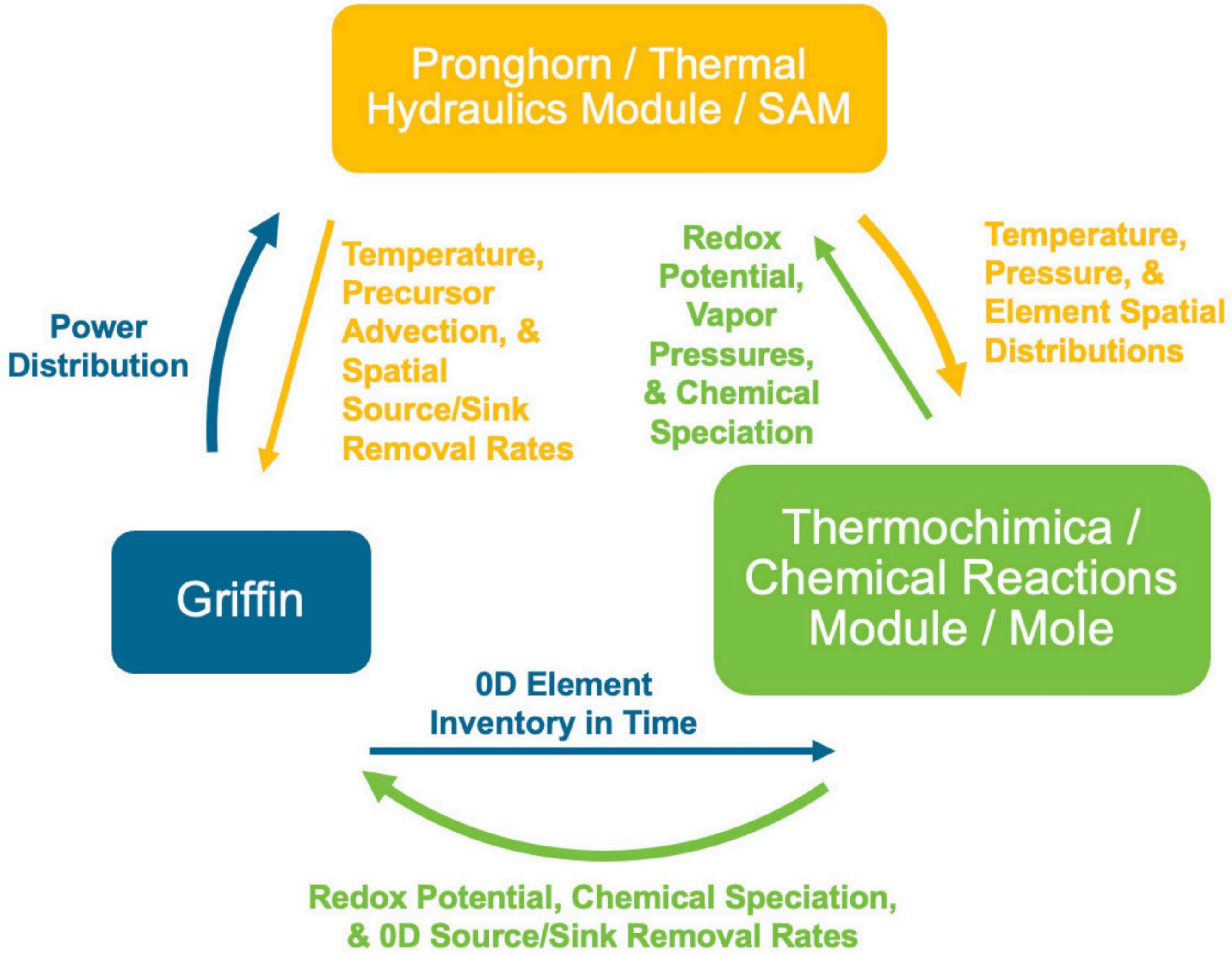
2D/3D Multiphysics Transients

Balance of Plant

Depletion & Fuel Performance

NEAMS Multiphysics Framework for Chemical Species Tracking in MSRs

- Specifically for chemical species tracking in MSRs the following framework has been developed using.
    1. Griffin: Neutronics & depletion
    2. Pronghorn: Thermal-hydraulics and species transport
    3. Thermochimica: Chemical equilibrium calculations and speciation.



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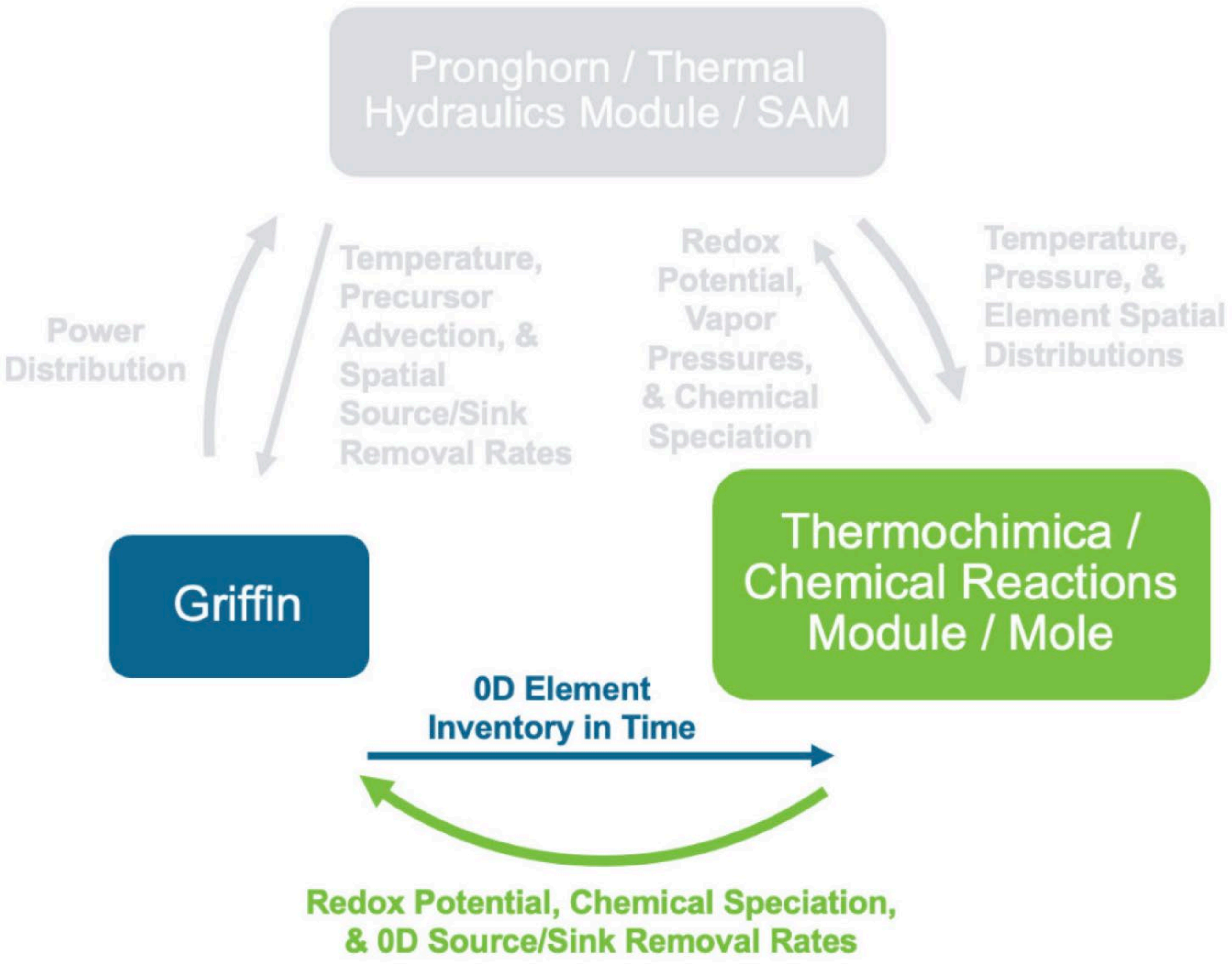
Depletion-Driven Thermochemistry

- Thermochimica – An open-source Gibbs Energy Minimizer developed by Markus Piro and team at McMaster University. (https://github.com/ORNL-CEES/Thermochimica)
  - Available now for easy coupling in the Chemical Reactions Module of MOOSE thanks to Parikshit Bajpai and Daniel Schwen at Idaho National Laboratory.
- Molten Salt Thermodynamic DataBase ThermoChemical – (MSTDB-TC) developed by Ted Besmann and team at South Carolina University. (https://mstdb.ornl.gov/)
  - An essential thermochemical database that Thermochimica uses to calculate chemical equilibrium.



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Study 1: Redox potential control for the Molten Salt Fast Reactor during Depletion

- Redox potential is controlled via the reaction $Be^0_{(s)} + 2UF_4 \leftrightarrow 2UF_3 + BeF_2$
  - $Be^0_{(s)}$ is a solid rod of beryllium immersed in the fuel salt,
  - UF_4 and UF_3 are a redox buffer pair which poise the chemical fluorine potential of the fuel salt system due to their relative chemical activities in the fuel salt
  - BeF_2 is the newly added beryllium which has dissolved reducing UF_4 to UF_3
- The interest of the control system is to protect the structural Ni in the steel structures, which may leach to the salt via the $Ni^0_{(s)} + 2UF_4 \leftrightarrow 2UF_3 + NiF_2$ reaction
- Also, Redox control can help mitigate the number of volatile species formed in the reactor, which assist in controlling the radiological source term
- Redox control is performed via a discrete PID system, where the signal u_k, which is the added mass of Be, and the error e_k, which is the difference between the desired and actual Fluorine potential, are governed as follows:

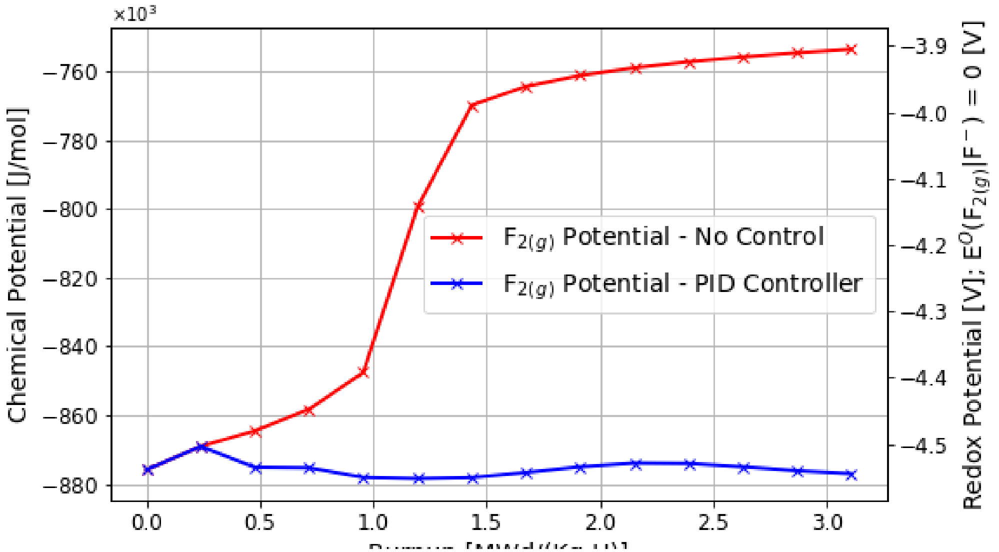
$$u_k = u_{k-1} + \left(K_p + K_i\Delta t + \frac{k_d}{\Delta T}\right)e_k + \left(-K_p - \frac{2K_d}{\Delta t}\right)e_{k-1} + \frac{K_d}{\Delta t}e_{k-2}$$

