Molten Salt Reactor Mechanistic Source Term

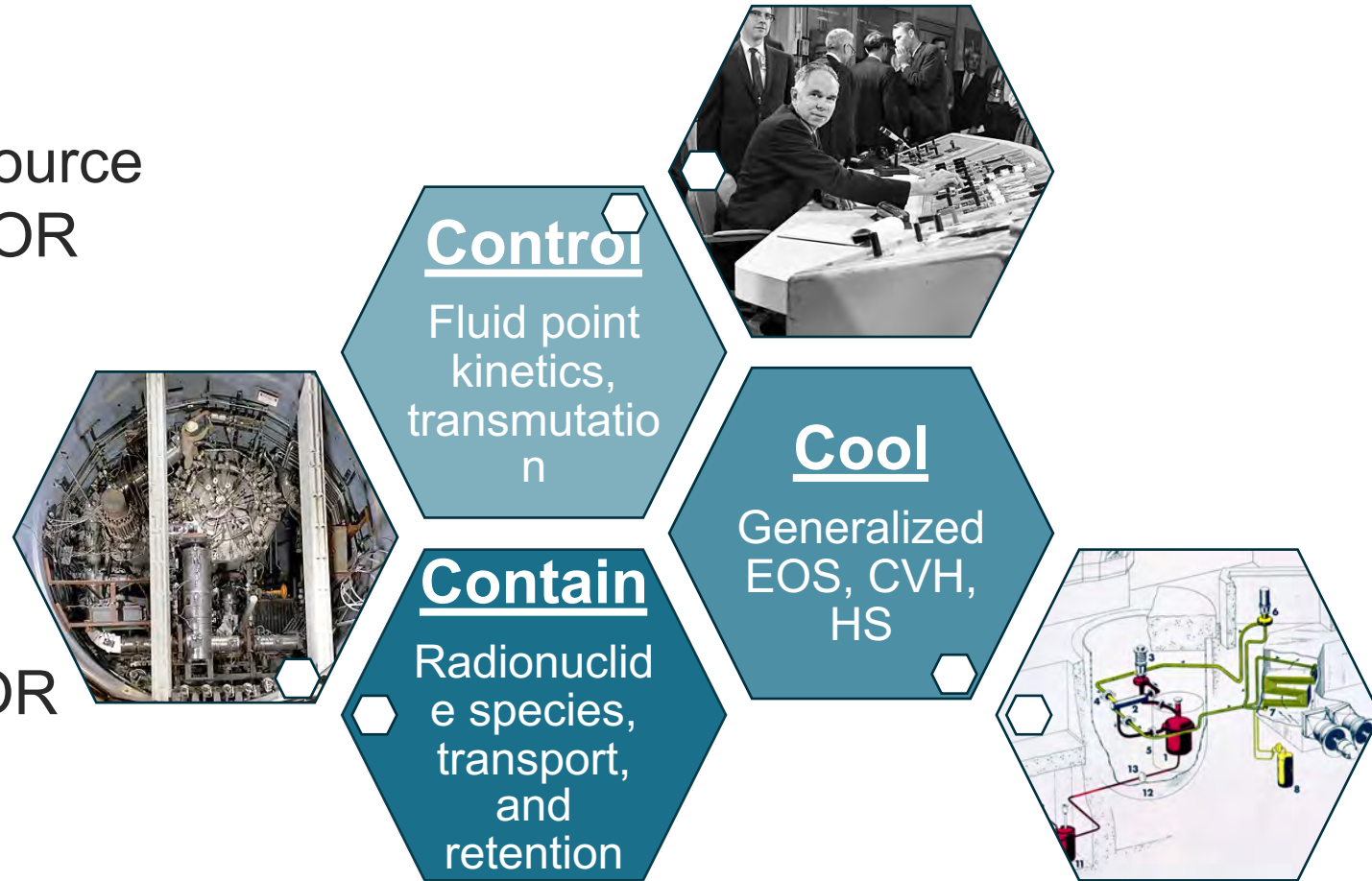**Lucas I. Albright, Troy Haskin, Rod Schmidt, David L. Luxat**

Sandia National Laboratories

SAND2023-0311PE

Annual MSR Campaign Review Meeting 2-4 May 2023

Molten Salt Reactor Modeling

- Molten Salt Reactor mechanistic source term modeling strategies in MELCOR boil down to 3 primary concepts:
  1. Control
  2. Cool
  3. Contain
- MELCOR capabilities have been primarily tested using the MELCOR model of the MSRE
  - Liquid Salt Test Loop model (under development) will be added to the MELCOR validation matrix as data becomes available

Control
Fluid point kinetics, transmutation

Cool
Generalized EOS, CVH, HS

Contain
Radionuclide species, transport, and retention

Images from https://en.wikipedia.org/wiki/Molten-Salt_Reactor_Experiment

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Control

- Fuel point kinetics – Derived from standard PRKEs and solved similarly
  - Feedback models
    - User-specified external input
    - Doppler
    - Fuel and moderator density
    - Flow reactivity feedback effects integrated into the equation set

$$\frac{dP(t)}{dt} = \left(\frac{\rho(t) - \beta_{eff}}{\Lambda}\right) P(t) + \sum_{i=1}^{6} \lambda_i C_i^C(t) + S_0$$

$$\frac{dC_i^C(t)}{dt} = \left(\frac{\beta_i}{\Lambda}\right) P(t) - \left(\lambda_i + \frac{2}{\tau_C}\right) C_i^C(t) + \left(\frac{V_L}{V_C}\right)\left(\lambda_i + \frac{2}{\tau_L}\right) C_i^L(t),$$
$$i = 1 \dots 6$$

$$\frac{dC_i^L(t)}{dt} = \left(\frac{V_C}{\tau_C V_L}\right) C_i^C(t) - \left(\lambda_i + \frac{1}{\tau_L}\right) C_i^L(t),$$
$$i = 1 \dots 6$$

Compensating Control System Reactivity [pcm] vs Time [s] — Guo Code, MSRE Data, MELCOR

- Transmutation – Depletion Module (under development)
  - Treats transmutation of the initial radionuclide inventory (radioactive decay and neutron interactions) during severe accidents including mass transfer between radionuclide classes

$$\frac{dN}{dt} = (A_\lambda + A_\phi \Phi) N(t) + S(t)$$

- $N(t)$: vector of isotope number densities
- A_λ: radioactive decay transition matrix
- A_ϕ: neutron interaction transition matrix
- Φ: scalar neutron flux
- $S(t)$: eternal isotope source

Cool
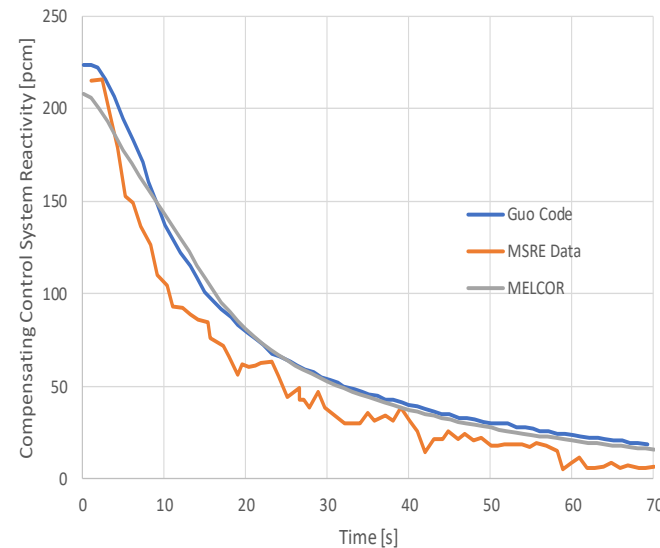
- Generalized EOS – Equations of state for multiple working fluids are presently available in MELCOR including water, sodium, and FLiBe.
  - MELEOS (under development) can generate equations of state for any fluid with the necessary data
- Thermal hydraulics – CVH/FL Packages
  - The CVH package defines control volumes (CV)
  - The FL package defines flow paths (FL)
- Heat Transfer – HS/CVH/COR Packages
  - The HS package defines heat structures (HS) that model radiative and conductive heat losses
  - The CVH package manages convective heat losses
  - The COR package controls heat losses of heat bearing and other core structures
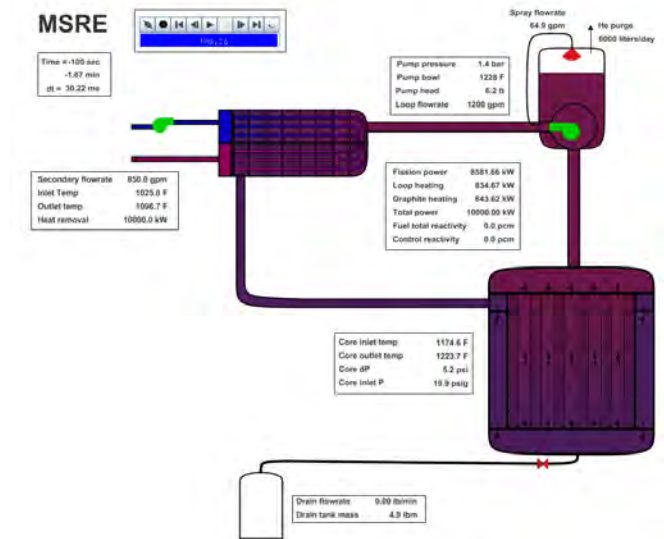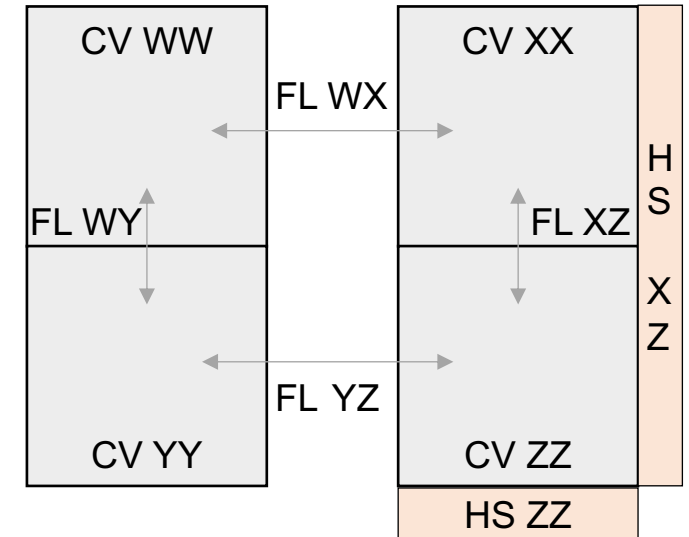
Contain