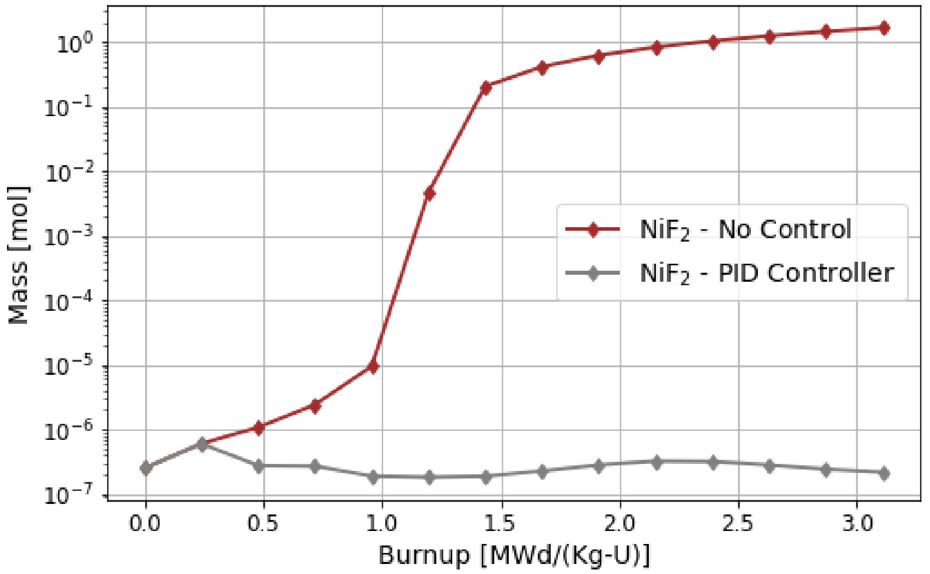
Walker et. al (2023) M&C 2023

Study 1: Impact of Redox Control on Ni Leaching

- Our models show that the Fluorine potential in the molten salt can be controlled by the addition of Be (previous experience in the MSRE confirms this hypothesis).

- Additionally, we see that the addition of Be stops the migration of Ni from the solid structures into the salt, confirming that Redox control is a key mechanism for maintaining structural integrity of the structures in the molten salt.





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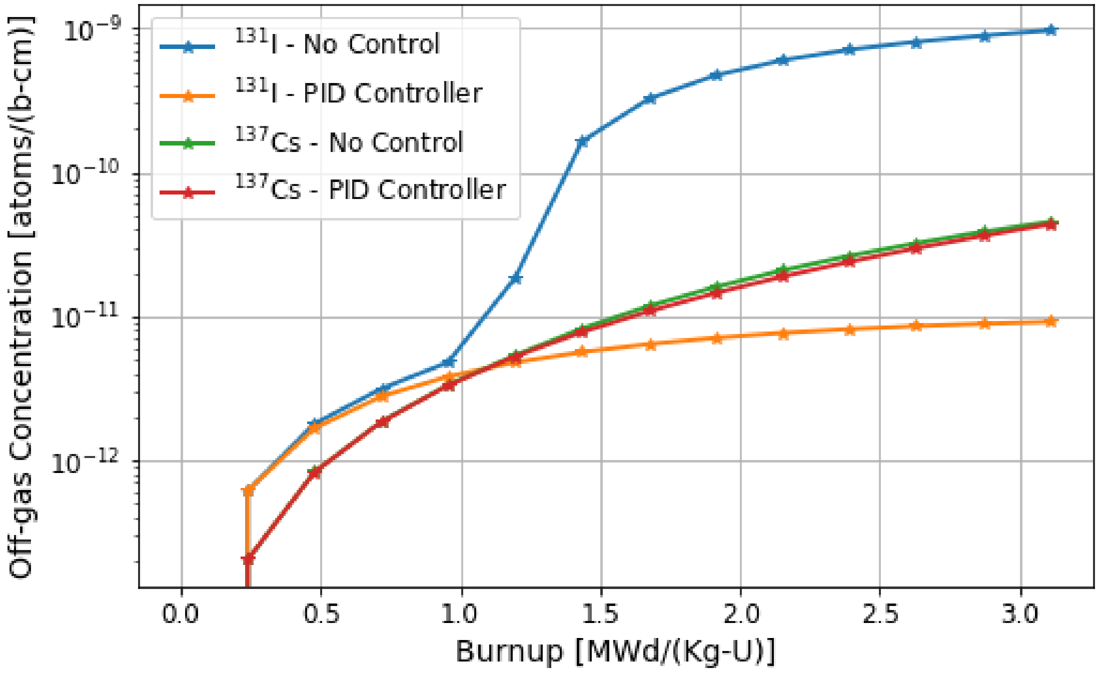
Study 1: Impact of Redox Control on Species Volatilization

- The volatilization of ^{131}I for the controlled case is mostly due to CsI(g) vapor formation and extraction to the off-gas system.
- For the uncontrolled situation, the formation of I_2(g) vapor becomes thermodynamically more favorable and significantly more iodine ends up in the off-gas system as a result.
- For the ^{137}Cs case, the impact of redox potential seems negligible. This is likely because ^{137}Cs vapor from CsI(g) formation can be significant, but a greater source for ^{137}Cs in the off-gas system is likely due to the extraction of noble gas ^{137}Xe which comes out of the fuel-salt more readily than CsI(g).



Percentage of Radionuclides in Off-gas System		Fluoride Salt Reactor
^{131}I	No Control	31.10 %
	PID Controller	0.29 %
^{37}Cs	No Control	0.11 %
	PID Controller	0.11 %

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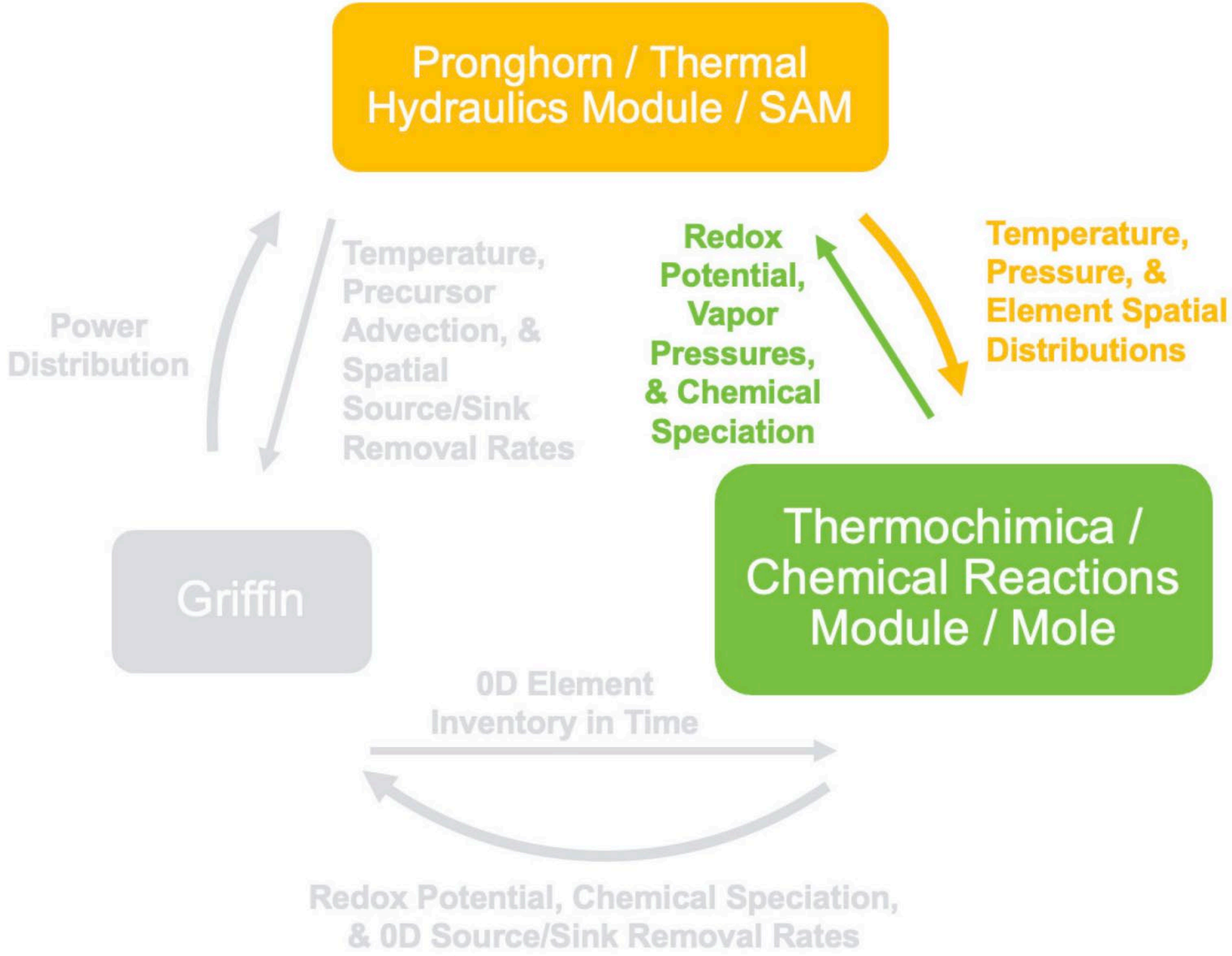
Spatially-Resolved Thermochemistry

- Pronghorn (a.k.a. Navier Stokes Module of MOOSE) – An open-source finite volume thermal hydraulics solver in MOOSE.
  - Reacting species transport and mass transfer capabilities for full partial differential equation resolution of nuclide decay chains.
  - New Two-Phase Flow capability for void tracking via a drift flux model.



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Study 2: Spatially Resolved Corrosion & Redox Potential Control for the Molten Salt Fast Reactor

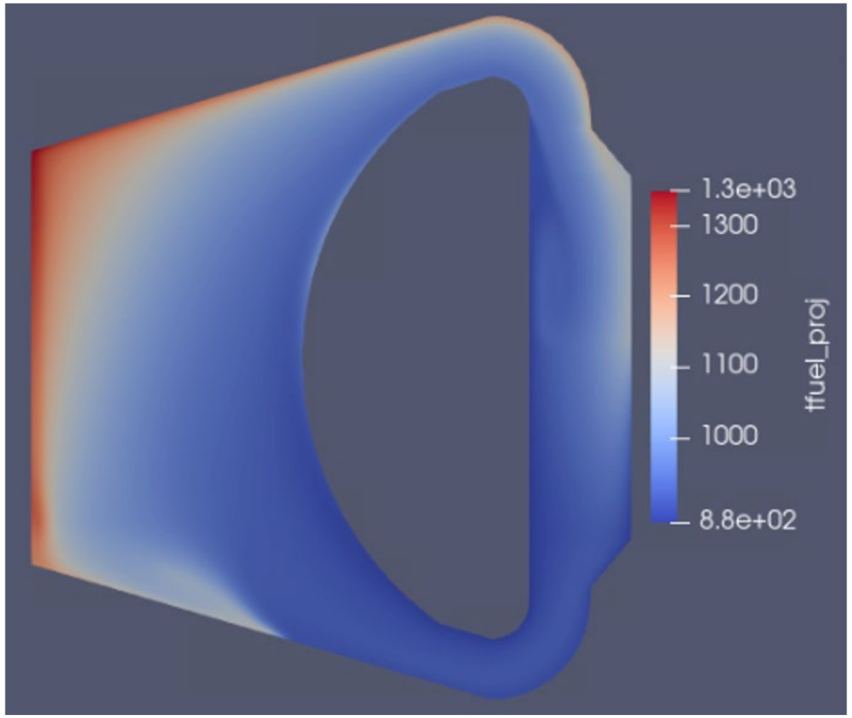

Figure: Multiphysics (Griffin + Pronghorn) steady-state Temperature [K] solution from VTB