- Generalized Radionuclide Species – DCH/RN1
  - Default 17 representative radionuclide species/elements (RN classes)
  - Users have the ability to redefine/add RN classes
  - Radionuclide species are categories of chemically similar (similar in transport and retention mechanisms) radioactive elements/compounds
  - Each radionuclide species is representative of a set of elements and the corresponding collapsed isotopic masses
  - MELCOR does not currently model RN class masses at the isotopic level or mass transfer between RN classes
- Generalized Radionuclide Transport and Retention (GRTR)
  - MELCOR treats transport and retention of RN classes by various physiochemical processes in vapor and aerosol form by default; including aerosol size distribution (called "sections")
  - The GRTR model enhances these capabilities to allow for user-defined RN forms and subsequent formwise/sectionwise transfer between RN forms
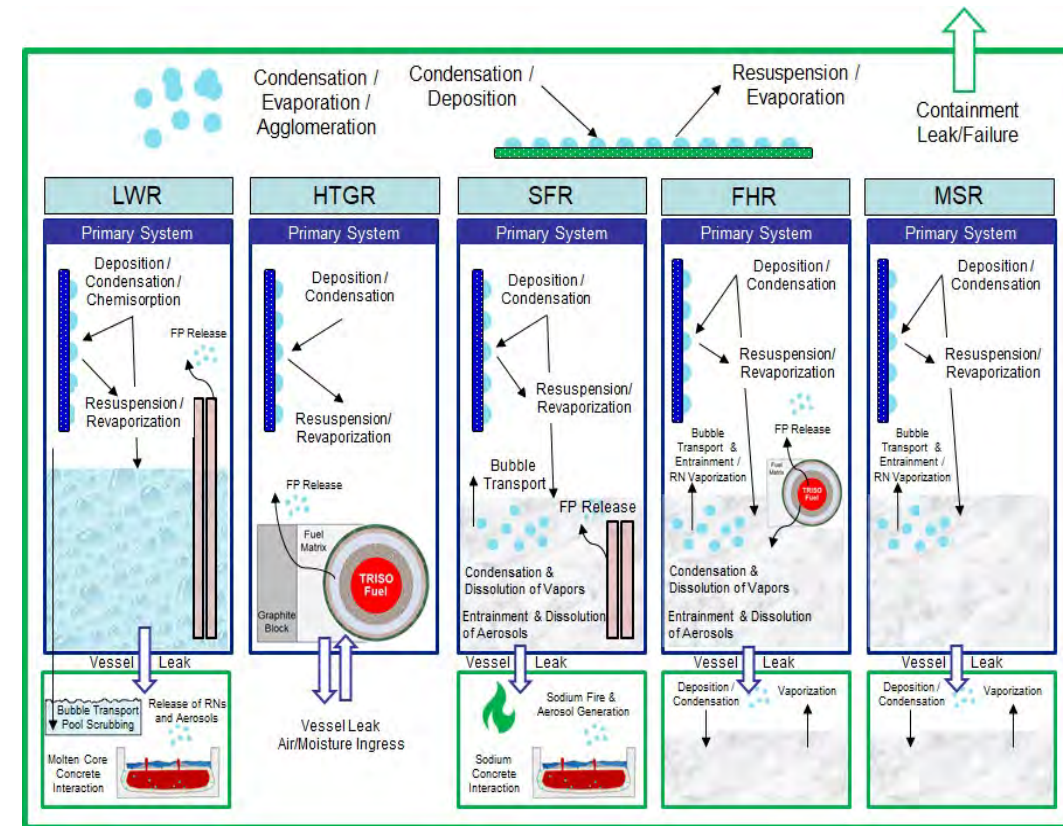
Liquid Salt Test Loop Model Development

Fig. 3. Picture of loop in hood.
Yoder, G.L. et al., *High-Temperature Fluoride Salt Test Loop*. Oak Ridge National Laboratory. ORNL/TM-2012/430, 2015.

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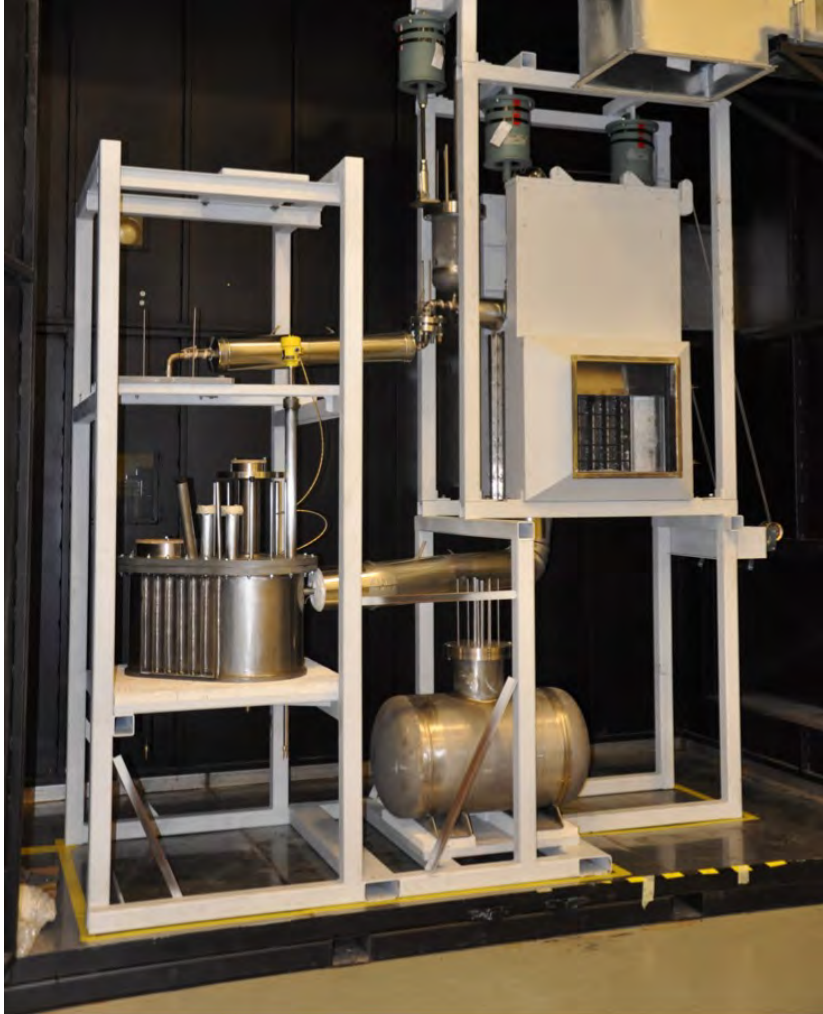# The ORNL High-Temperature Fluoride Salt Test Loop

Fig. 3. Picture of loop in hood.

- Test facility designed for measuring fluoride salt heat transfer inside a heated pebble bed similar to that used for the core of the pebble bed–advanced high-temperature reactor.

- Incorporates a centrifugal pump to circulate FLiNaK salt through a removable test section.

- Inductive heating used to apply heat to the test section.

- Supporting loop infrastructure includes a pressure control system; trace heating system; and complement of instrumentation to measure salt flow, temperatures, and pressures around the loop.

- Details of the experimental design provided in:

  Yoder, G.L. et al., *High-Temperature Fluoride Salt Test Loop.* Oak Ridge National Laboratory. ORNL/TM-2012/430, 2015.

- Consultation with current ORNL staff provided helpful additional understanding and data!

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Fig. 1. Salt loop CAD model.