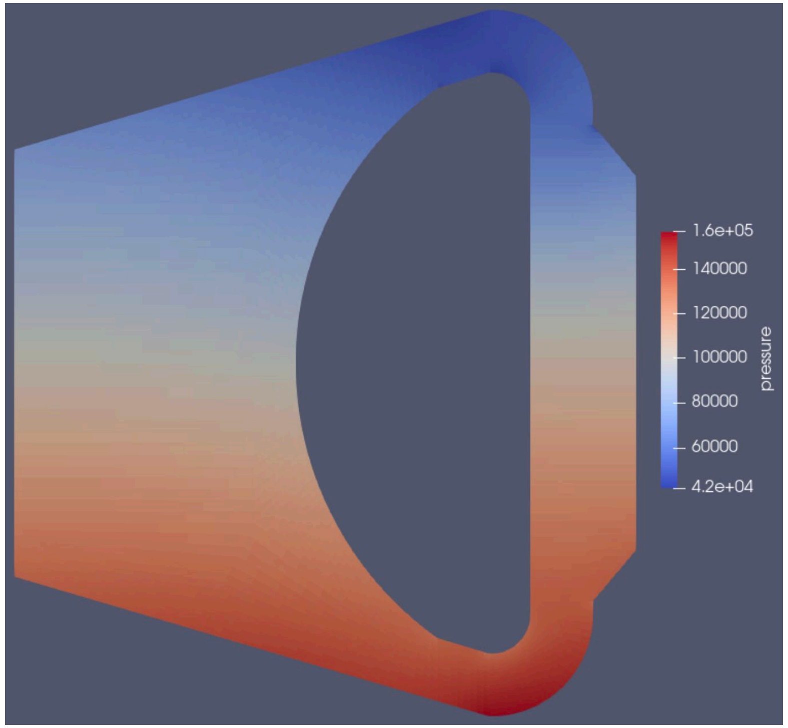

Figure: Multiphysics (Griffin + Pronghorn) steady-state Pressure [Pa] solution from VTB

https://mooseframework.inl.gov/virtual_test_bed/

Study 2: Spatially Resolved Redox Potential Control Transient for the Molten Salt Fast Reactor

- Redox control via reducing metal addition.

  - Adding in Beryllium at lower plenum (mixing into system)

  - Reducing Effect on Fluorine potential

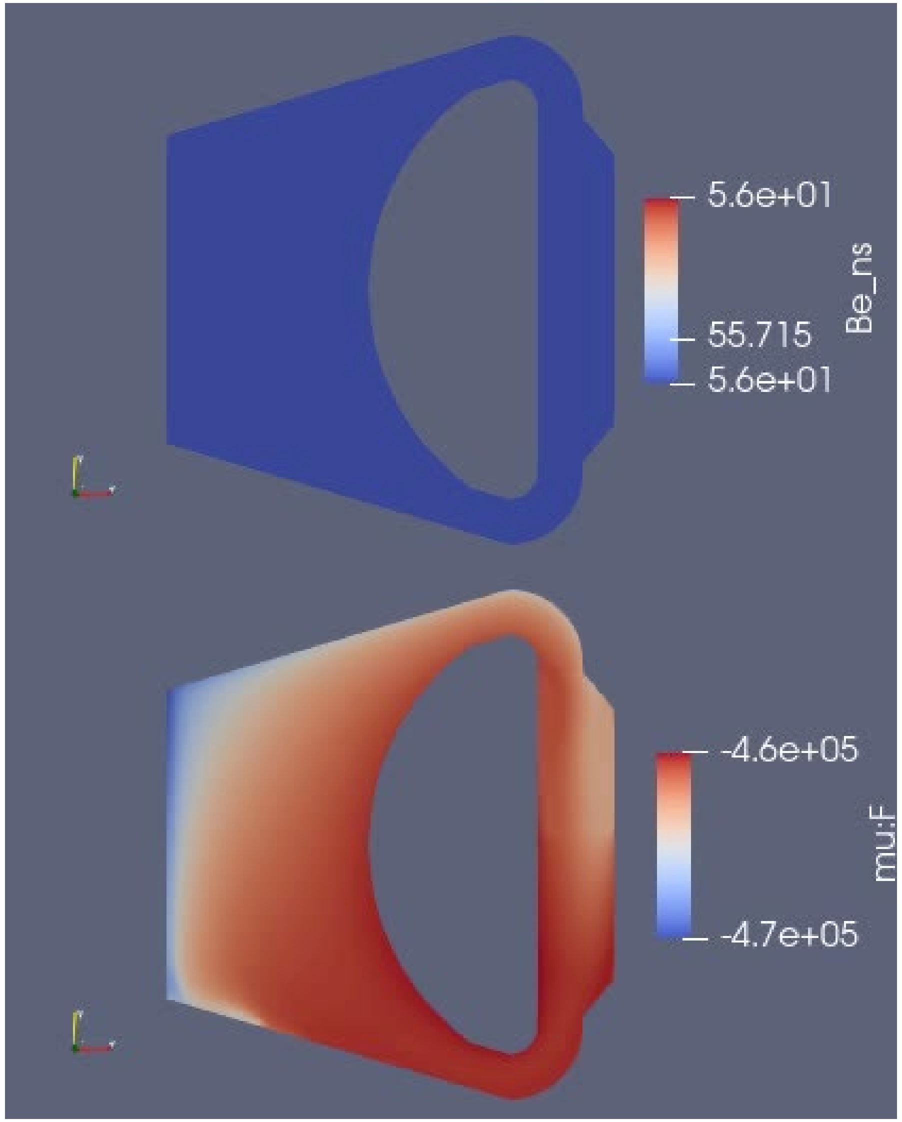

Figure: (top) Beryllium addition at lower plenum and mixing in MSFR. (bottom) Reducing effect on decreasing the fluorine potential.

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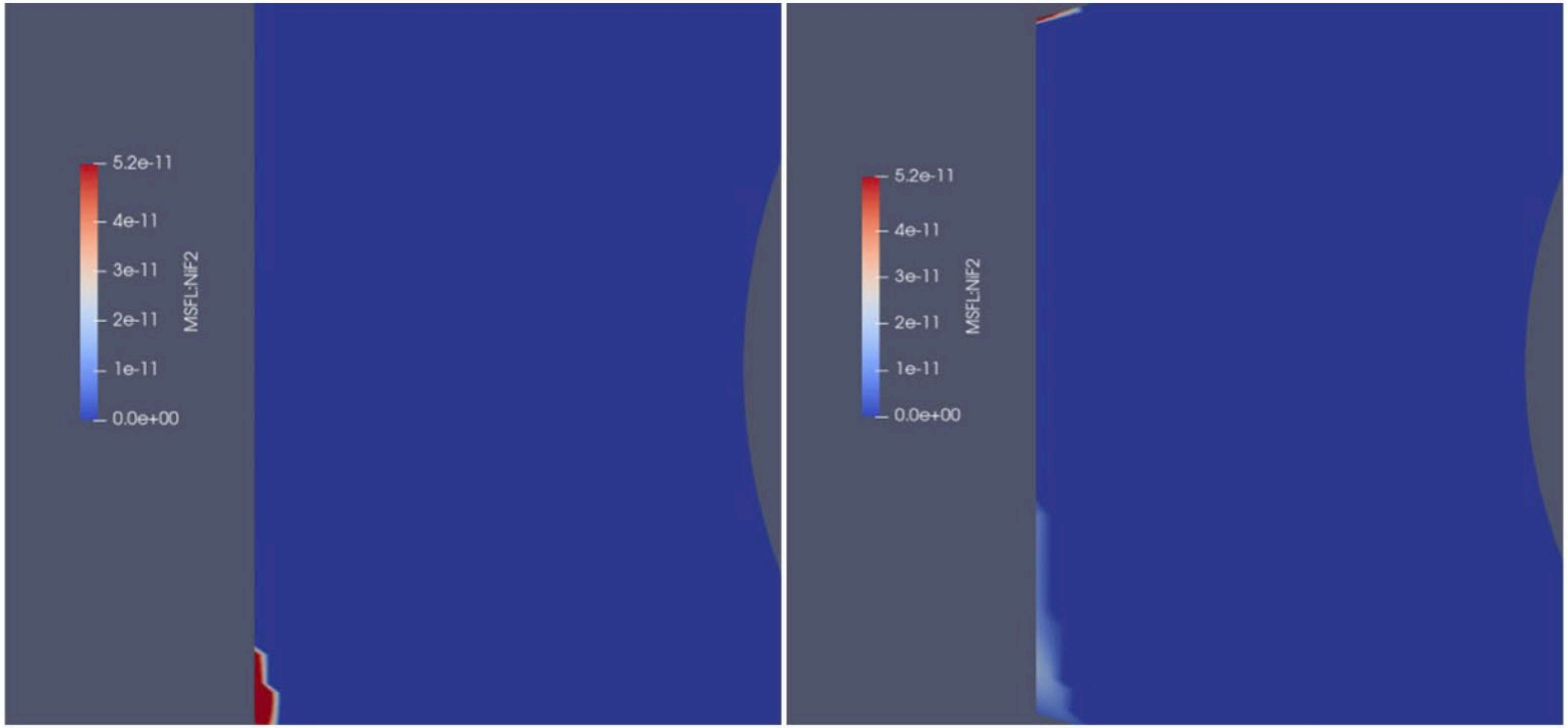
Study 2: Effect of NiF_2 reduction due to Beryllium Addition

- As additional Beryllium is dissolved into fuel-salt the presence of corrosion products (e.g., NiF_2) is reduced due to reduction.



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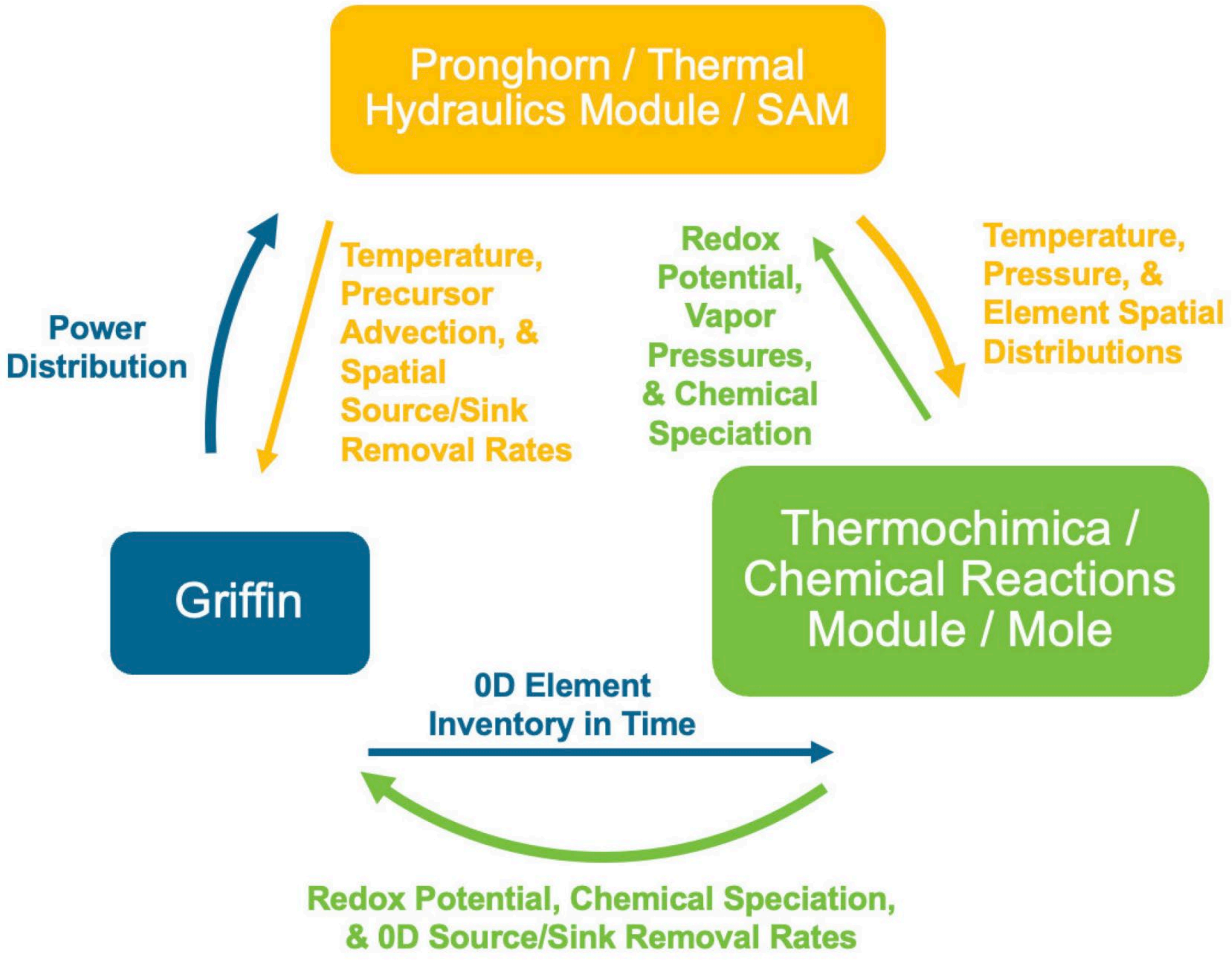
Depletion-driven Spatially Resolved Thermochemistry

- Griffin + Pronghorn calculate multiphysics steady state solution of fission source, temperature, and pressure.
- Griffin + Pronghorn depletion calculates 0D nuclide concentration by incorporating effect of nuclide flow on reaction rates.
- Pronghorn species tracking incorporates explicitly tracked nuclides & homogenizes 0D calculations from Griffin depletion to determine elemental distributions.
- Thermochimica given the elemental distributions calculates speciation and source and sink terms for Pronghorn species tracking.



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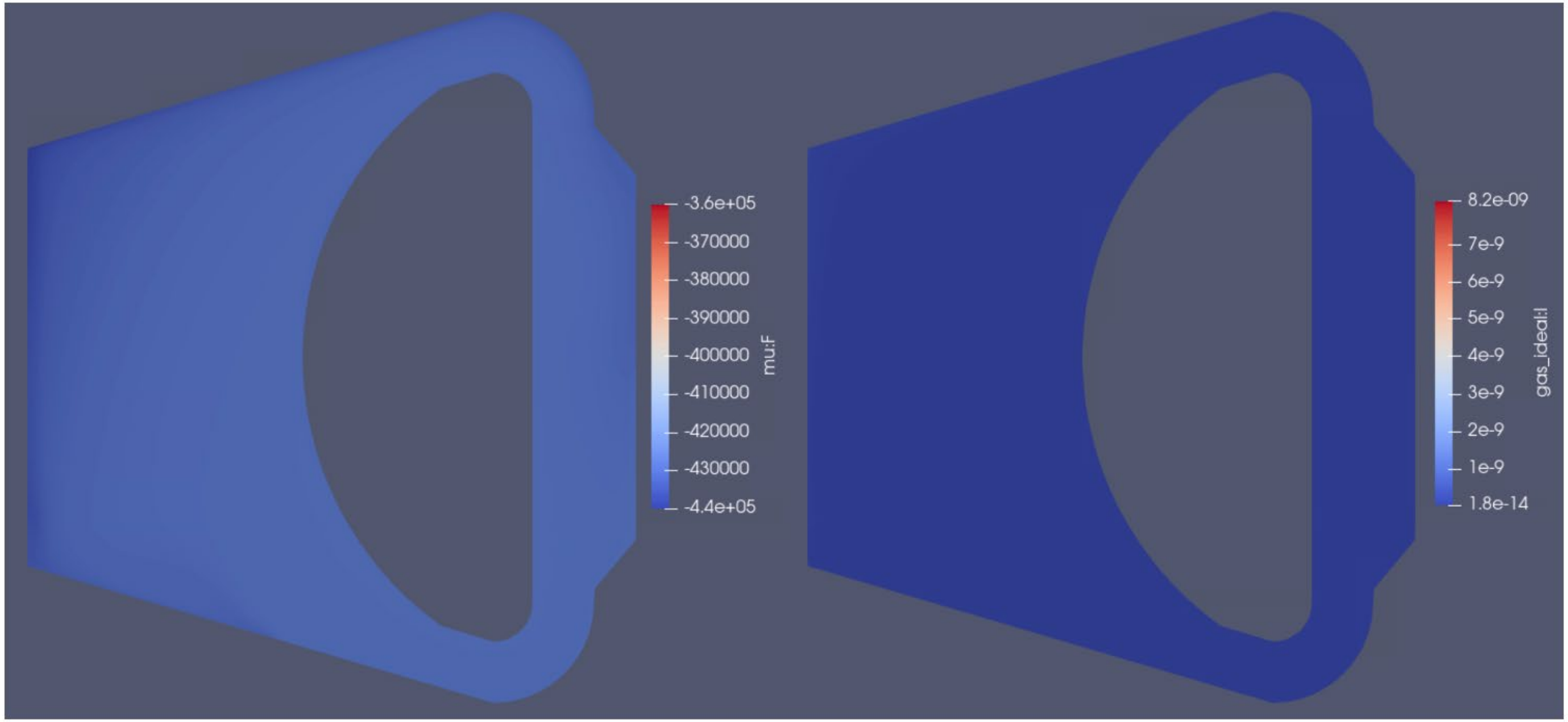
Study 3: Depletion affecting chemical species volatilization



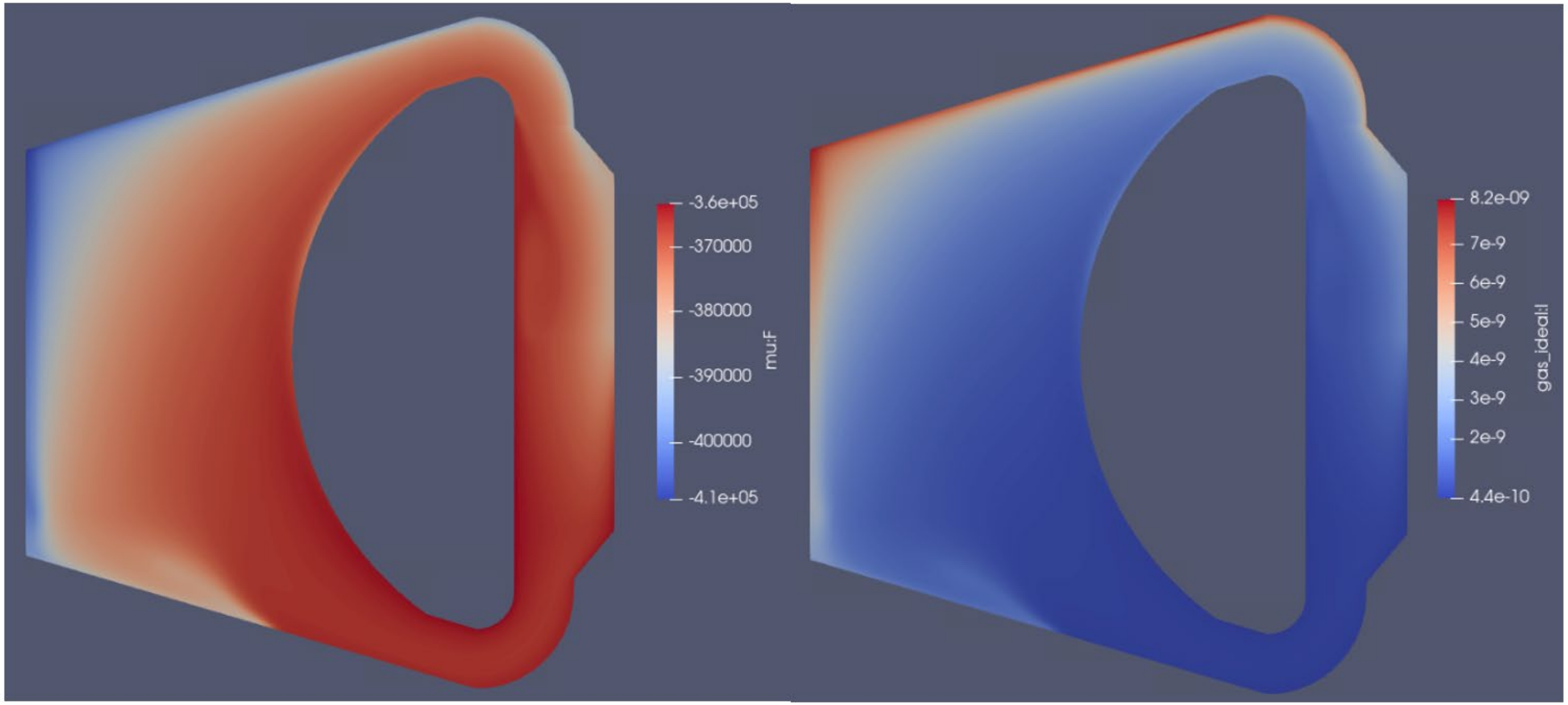
- Result: Iodine and Cesium are extremely soluble in fluoride-based fuel salt.

Figure: (left) Fluoride (F-) potential [J/mol] and corresponding iodine in stable ideal gas phase (right) [mol] with fresh fuel salt – i.e., reducing.

Walker et. al (2023) INL/RPT-23-74376

Walker et. al (2023) INL/RPT-23-74376

Study 3: Depletion effecting chemical species volatilization



- Result: Iodine and Cesium begin to volatilize as Fluorine potential increases due to depletion.
- Informs source term calculations during normal operation and accident scenarios.

Figure: (left) Fluoride (F-) potential [J/mol] and corresponding iodine in stable ideal gas phase (right) [mol] at 2.07 MWd/Kg-U burnup without chemistry control – i.e., oxidizing.

Walker et. al (2023) INL/RPT-23-74376

Study 3: Depletion affecting chemical species volatilization

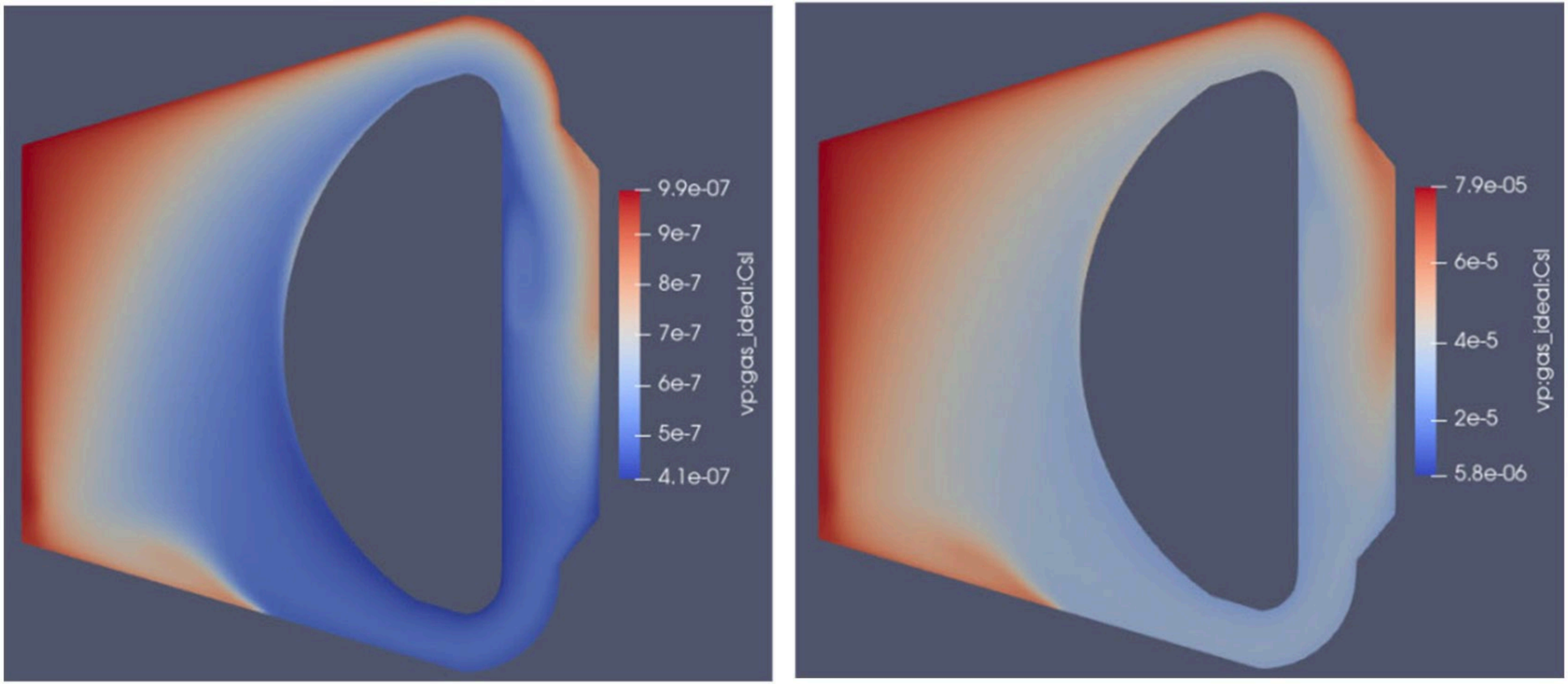


Figure: (left) CsI vapor pressure [atm] at beginning of life and (right) at 2.07 MWd/Kg-U burnup without chemistry control – i.e., oxidizing.