From: *Avigni, Wysocki, and Yoder, "Liquid Salt Test Loop modeling using Trace", Annals of Nuclear Energy 106 (2017) 170-184*
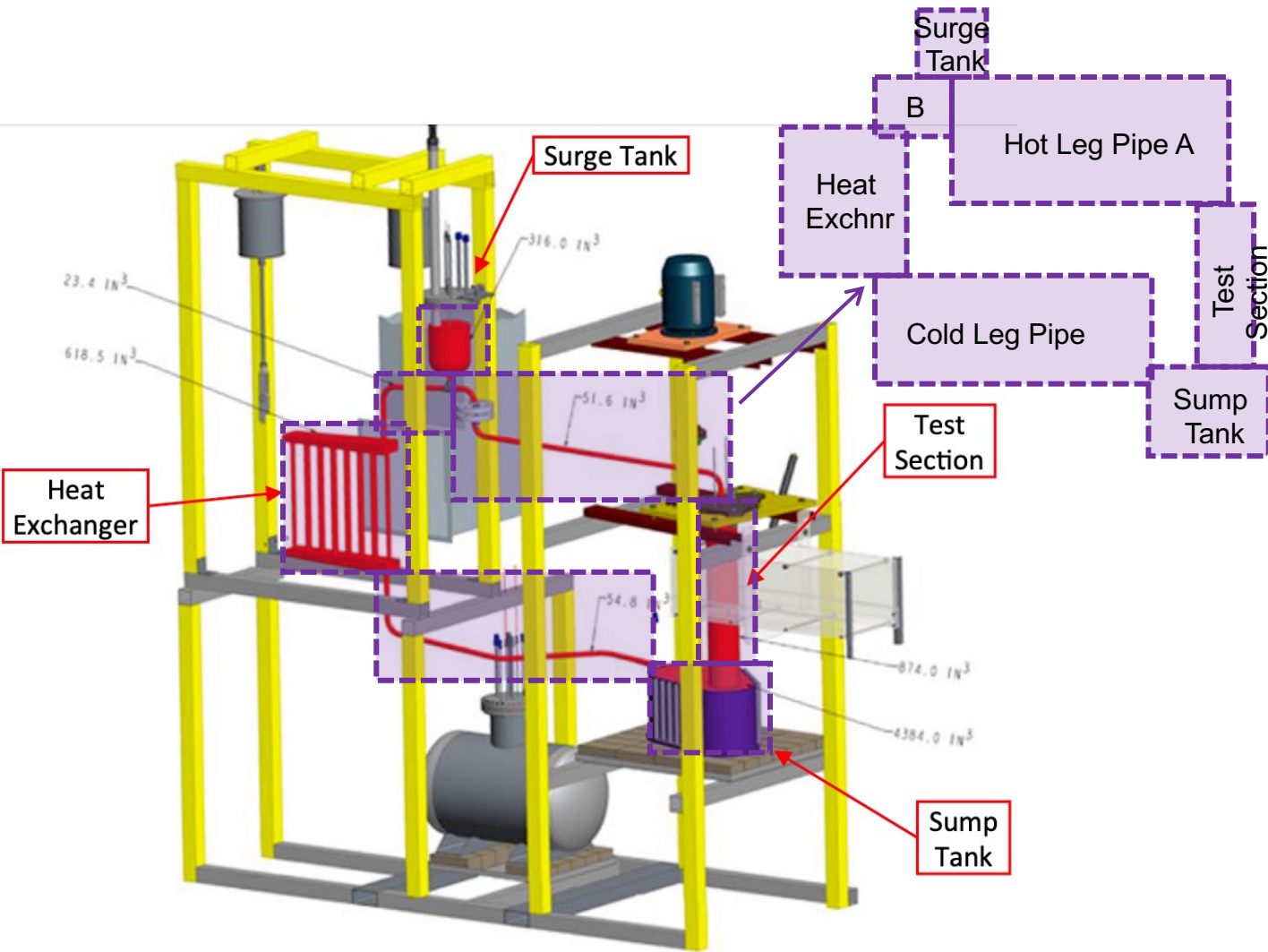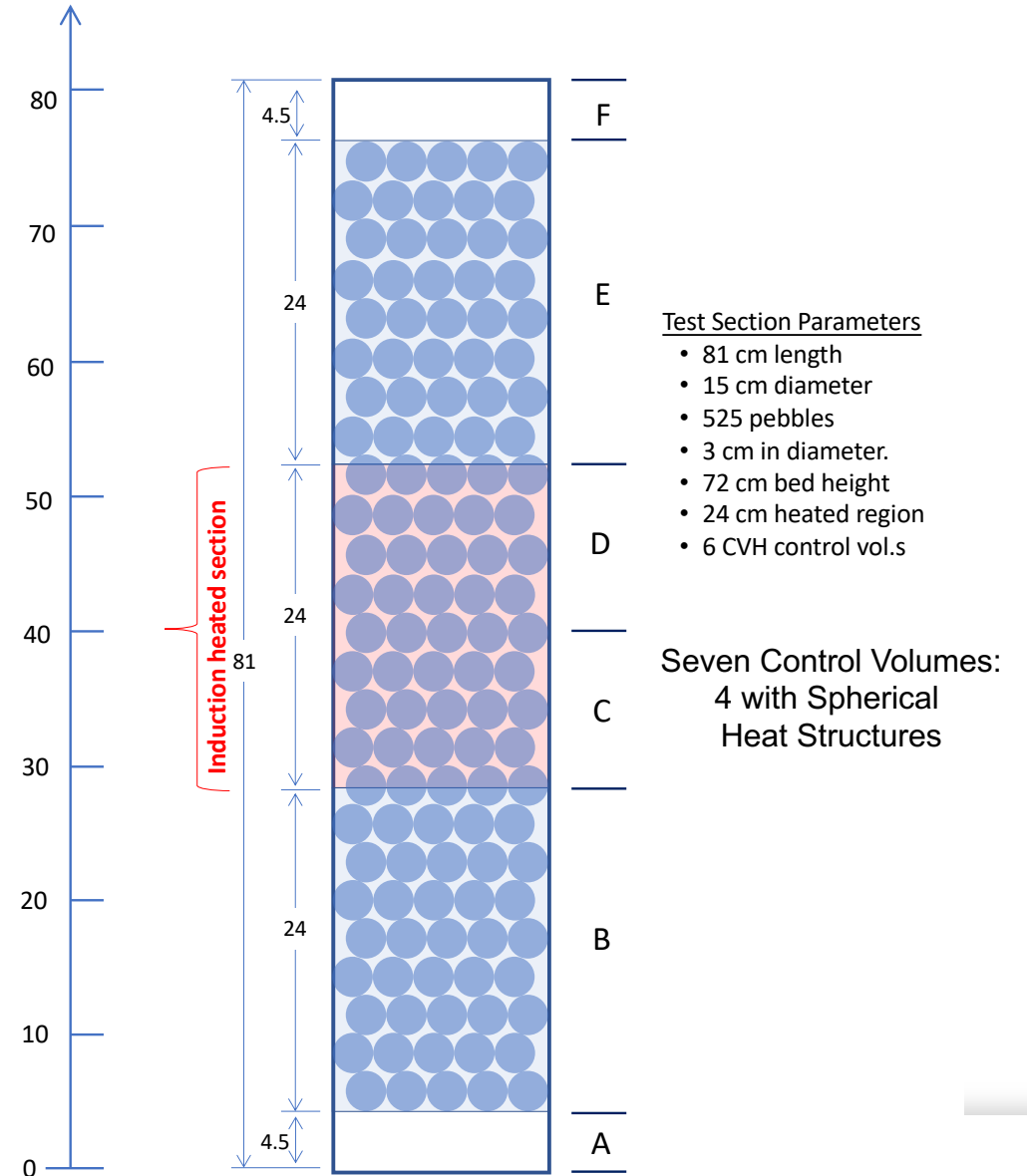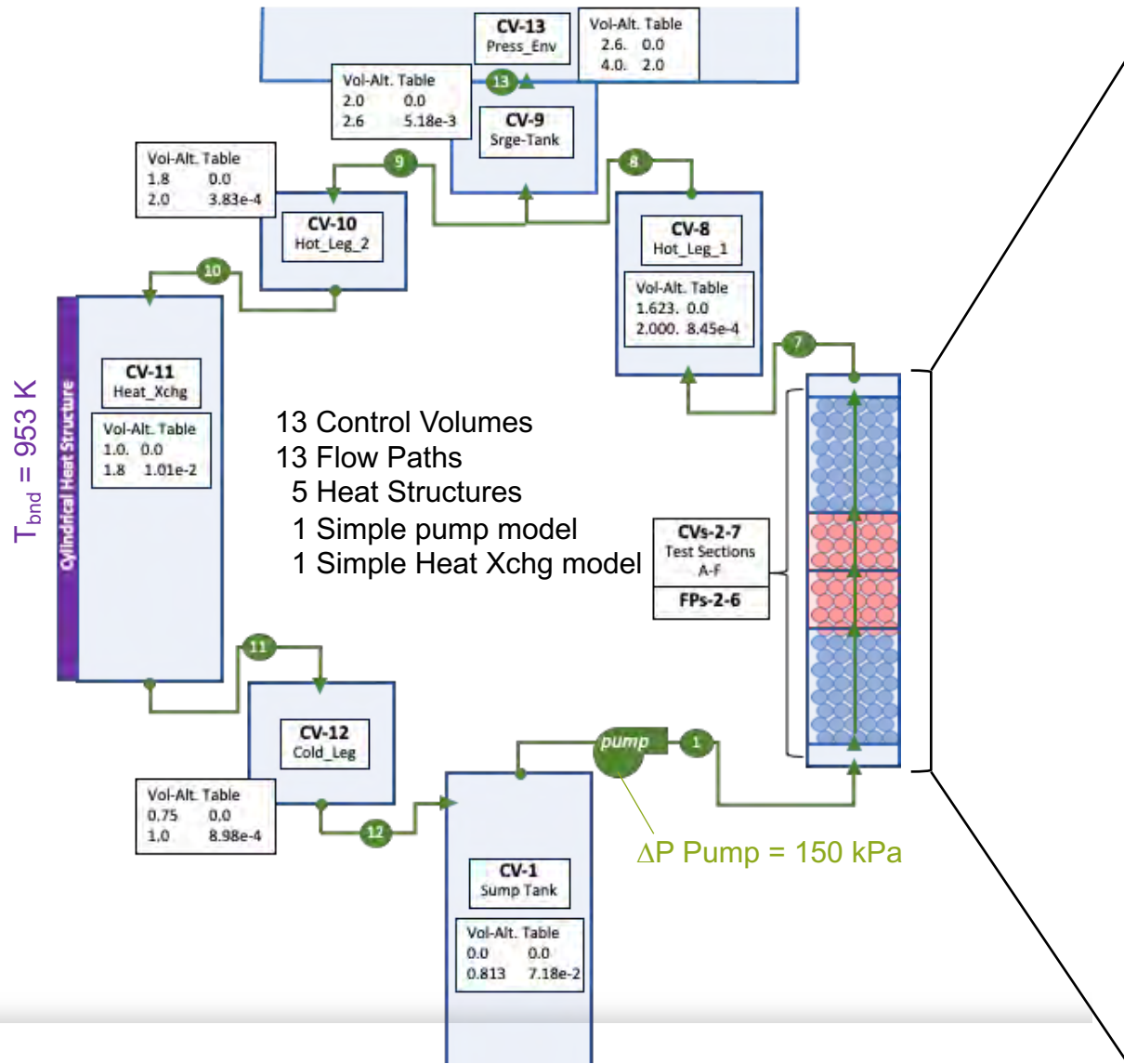
- MELCOR models represent a reactor or experiment with a collection of interacting and interconnected control volumes, flow paths, COR components, heat structures, etc.

- As a stepping stone to modeling liquid salt based reactors, this work focused on developing and demonstrating how MELCOR models can be used to model the steady state operation of the Liquid Salt Test (LST) loop

  - Liquid-salt-based coolant (but FLiBe instead of FLiNaK)
  - Heat-structure-based representation of Pebble Bed test section.
  - Testing of "in-development" MetaVolume approach in CVH to our standard approach.

- Aspects of the test not central to the purpose of this work are represented very simply: e.g., heat exchanger, pump, sump tank, some material properties.

- Some dimensions and geometric details had to be estimated as they were not available.

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Diagram of MELCOR Control Volumes, Flow Paths, and Heat Structures

13 Control Volumes
13 Flow Paths
 5 Heat Structures
 1 Simple pump model
 1 Simple Heat Xchg model

T_{bnd} = 953 K

ΔP Pump = 150 kPa

Test Section Parameters
- 81 cm length
- 15 cm diameter
- 525 pebbles
- 3 cm in diameter.
- 72 cm bed height
- 24 cm heated region
- 6 CVH control vol.s

Seven Control Volumes:
    4 with Spherical
    Heat Structures

Induction heated section

Spherical Heat Structure Modeling of Pebble Bed as a Porous Medium

- Until now, only the core package could be used to model a region as a porous medium.
- As part of this work, the MELCOR source code was adapted and modified to leverage the spherical heat structure model in the "HS" package as an alternate basis for modeling flow through a porous medium.
- Flow paths can now be designated as associated with flow through a heat-structure-based pebble bed through a new option (PBD-HS) in the "fl_blk" input card.
- Friction factors are computed using the Ergun equation as described in the MELCOR theory manual for flow through degraded geometries.
- The new capability makes setting up such problems simpler than before and also allows for the temperature gradient within the spherical pebbles to be modeled.
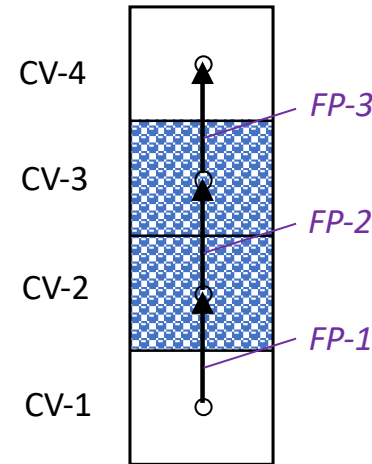
- Either the "From" or "To" CV must contain a spherical heat structures, here called pebbles.
- The FP must contain only one field (pool/ Atm)
- The FP will always have two segments, one associated with each of the two CVs. The segment length in any pebble-filled CV will always extend to the center of the CV.
- The user-specified fluid volume in the CVs containing the pebbles **does not** account for the pebbles. It is considered the free volume of the "pores".
- The multiplicity and dimensions of the spherical heat structure are used to compute the pebble volume.
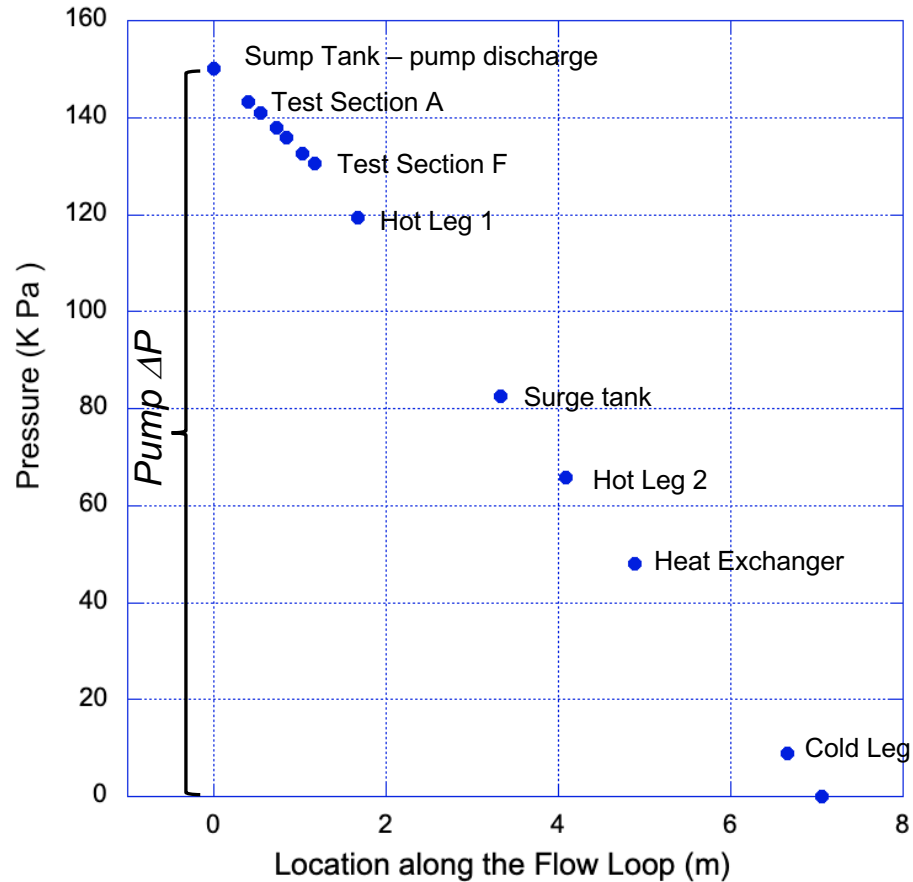- The porosity ε of the bed is calculated as:

(fluid volume) / (fluid volume + pebble volume)

CV-4
FP-3
CV-3
FP-2
CV-2
FP-1
CV-1

Ergun Equation:
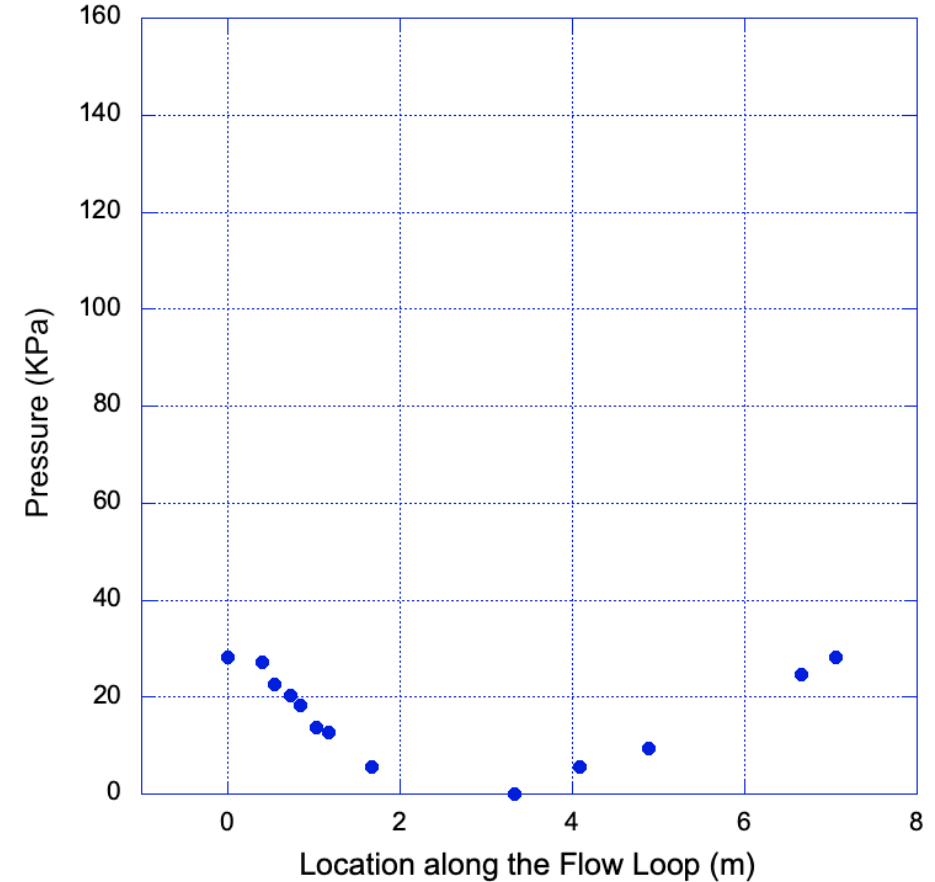
$$\Delta P_j^f = \frac{1}{2} K_j^* \rho_j |v_j| v_j$$

$$K^* = K_{empty} + \left[C_1 + C_2 \left(\frac{(1-\varepsilon)}{Re} \right) \right] \frac{(1-\varepsilon)L}{\varepsilon D_p}$$

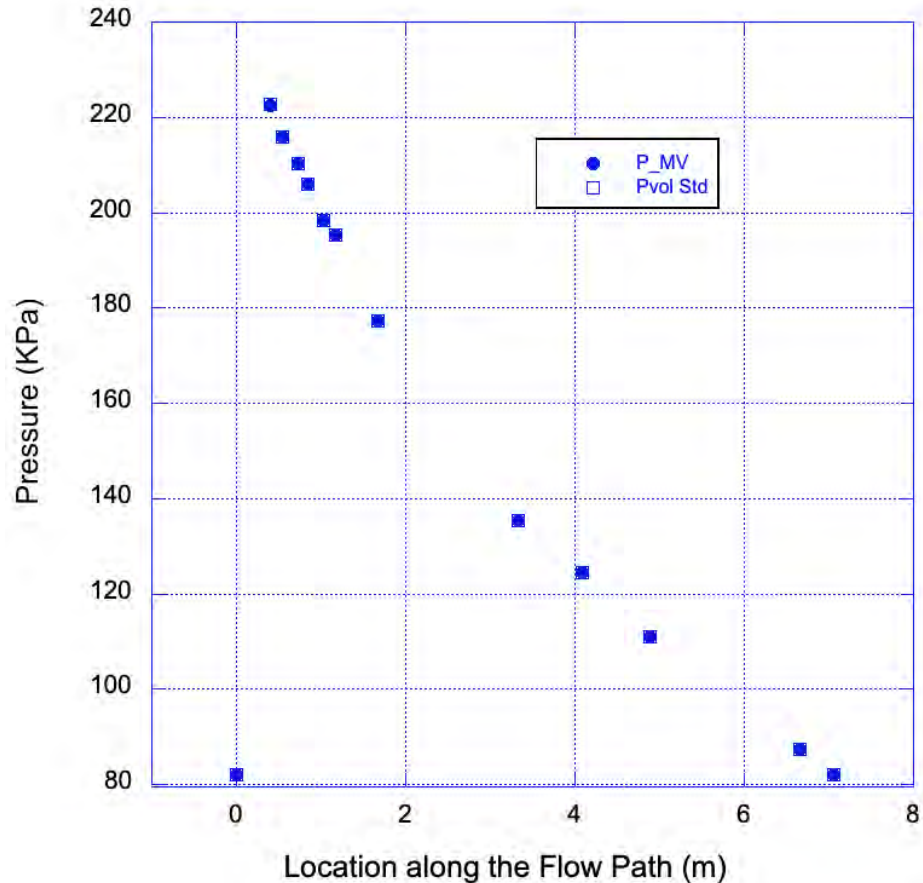$$Re = \frac{\rho \, (\varepsilon \, v_j) D_p}{\mu}$$