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Study 3: Depletion affecting chemical species volatilization

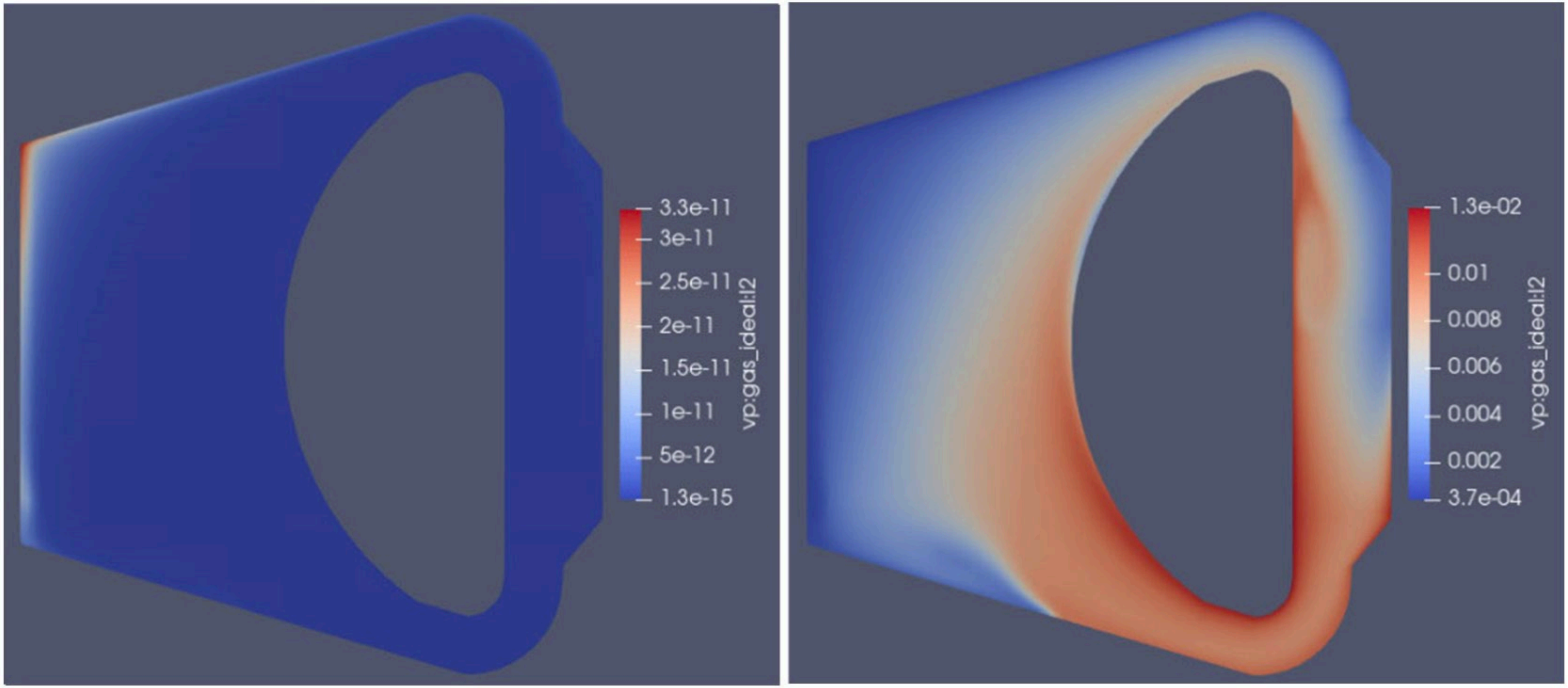

Figure: (left) I_2 vapor pressure [atm] at beginning of life and (right) at 2.07 MWd/Kg-U burnup without chemistry control – i.e., oxidizing. Most sensitive to redox potential change more than CsI.

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Study 3: Depletion affecting corrosion

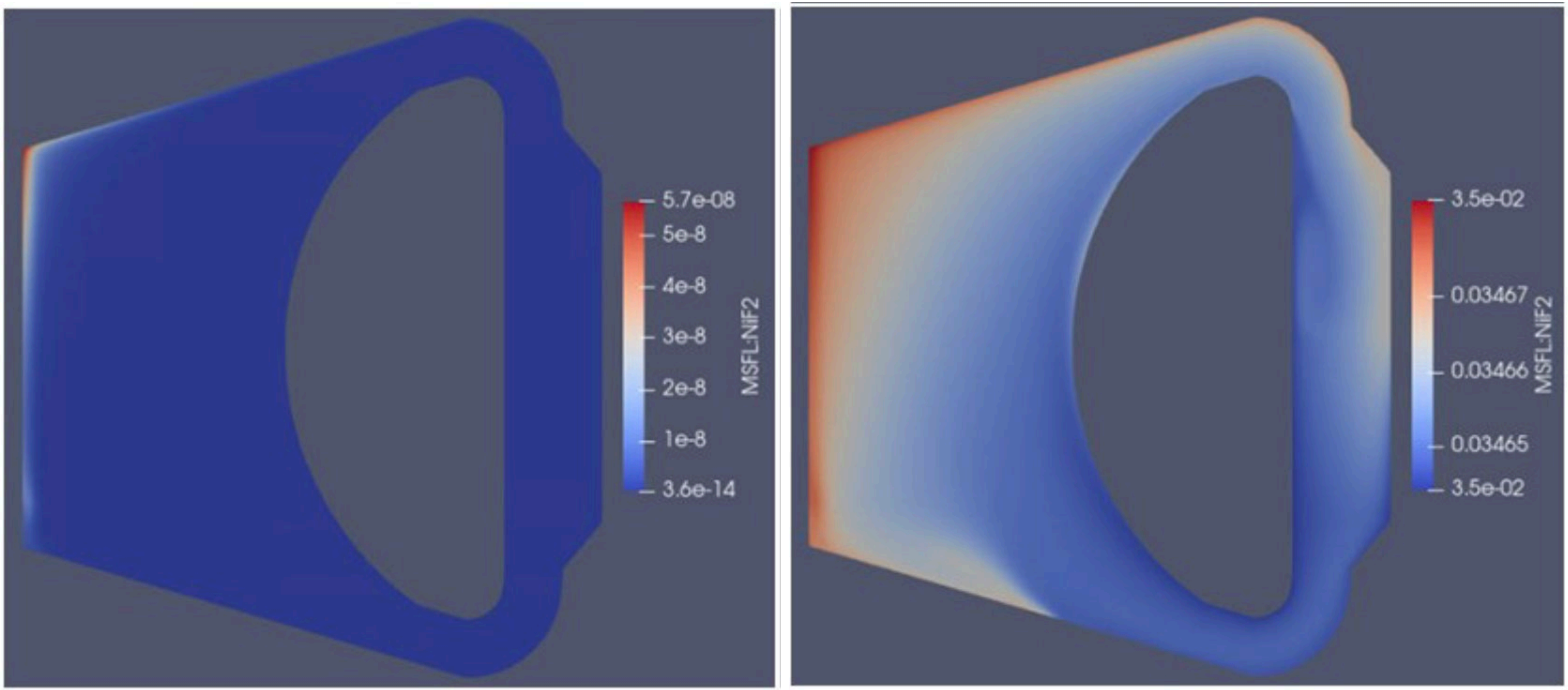
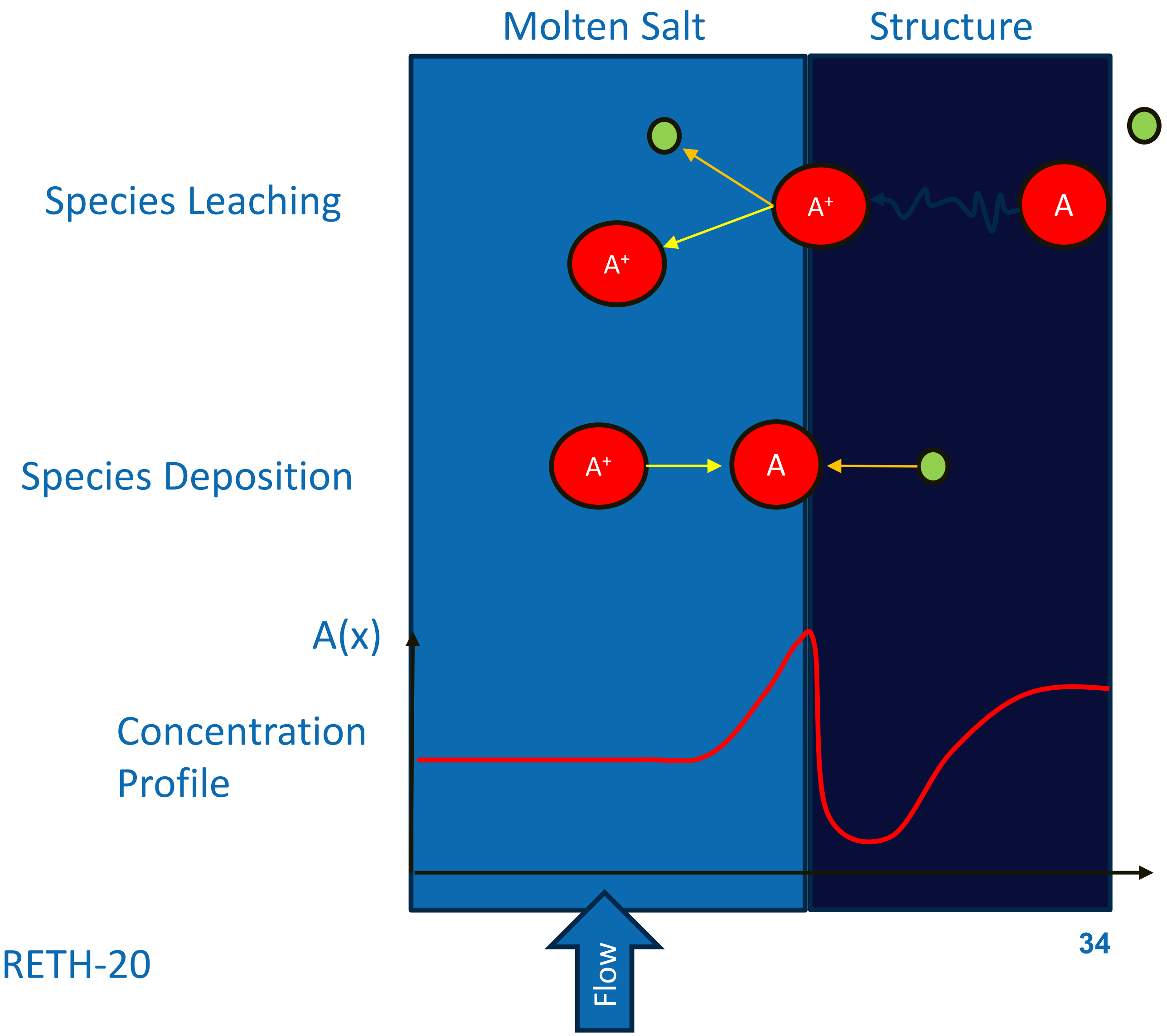


Figure: (left) NiF_2 concentration [mols] at beginning of life and (right) at 2.07 MWd/Kg-U burnup without chemistry control – i.e., oxidizing. Corrosion products greatly increasing and mixing into reactor.