Pump ΔP and Friction-loss Balance

Hydrostatic Pressure Field

- Verification of expected friction losses across model volumes
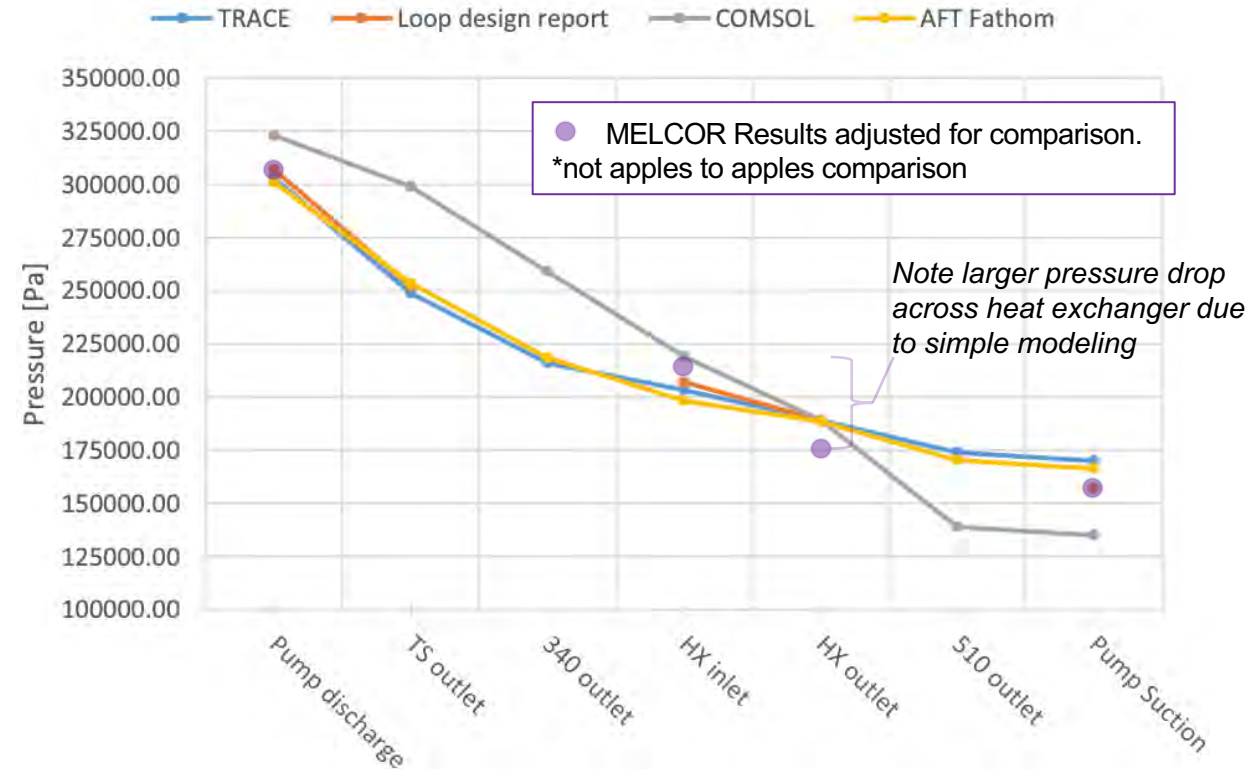- Verification of expected hydrostatic pressure field

Absolute Pressure (includes hydrostatic)

P. Avigni et al./Annals of Nuclear Energy 106 (2017) 170–184

MELCOR Results adjusted for comparison. *not apples to apples comparison

Note larger pressure drop across heat exchanger due to simple modeling

MELCOR result comparison with Avigni et al. figure 12 results if the MELCOR pressure range is adjusted.
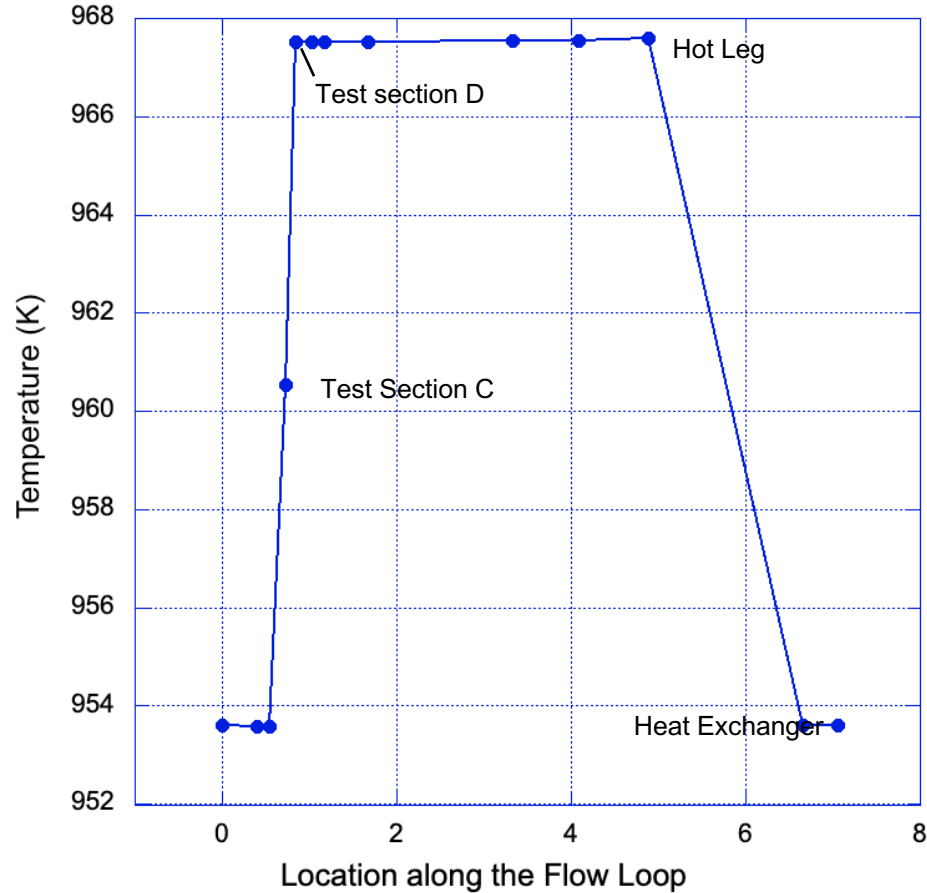
- Absolute pressure demonstrates variation in pressure across the loop domain
- Results compare favorably with previous calculations given modeling assumptions

Results: Temperature Plots


Control Volume Liquid Temperatures
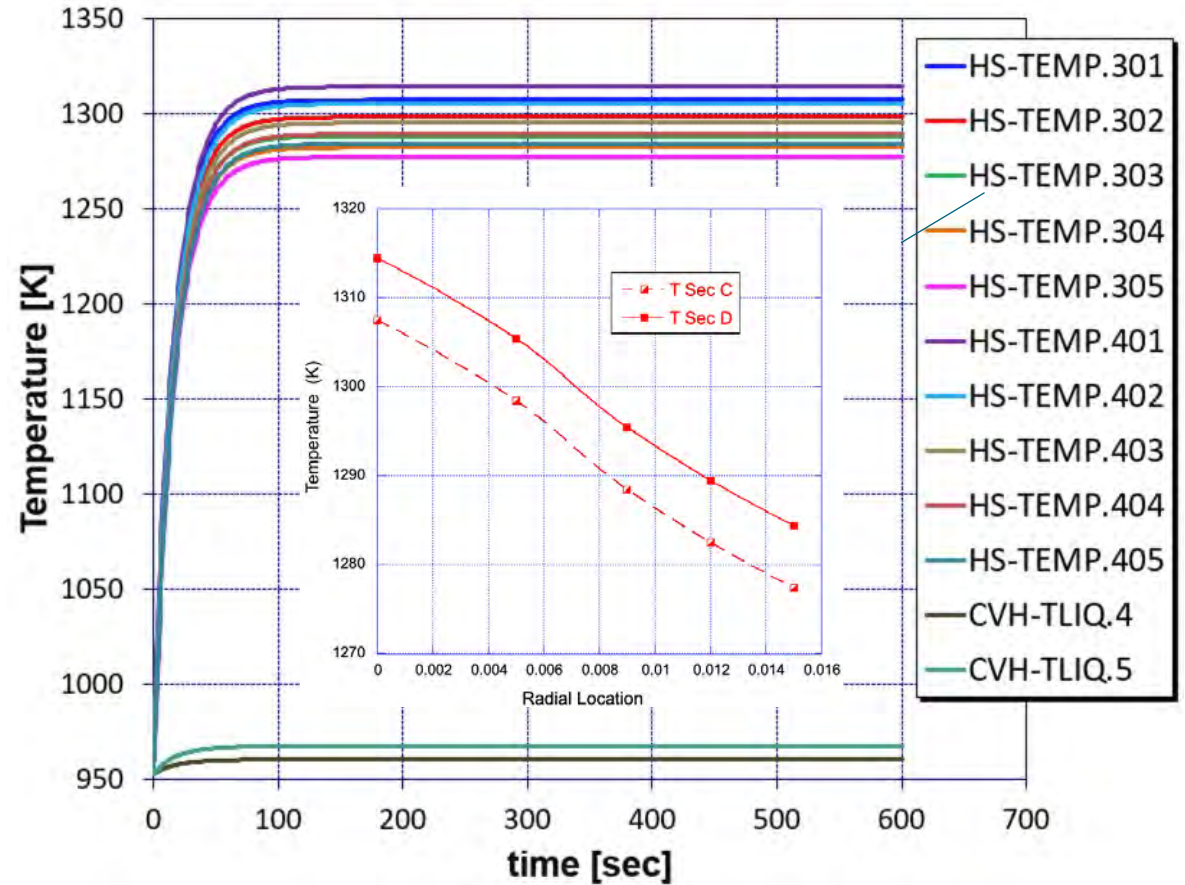
Transient thermal response and steady state radial variation of temperatures within the spherical heat structures

- Only two heated sections, and pipes assumed insulated
- Temperature profile generally compares favorably with expectations given model assumptions

Comments and Discussion

- MELCOR has successfully been used to model the steady state operation of the Liquid Salt Test (LST) loop. This was facilitated by helpful consultations with ORNL staff.

- The focus of this work has been on developing and testing several MELCOR models important for representing liquid salt based reactors.
  - Liquid-salt-based coolant
  - Heat-structure-based representation of Pebble Bed test section.
  - Testing of "in-development" MetaVolume approach in CVH to our standard approach.

- Other aspects of the actual LST experiment were either treated differently or modeled very simply, thus differences with available data were expected.
  - FLiBe instead of FLiNaK
  - Simplified modeling of heat exchanger, pump and sump tank.
  - Details of piping (elbows, exact lengths., etc) neglected.
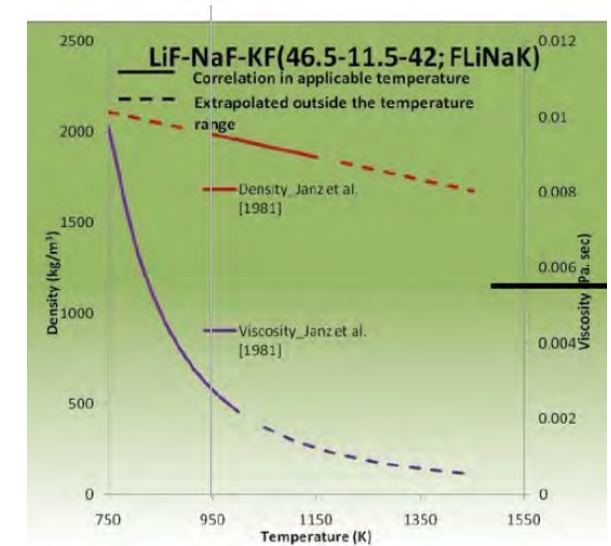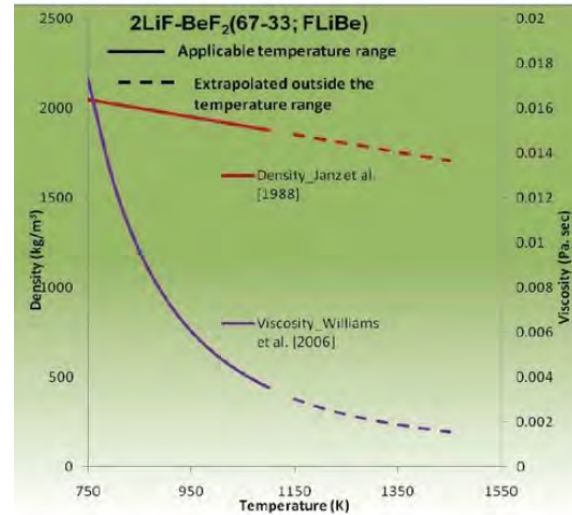
Comparison of FLiBe vs FLiNaK Properties

- Densities are fairly close
- FLiBe viscosity is larger than that of FLiNaK (note different scales)

MELEOS – Generalized Equation of State Generator for MELCOR

MELCOR EOS

- Default Fluid: Water
- Implementation
  - Basis: Density-Temperature
  - Single phase: functional implementation (Keenan and Keyes)
  - Two-phase: pre-computed table of vapor dome values
- Properties:
  - Pressure (and derivatives with respect to the basis)
  - Internal Energy
  - Entropy
  - Enthalpy
  - Isochoric Heat Capacity

- Two Extensions
  - Sodium Chemistry Package
    - SIMMER EOS implementation
    - Function-based (no tables) for one- and two-phases
  - External file-read
    - Table-based EOS, cubic/linear interpolation
    - Requires viscosity, thermal conductivity, surface tension closures in codebase
    - Fluids:
      - Sodium
      - Lead-Lithium
      - FLiBe
      - … others

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Goals

- Primary: Generate MELCOR an EOS for Uranium Chloride

- Chosen path
  - Develop a library to generate the above EOS
  - Ensure library is general to data input and functional forms
  - Provide both functional and table implementations
  - Based EOS on Helmholtz potential theory

- Influences
  - *Multiparameter Equations of State* by Roland Span
  - Work of Humrickhouse and Merrill
    - *Revised Equation of State for FLiBe in MELCOR* (INL/EXT-17-44148)
    - *An Equation of State for Liquid $Pb_{83}Li_{17}$* (INL/CON-16-38063)

Helmholtz Potential

- A fundamental relation from thermodynamic theory

- Natural function of density and temperature

- Related to all thermodynamic variables

- Can be made accurate over the entire fluid range (liquid and gas phases)

- Differential definition of Helmholtz Free Energy f in terms of the entropy s, temperature T, pressure P, and volume v / density ρ

- $df = -s\,dT - P\,dv = -s\,dT - P\,d(1/\rho)$

- Integral definition of Helmholtz Free Energy f in terms of the internal energy e, temperature, and entropy

- $f = e - Ts$

- Helmholtz Relations

  - $P = \rho^2 \dfrac{\partial f}{\partial \rho}$

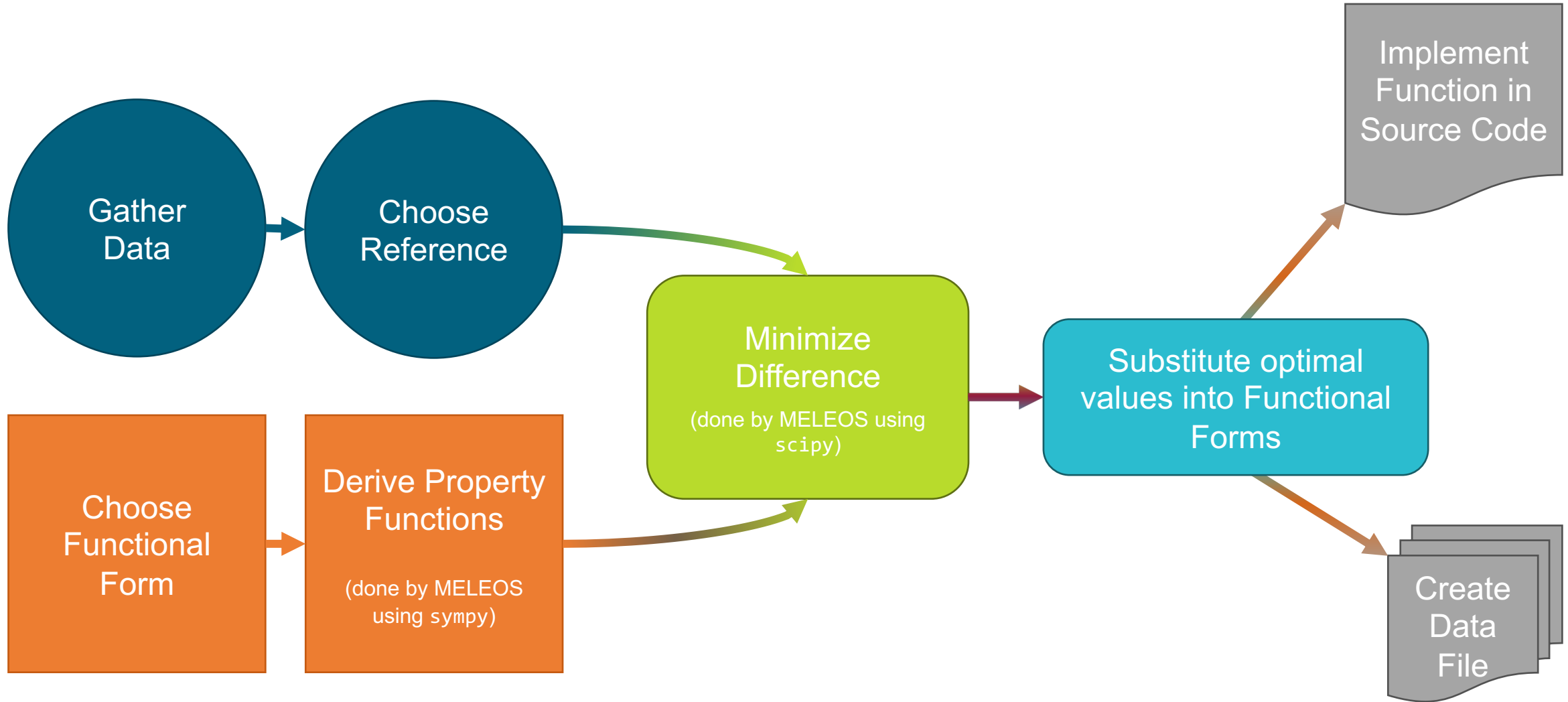 - $e = -T \dfrac{\partial f}{\partial T} + f$

  - $s = -\dfrac{\partial f}{\partial T}$

  - $c_p = -T \dfrac{\partial^2 f}{\partial T^2} + \dfrac{\rho T \left(\frac{\partial^2 f}{\partial \rho \partial T}\right)^2}{\left[2\frac{\partial f}{\partial \rho} + \rho \frac{\partial^2 f}{\partial \rho^2}\right]}$ (Isobaric Heat Capacity)

  - $w^2 = \rho \left[2\dfrac{\partial f}{\partial \rho} + \rho \dfrac{\partial^2 f}{\partial \rho^2} - \rho \dfrac{\left(\frac{\partial^2 f}{\partial \rho \partial T}\right)^2}{\frac{\partial^2 f}{\partial T^2}}\right]$ (Isentropic sound speed)

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MELEOS

- Python Library written to generate equations of state
  - Currently only has Helmholtz definitions, easy to add others (e.g., Gibbs)

- Leverages three popular Python packages
  - `numpy` (arrays and calculation)
  - `scipy` (optimization and root finding)
  - `sympy` (symbolic computation of expressions and derivatives)

- Creation of generic correlation objects
  - Arbitrary symbolic form (limited only by `sympy`'s capability)
  - Distinction between variables (e.g., density, temperature) and curve-fitting parameters for Jacobian creation

- Creation of Helmholtz objects from correlations
  - Generates thermodynamic identities for each property
  - Generates the functional form of each property as function of density, temperature, and fit parameters

- Creation of binary data files compatible with MELCOR's requirements

- Process
  1. Gather Data with complete thermodynamic states, typically three state properties for pure substances (e.g., ρ-T-P or ρ-T-c_p)
  2. Choose Reference state to gauge non-absolute properties like internal energy and entropy
  3. Choose Form: choose a functional form of the potential that is a function of its state variables (e.g. ρ and T) and fitting parameters
  4. Derive Properties' functions from potential's functional form
  5. Minimize the distance/difference between the property data and the property functions evaluated at the corresponding state points by varying the fitting parameters
  6. Substitute optimal fitting parameter values into function form
  7. Implement the function in source code or create external data file

MELEOS

Gather Data → Choose Reference

Choose Functional Form → Derive Property Functions (done by MELEOS using sympy)

Minimize Difference (done by MELEOS using scipy)

Substitute optimal values into Functional Forms

Implement Function in Source Code

Create Data File

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Perfect Gas Example

- Air properties
  - $c_v = 716$ [J/kg-K]
  - $R_{air} = 287.79$ [J/kg-K]
- Surrogate state data:
  - $T = [300\text{ K}, 400\text{ K}]$
  - $P = [1\text{ atm}, 2\text{ atm}]$
- Surrogate property data
  - Ideal gas law for density
  - Constant specific heat relations for internal energy and entropy
- Functional form
  - $\dfrac{f(\rho, T; c^0, c^I, c^{II})}{R_{air} T} = c^{II} + c^I \left(\dfrac{T_r}{T}\right) + \log\left[\dfrac{\rho}{\rho_r}\left(\dfrac{T_r}{T}\right)^{c^0}\right]$
  - $T_r = 300\ K$
  - $\rho_r = 1.17$ [kg/m³] are nondimensionalizing values