Study 4: Corrosion Kinetics via Poisson-Nernst-Planck

- During species leaching, an atom of structure constitutive element A migrates to the surface, where it loses an electron an is released to the salt
- During species deposition, a cation gets and electron and deposits in the solid structure
- The rate at which this process happens in dependent of the concentration profile of the species and temperature



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Study 4: Corrosion Kinetics via Poisson-Nernst-Planck

- The PNP model solves a coupled mechanism between species diffusion and development of the electrochemical potential:

$$\frac{\partial c_i}{\partial t} + \nabla\cdot(\boldsymbol{u}c_i) - \nabla\cdot(D_i\nabla c_i) - \frac{FZ_i}{RT}\nabla\cdot(D_i c_i \nabla\phi) = 0$$

$$\nabla\cdot(\epsilon_r\epsilon_0\nabla\phi) - \frac{F}{\epsilon_r\epsilon_0}\sum_i z_i c_i = 0$$

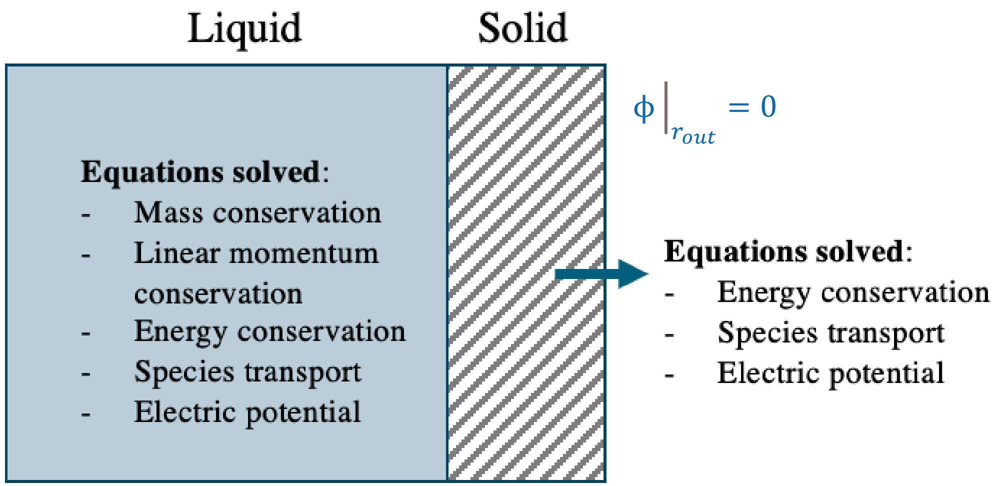
- The interface kinetics must be added into this model, which can be modeled via the Butler-Volmer equation:

$$j_{r\to o,i} = k_0 c_r^{\alpha} c_o^{1-\alpha}\left[\frac{c_{r,i}}{c_{r,b}} e^{-\frac{\Delta G_r}{RT}_i} e^{\frac{(1-\alpha)ZF}{RT_i}(E_{ro}-\phi_i)} + \frac{c_{o,i}}{c_{o,b}} e^{-\frac{\Delta G_o}{RT}_i} e^{\frac{(1-\alpha)ZF}{RT_i}(E_{ro}-\phi_i)}\right]$$

- The effective electric potential can be modeled via the Nernst equation:

$$E_{ro} = E_{ro}^{*} + \frac{RT}{ZF}\ln\left(\frac{c_{r,i}}{c_{o,i}}\right)$$

Model 2



Reaction Modeled:

$$F_2Fe + Cr \to F_2Cr + Fe,$$
$$F_2Ni + Cr \to F_2Cr + Ni,$$
$$F_2Ni + Fe \to F_2Fe + Ni,$$
$$2LiF + Cr \to F_2Cr + 2Li,$$
$$2LiF + Fe \to F_2Fe + 2Li,$$
$$2LiF + Ni \to F_2Ni + 2Li,$$
$$2NaF + Cr \to F_2Cr + 2Na,$$
$$2NaF + Fe \to F_2Fe + 2Na,$$
$$2NaF + Ni \to F_2Ni + 2Na,$$
$$2KF + Cr \to F_2Cr + 2K,$$
$$2KF + Fe \to F_2Fe + 2K,$$
$$2KF + Ni \to F_2Ni + 2K,$$
$$2FH + Cr \to F_2Cr + H_2,$$
$$2FH + Fe \to F_2Fr + H_2,$$
$$2FH + Ni \to F_2Ni + H_2$$

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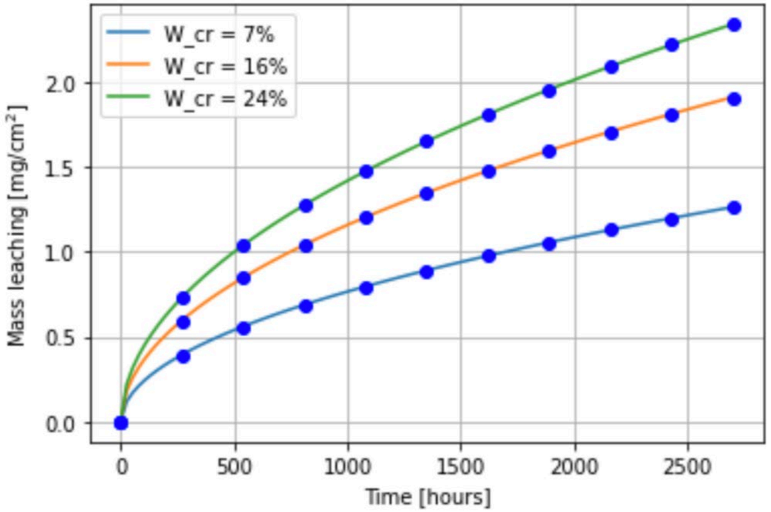
Study 4: Model Verification

Species Leaching: Leaching of Cr alloy to an infinite sink



<u>Analytical Model</u>:

$$D_{leach,Cr} = \frac{k_0 W_{Cr}}{7} e^{-Q/T}$$

$$j_{Cr} = \left(D_{leach,Cr} t\right)^{1/2}$$

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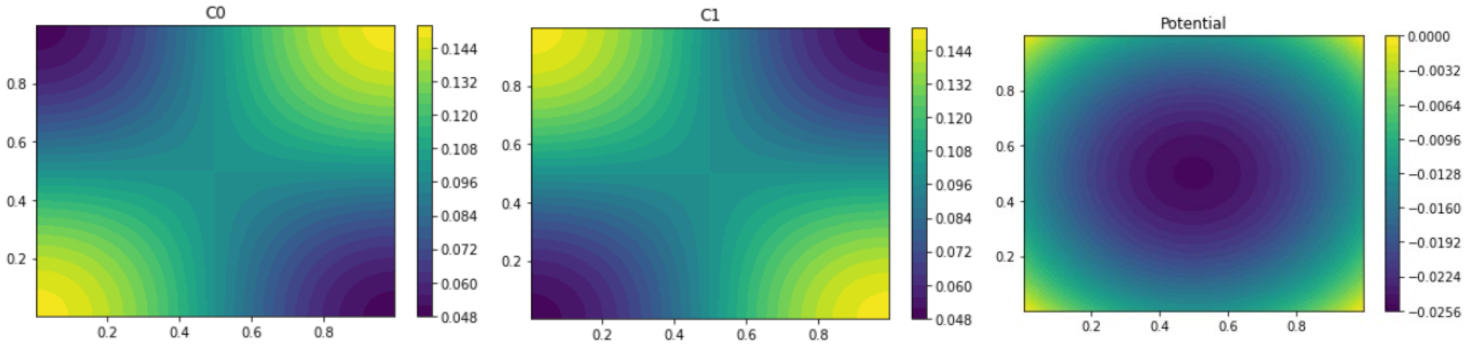
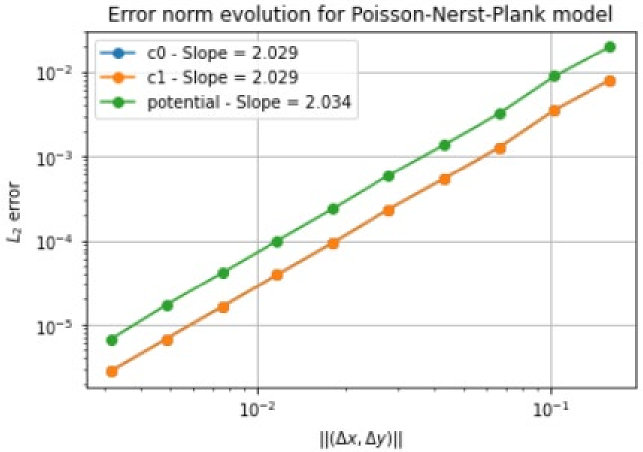
PNP: Manufactured solution