- Equations to minimize
  - $e_{data} = R_{air}(c^0\,T + c^I\,T_r)$
  - $s_{data} = -R_{air}\left(-c^0 + c^{II} + \log\left[\dfrac{\rho}{\rho_r}\left(\dfrac{T_r}{T}\right)^{c^0}\right]\right)$
- (Optimal) Fit Parameter values:
  - $c^0 = 2.48790$
  - $c^I = -2.48790$
  - $c^{II} = 2.48790$

- Substituting into property relations
  - $e(T) = 716.0\,T - 214800.0$
  - $s(\rho, T) = -287.79 \log\left[1239720 * \dfrac{\rho}{T^{2.4879}}\right]$

Perfect Gas Example
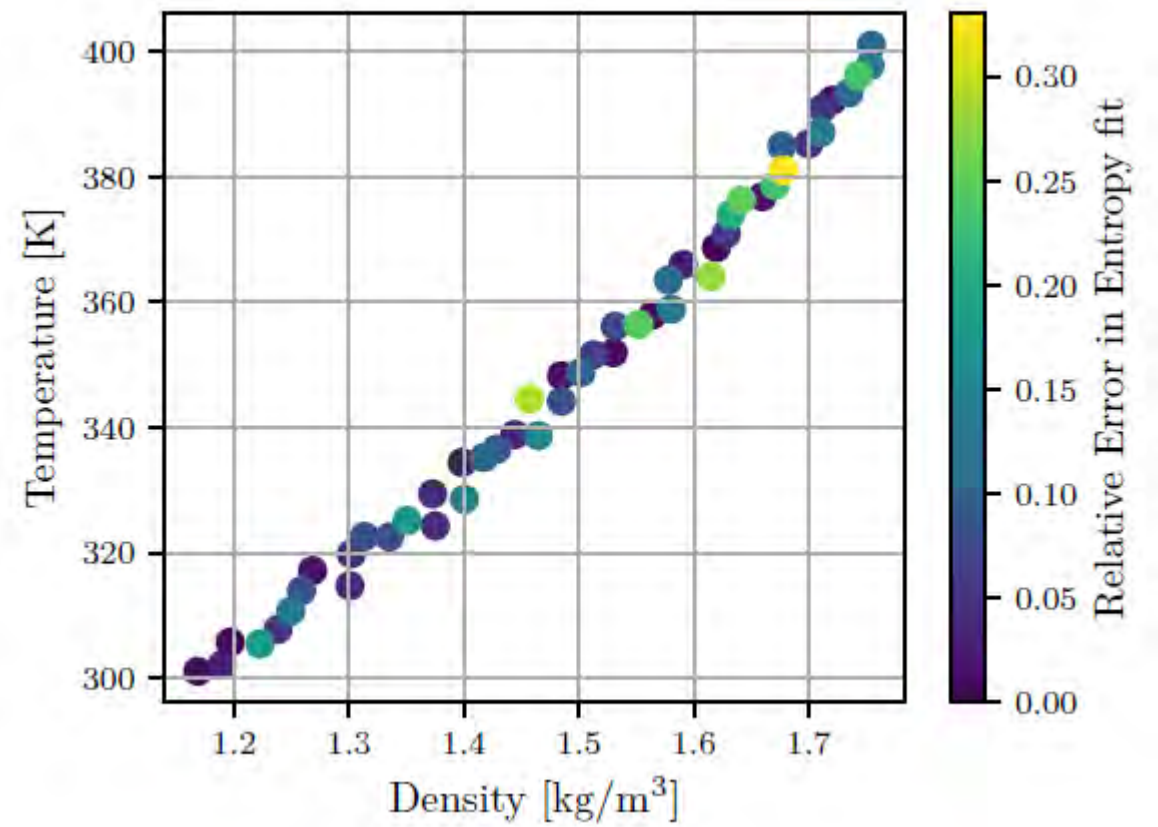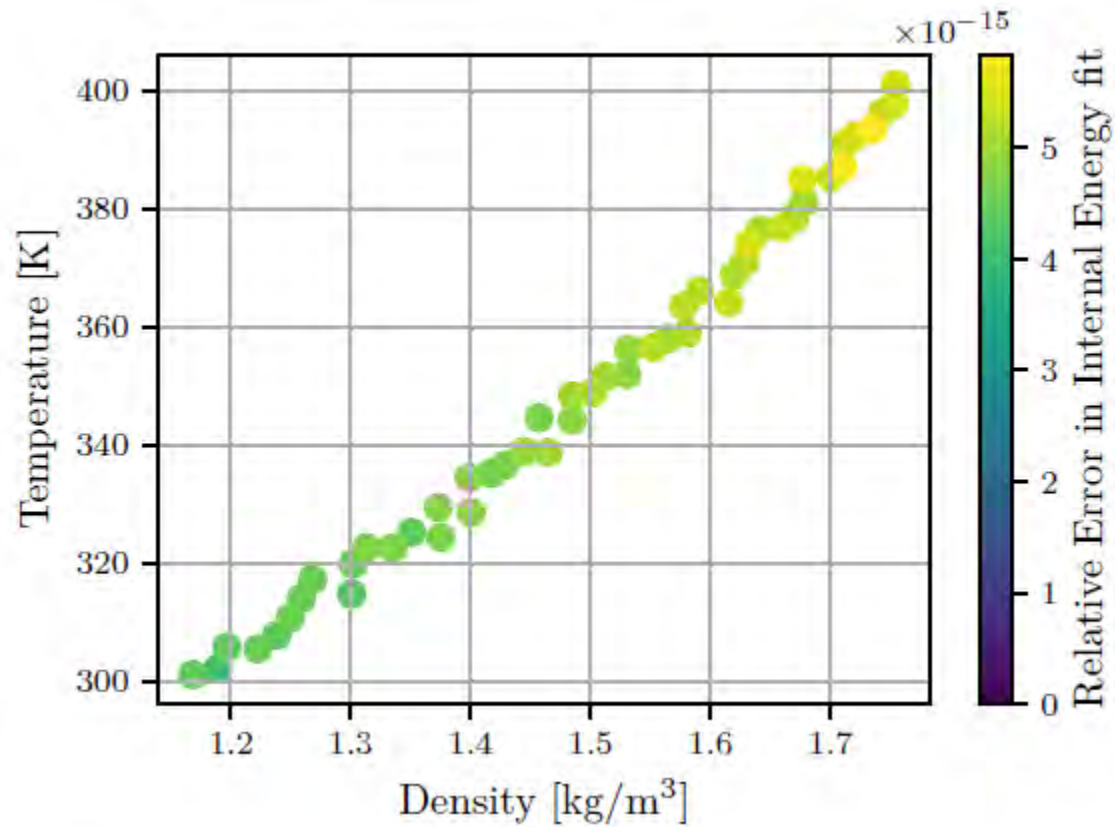
Current Uranium-Chloride EOS

- Experimental data from Desyatnik et al.
  - Several molar fractions: $(1-x)\mathrm{NaCl}, x\mathrm{UCl}_3$
  - Density-temperature (assumed at 1 atmosphere)
  - Surface tension-temperature
  - Viscosity-temperature
- Density data for 0.495-UCl_3:
  $$\rho(P = 101325[Pa], T) = 4.4738 \cdot 10^3 - 0.9304\,T$$
- Took six equally-spaced value from temperature range:
  $$T \in [951K, 1943K]$$
- Caloric value from Kasam and Shwageraus:
  - $c_p = 435.6$ [J/kg−K]
  - Assumed constant over temperature range
- Reference state
  - $(T = 951, P = 101325)$
  - internal energy and entropy zero point.

- $$\frac{f(\rho, T)}{R_{salt}T} = c^{II} + c^{I}\left(\frac{T_r}{T}\right) + Z \log\left[\frac{\rho}{\rho_r}\left(\frac{T_r}{T}\right)^{c^0}\right]$$
  $$+ m \log\left[1 - \exp\left(-\vartheta\,\frac{T_r}{T}\right)\right]$$
  $$+ n \left(\frac{\rho}{\rho_r}\right)^{d}\left(\frac{T_r}{T}\right)^{t}$$
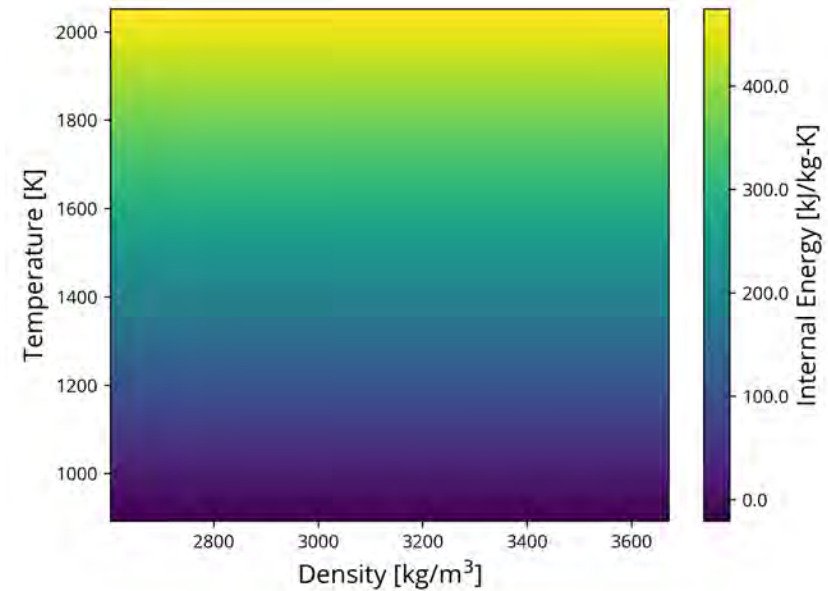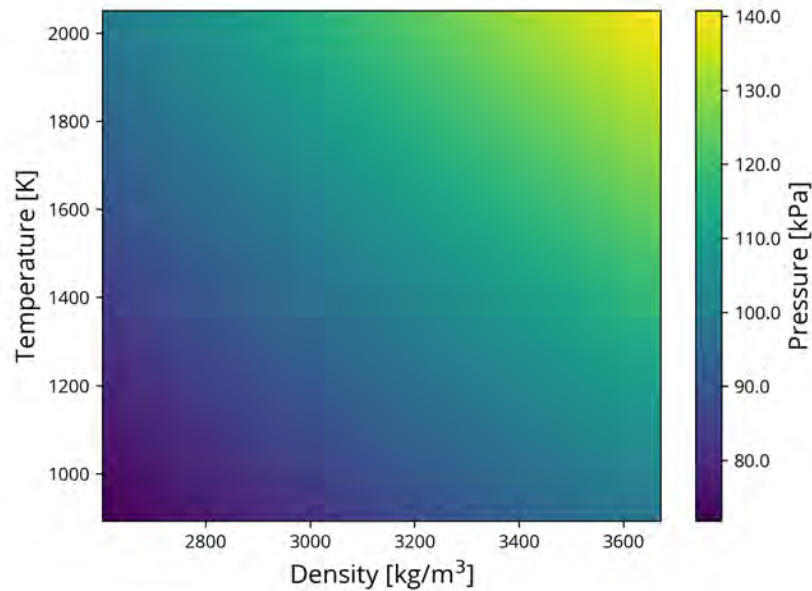
- Fit values from data
  - $c^0 = -32.01$
  - $c^I = -10.78$
  - $c^{II} = 81.09$
  - $Z = 0.0002325$
  - $m = 10.48$
  - $\vartheta = 0.001187$
  - $n = 0.3109$
  - $d = 0.001524$
  - $t = 0.9997$