$$c_0(x,y) = \tau + \frac{\tau}{2}\cos(\pi x)\cos(\pi y)\,,$$

$$c_1(x,y) = \tau - \frac{\tau}{2}\cos(\pi x)\cos(\pi y)$$

$$\phi = -\frac{\tau F}{2\epsilon}\left[(1-x)x + (1-y)y\right]$$





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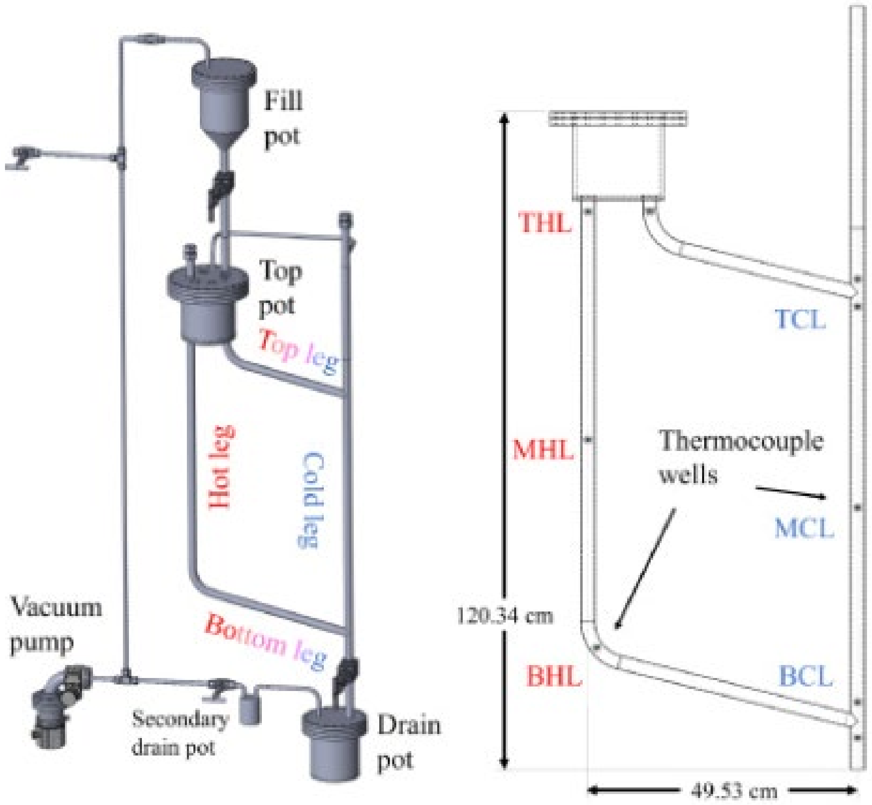
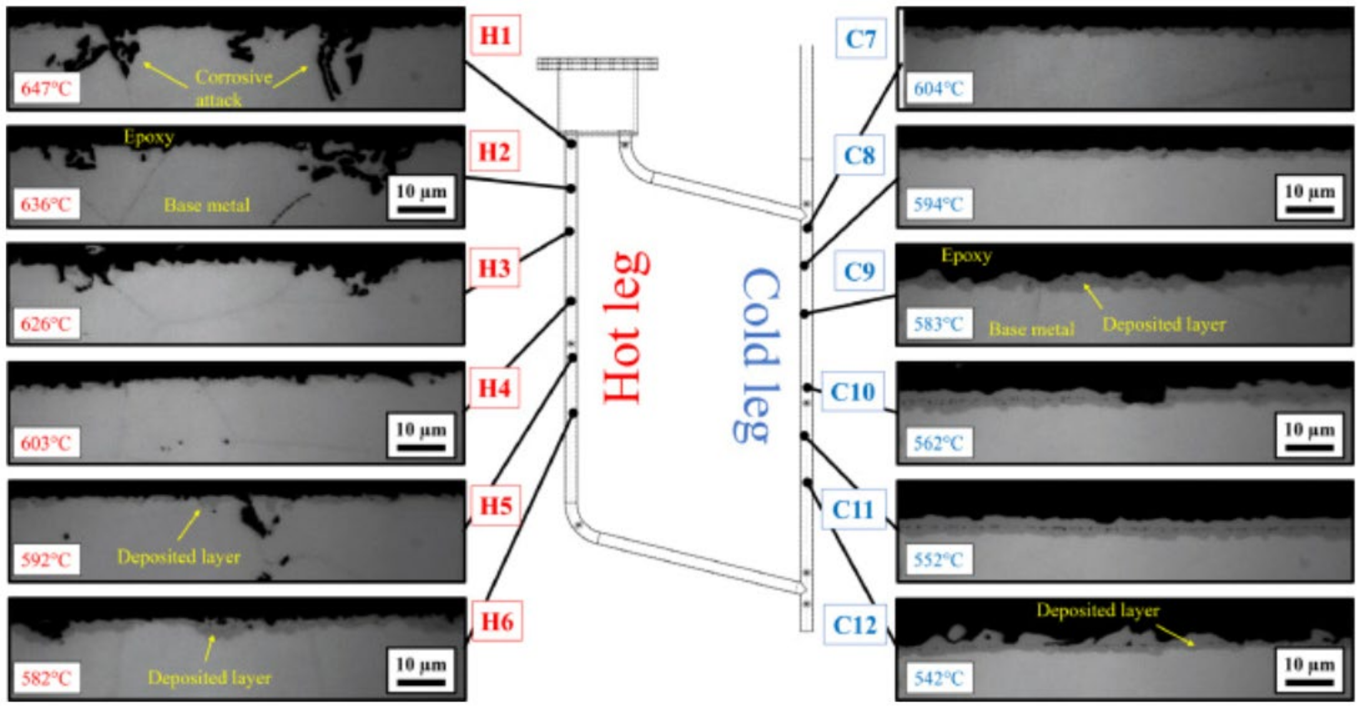
Study 4: Validation - Molten Salt Loop Experiment





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.

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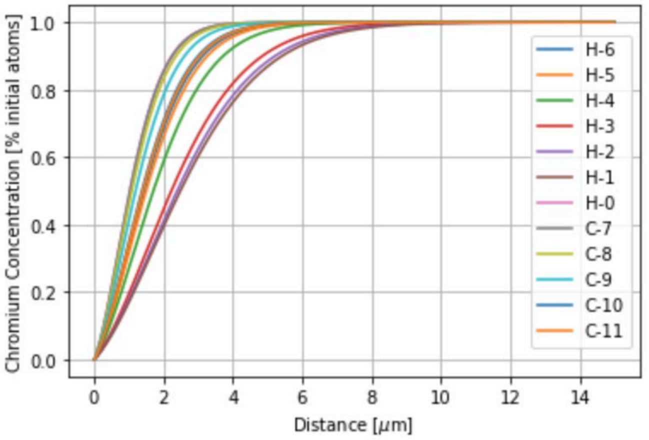
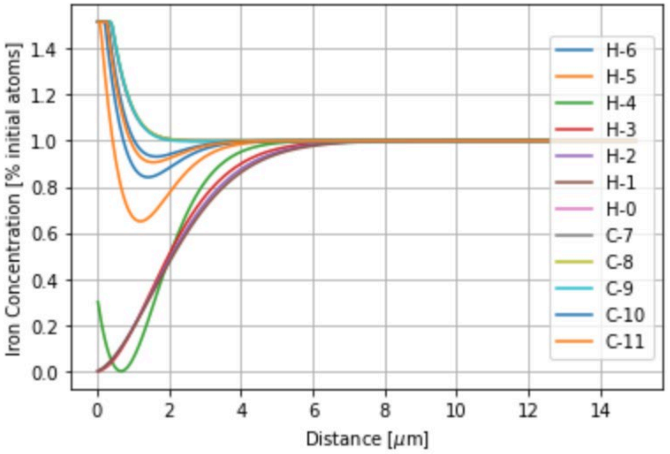
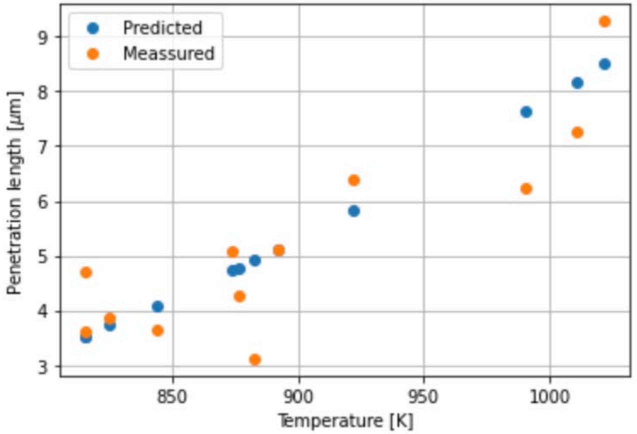
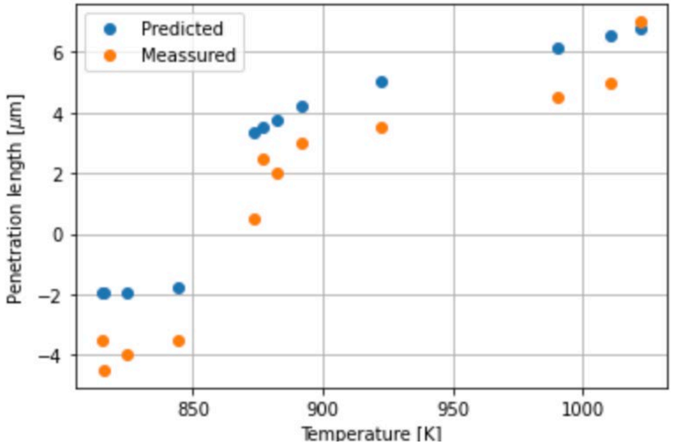
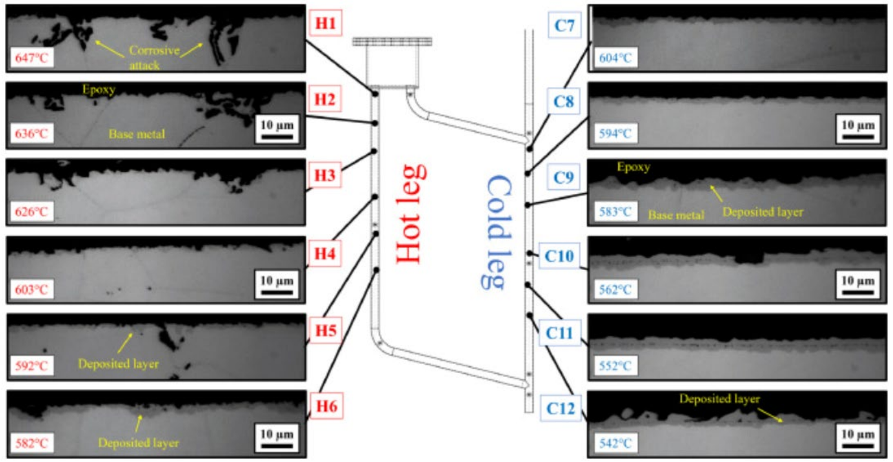
Study 4: Validation - PNP Model











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

Remaining amounts in the salt

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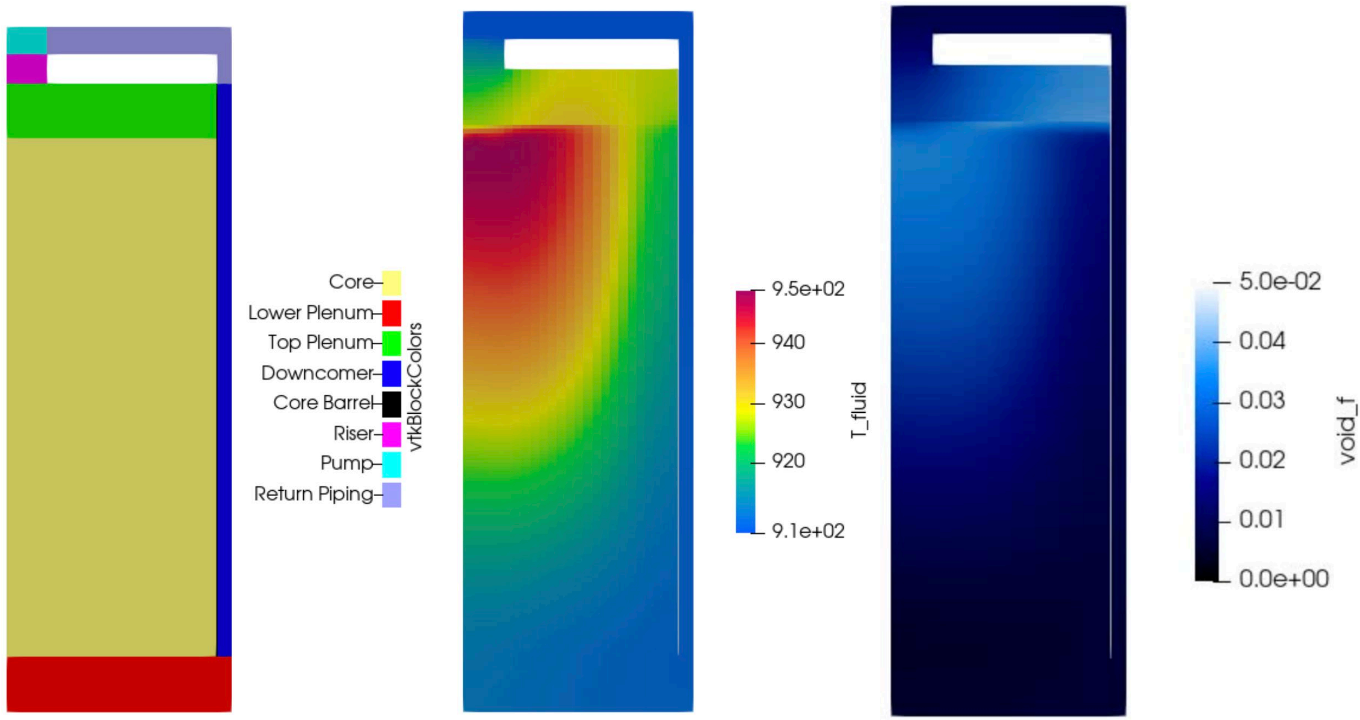
Ongoing Work: Two-Phase Flow for MSRE Analysis

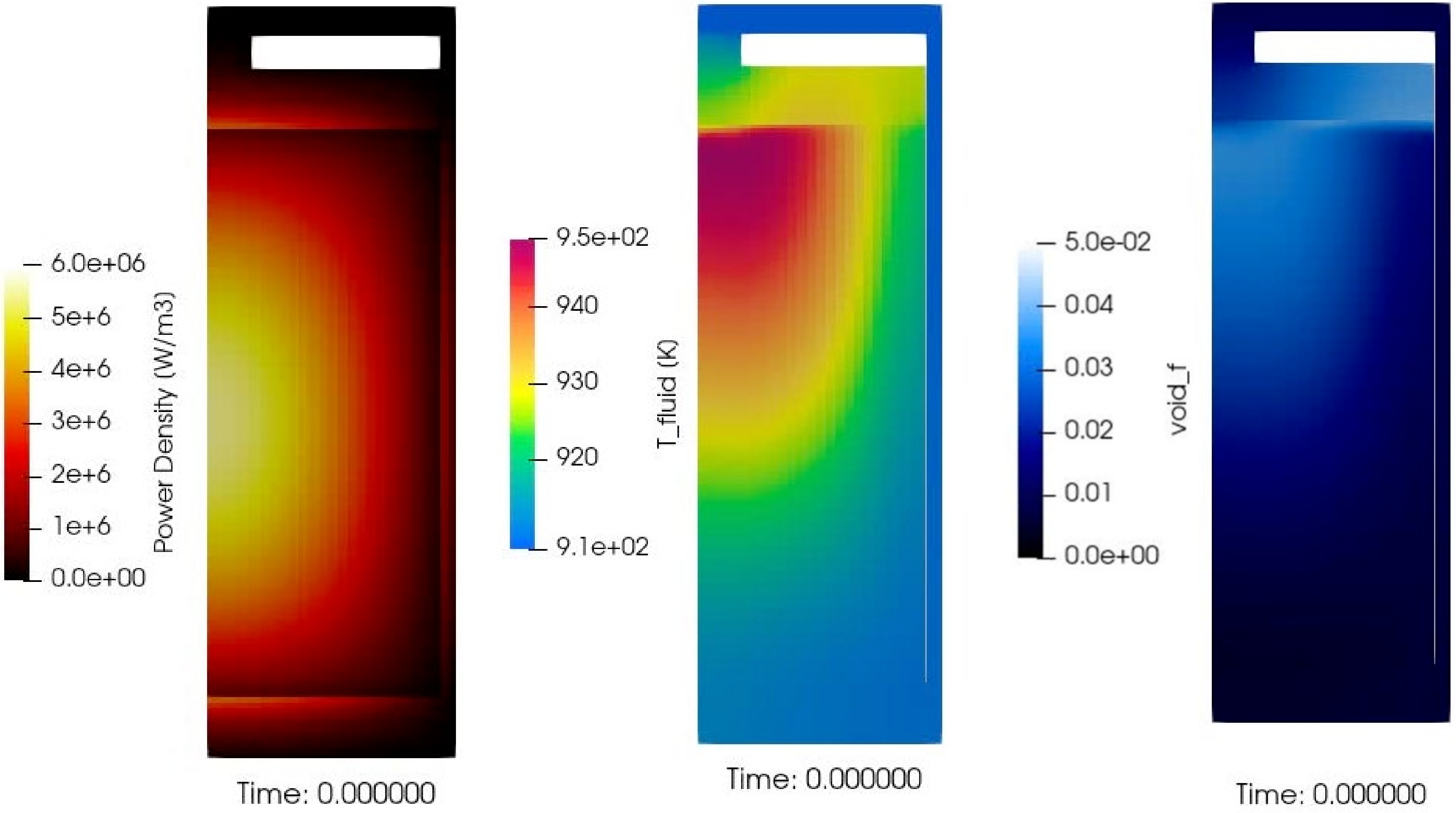
- 5-10% of fission products are gaseous in the fuel salt
- During operation of MSRs, gaseous fission products will result in power shifts, changes in the velocity and temperature fields, and non-uniform species transport
- Additionally, void fraction distribution plays an important role during reactor transients



Example: Molten Salt Reactor Experiment Axisymmetric Model

Ongoing Work: Two-Phase Flow for MSRE Analysis

- 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



Ongoing Work: Advanced Reactor International Safeguards Engagement (ARISE) Project on Multiphysics Chemical Species Forensics

- Perform safeguard analyses using new multiphysics framework.
- Perturb Multiphysics models within normal reactor operational parameters using Stochastic Tools Module of MOOSE.
- Feed Large Perturbed Data Sets to Machine Learning Algorithm to define "Normal operation Space" and "Diversion or misuse operation space"

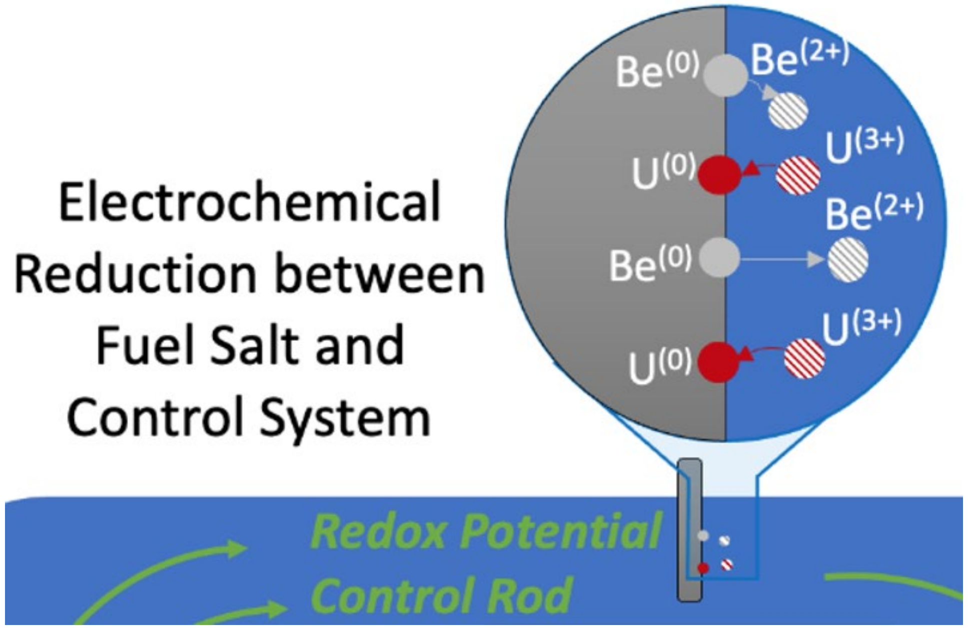


Figure: Eventual electrochemical plating of Uranium via over-reducing fuel salt with beryllium redox potential control rod.

Future Work: Liquid Breeding Blanket Tritium Modeling

- Breeding Material Depletion (Tritium Generation from ^6Li)
  - Griffin Depletion
- Depletion-Driven Thermochemistry of Molten Salt Breeding Blanket (Corrosion and speciation of Tritium)
  - Thermochimica
- Tritium Diffusion, Advection, Mass Transfer, and Extraction in liquid (gas sparging)
  - Pronghorn (a.k.a. Navier-Stokes Module in MOOSE)
- Tritium Interactions on Interfaces and Diffusion Through Materials
  - TMAP8

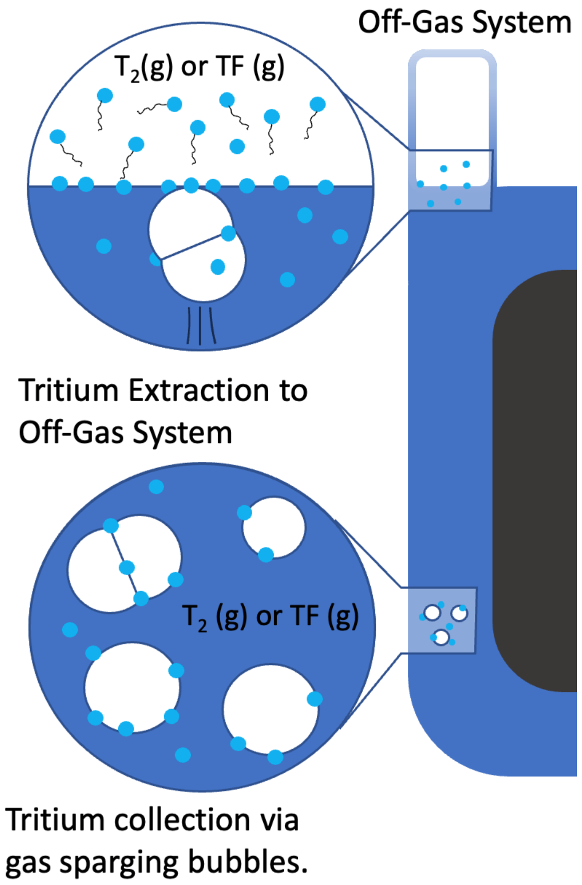


Figure: Tritium extraction via gas sparging system

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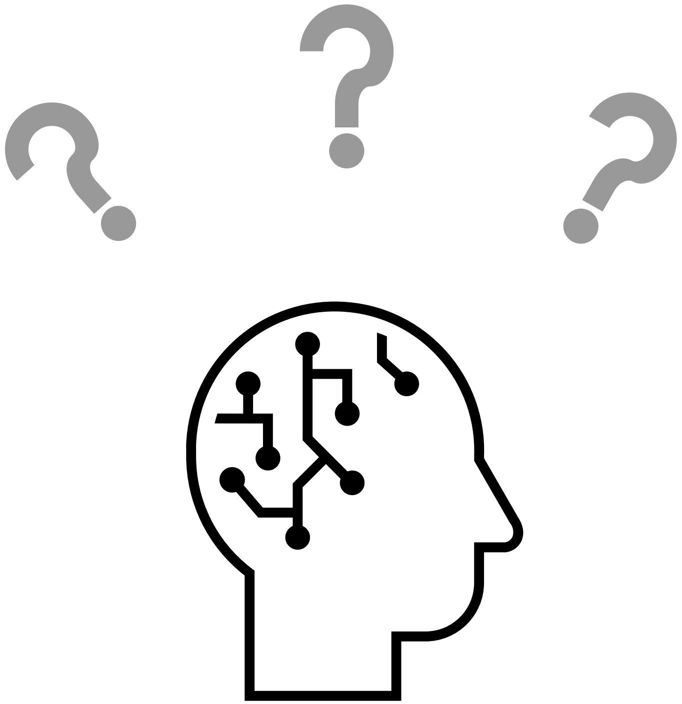
- Collaborators: **Mauricio Tano**, Parikshit Bajpai, Abdalla Abou-Jaoude, Rodrigo de Oliveira, Ryan Stewart, Odera Dim (Brookhaven National Laboratory), Thomas Fuerst and Olin Calvin.

Thank you for your attention – Questions?



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Upcoming Webinars

Date	Title	Presenter
22 May 2024	Joint GIF/IAEA Webinar: Regulatory Activities in support of SMRs and Advanced Reactor Systems	Paula Calle Vives, IAEA Greg Oberson, NRC, USA Tarek Tabikh, CNSC-CCNS, Canada
05 June 2024	Directed Energy Deposition Process of Corrosion Resistant Coating for Lead-Bismuth Eutectic Environment	Gidong Kim, UNIST, Korea
31 July 2024	Online monitoring development in support of the nuclear fuel cycle	Amanda Lines and Sam Bryan, PNNL, USA