Current Uranium-Chloride
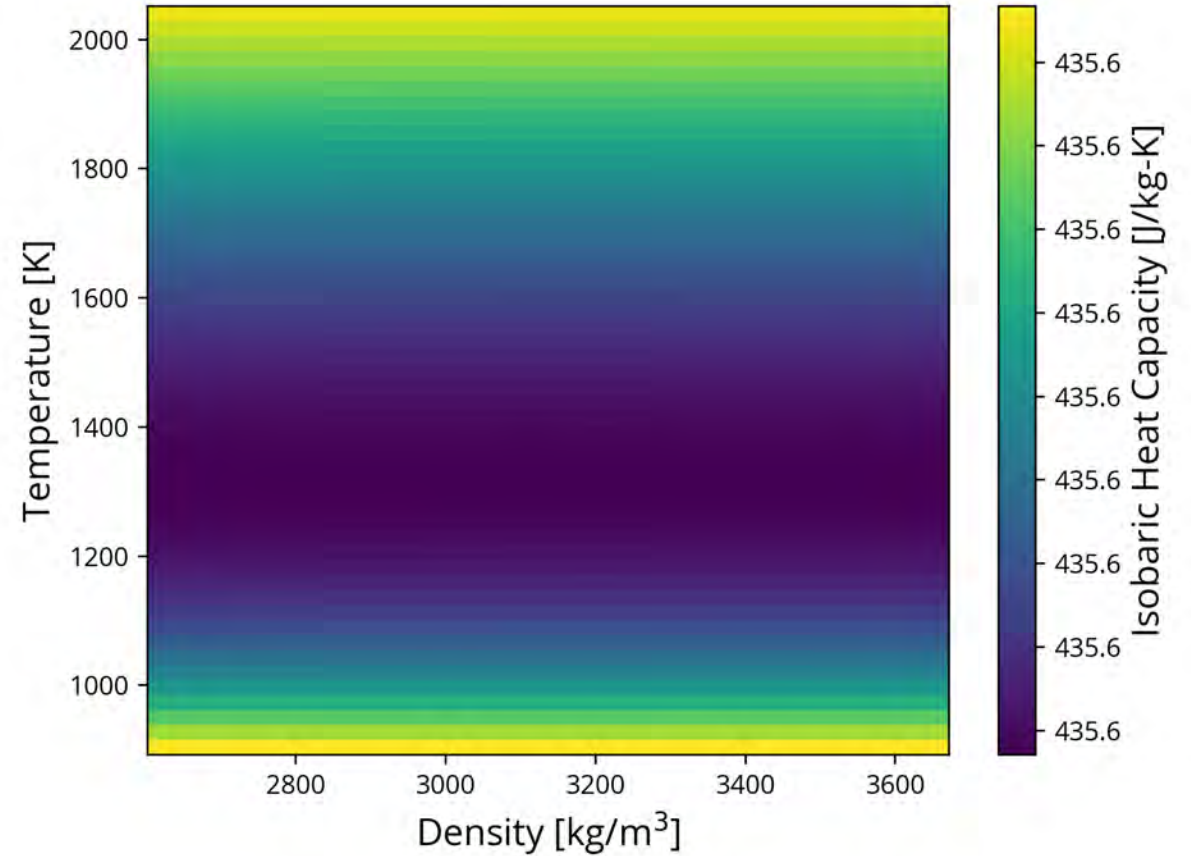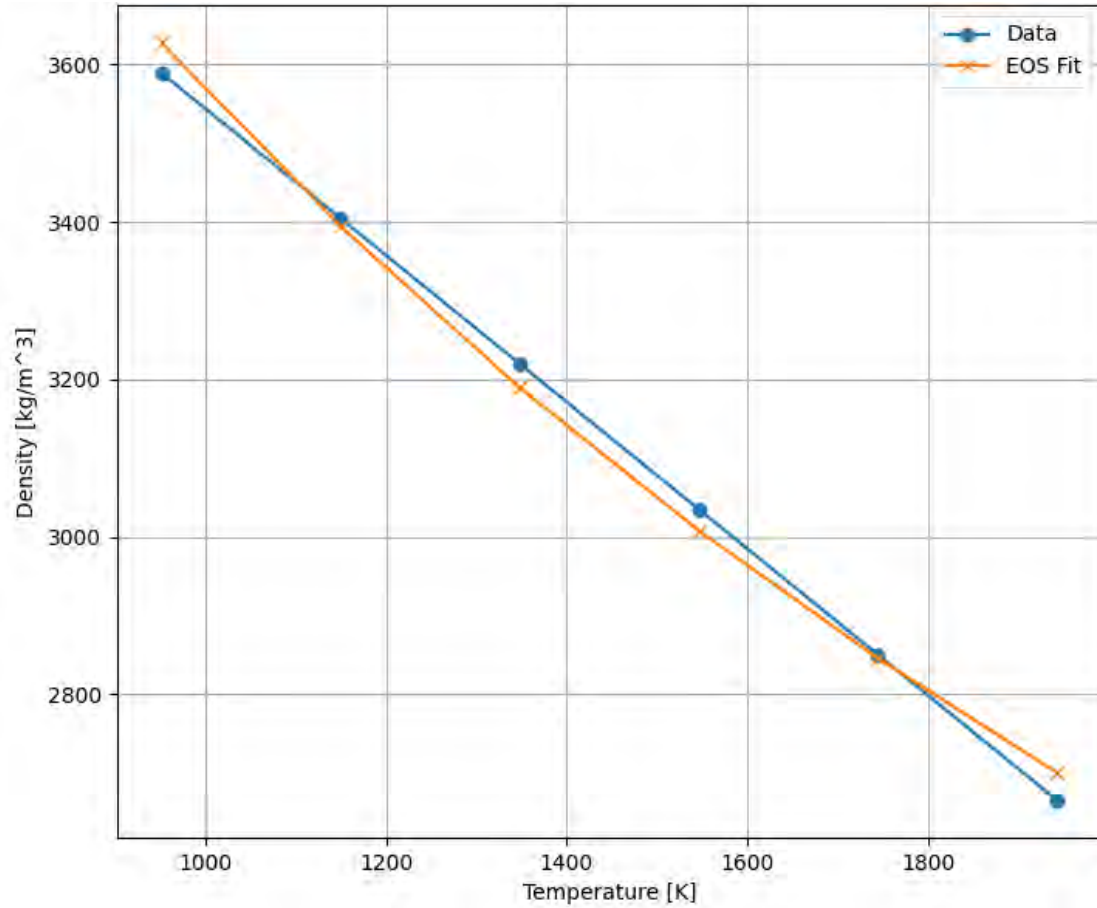
Current Uranium-Chloride

ρ [kg/m^3]	T [K]	P [kPa]	e [J/kg]	s [J/kg-K]	c_p [J/kg-K]	w [m/s]	α [1/K]
3589	**951.0**	**100**	$\mathbf{6.92 \cdot 10^{-6}}$	$\mathbf{-1.26 \cdot 10^{-9}}$	**435.6**	**5.29**	$\mathbf{3.46 \cdot 10^{-4}}$
3589	1046	103	$4.14 \cdot 10^{4}$	$4.15 \cdot 10^{1}$	435.6	5.37	$3.35 \cdot 10^{-4}$
3553	951.0	99.3	$-1.88 \cdot 10^{-1}$	$9.72 \cdot 10^{-5}$	435.6	5.29	$3.46 \cdot 10^{-4}$
3553	1046	103	$4.14 \cdot 10^{4}$	$4.15 \cdot 10^{1}$	435.6	5.37	$3.35 \cdot 10^{-4}$

Next Steps

- Produce a binary external file for use with MELCOR
  - Make needed source code changes for non-thermodynamic data
  - Perform a demonstration calculation
  - Explore incorporating Helmholtz fit directly into source
- Gather other sources of data to improve current Chloride EOS
  - Sound speed is too low
  - Need variable pressure
- Fit other salts and compositions using MELEOS for more applications

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Summary and Next Steps

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Summary

- ## LSTL Model
  - MELCOR model of LSTL has been developed and shaken down
  - Enhanced MELCOR HS package to allow for heat-structure-based pebble bed
    - Spherical heat structures are used with the FL package to model flow through a porous medium
    - Previously only possible with full COR package definition (significant increase in model development effort)
    - The new capability simplifies input model development when COR package modeling is unnecessary and reduces input development costs
  - Collaboration with and support from ORNL staff has been very beneficial in this effort

- ## MELEOS
  - A python package to develop equations of state for use by MELCOR is under development
  - A first UCl_3 equation of state has been developed using MELEOS
  - This tool has significant capacity to support a broad range of current and upcoming MELCOR activities

Looking Forward

- LSTL Model
  - Continued verification of LSTL model
    - Implement FLiNaK as working fluid (dependent on MELEOS)
    - Etc.
  - Benchmark LSTL model with ORNL SAM model
  - Incorporate LSTL into validation matrix for MSR models as data becomes available
- MELEOS
  - Develop method for binary external file generation for use with MELCOR
  - Revisit chloride salt data sources, investigate use of synthetic data from MSTDB to improve resolution of the EOS
  - Fit other salts and compositions using MELEOS for MSR mechanistic source term applications
- Radionuclide Transport Modeling
  - Continue to engage with partners to identify opportunities for NEAMS tools to develop/inform/improve correlations implemented in MELCOR

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