Progress Toward Eliminating Use of HEU in ^{99}Mo Production with a Multi-Physics Simulation

A solution vessel design optimization

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75 Los Alamos
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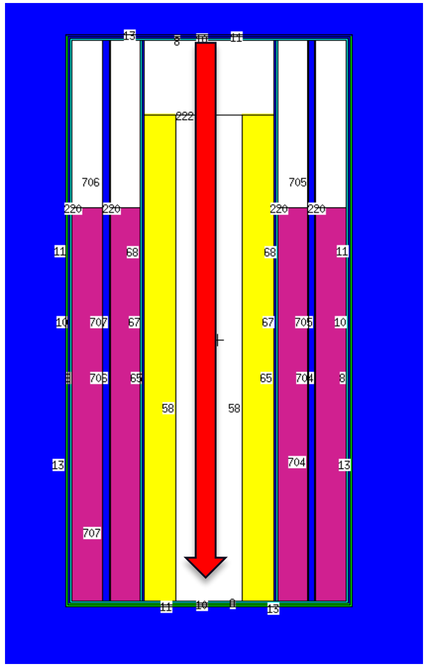
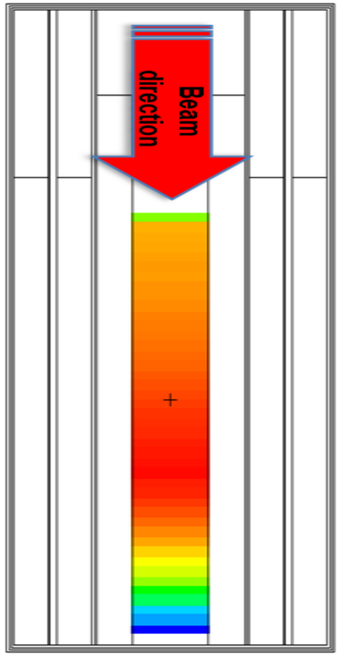
NNSA
National Nuclear Security Administration

Accelerator driven fissile solution system for Mo99

Fissile Solution Vessel model in MCNP

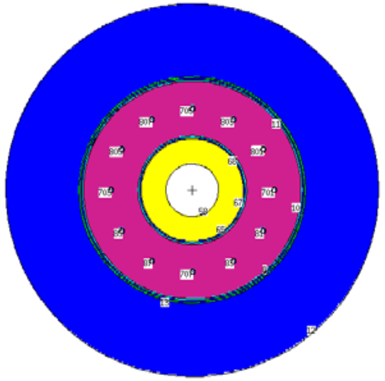
Side view

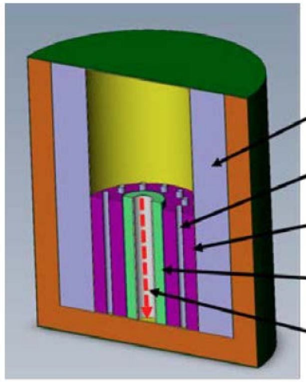




14Mev Mono energy Neutron generation by DT accelerator

Top view





Background:

- ❑ *Mo99 is a critical isotope in medical applications*
- ❑ *Domestic Mo99 needs are demanding (No production in USA)*

Innovations:

- ❑ *LEU based Mo99 production (No proliferation risk)*
- ❑ *Accelerator driven neutron source (subcritical, easy to turn off)*
- ❑ *Self-regulating feature (Liquid fuel reactor concept)*

Key Modeling Challenges:

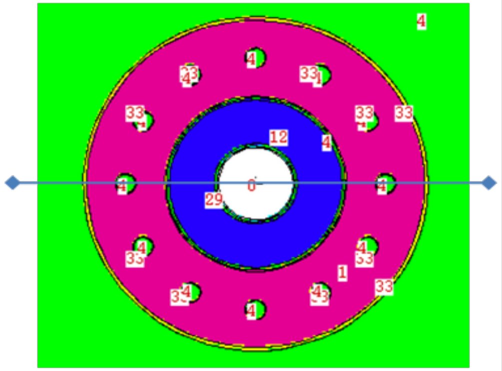
- ❑ ***Multi-physics coupled calculation for system evaluation***
- ❑ ***Bubbly flow modeling in TH code (i.e. Radiolytic gas)***
- ❑ ***Fissile Solution height adjustment (temp, void)***
- ❑ ***Non-uniform Fissile solution density***

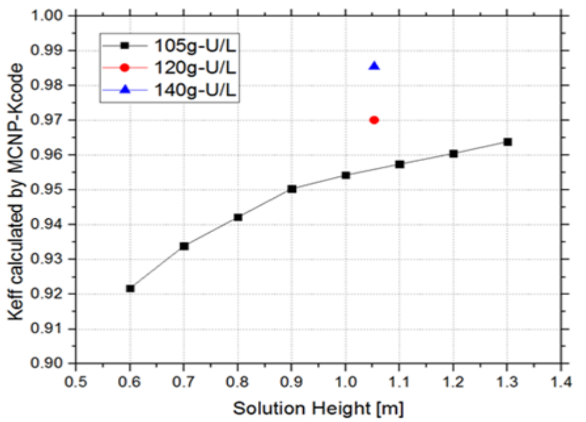
Applied simulation tools:

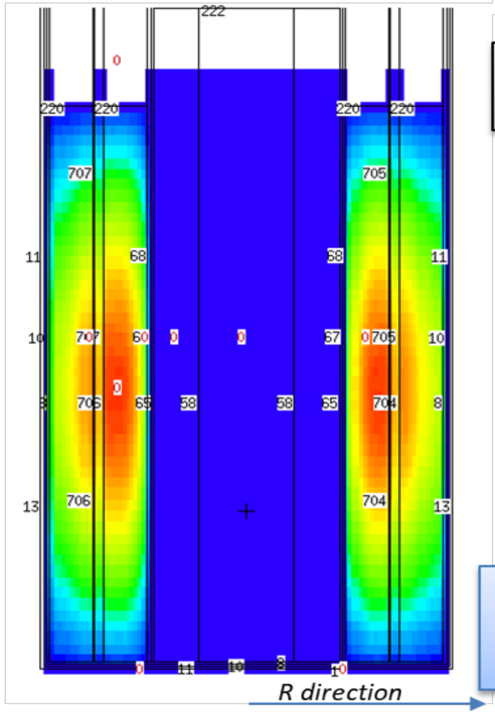
- ❑ *Neutronic calculation : MCNP6.2*
- ❑ *Thermal Hydraulic calculation : Fluent 17.2*
- ❑ *Automated coupling calculation frame toolkit : Python*
- ❑ *All calculation is performed in LANL HPC*

Neutronic Solver (MCNP calculation)

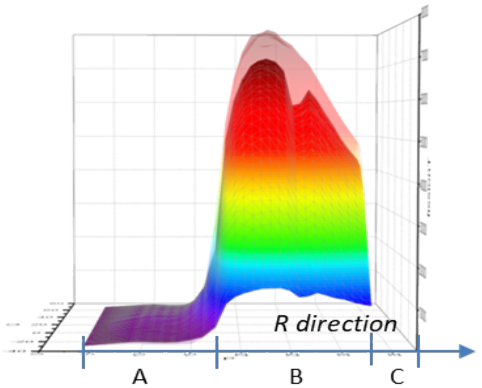
→ (Energy deposition calculation from heating tally







- *Energy deposition by FMESH tally*
- *Power profile (mapping into MCFD)*



A: Vacuum + moderator zone (no heating)
B: Fissile solution zone (heating source)
C: Outer water pool (no heating)

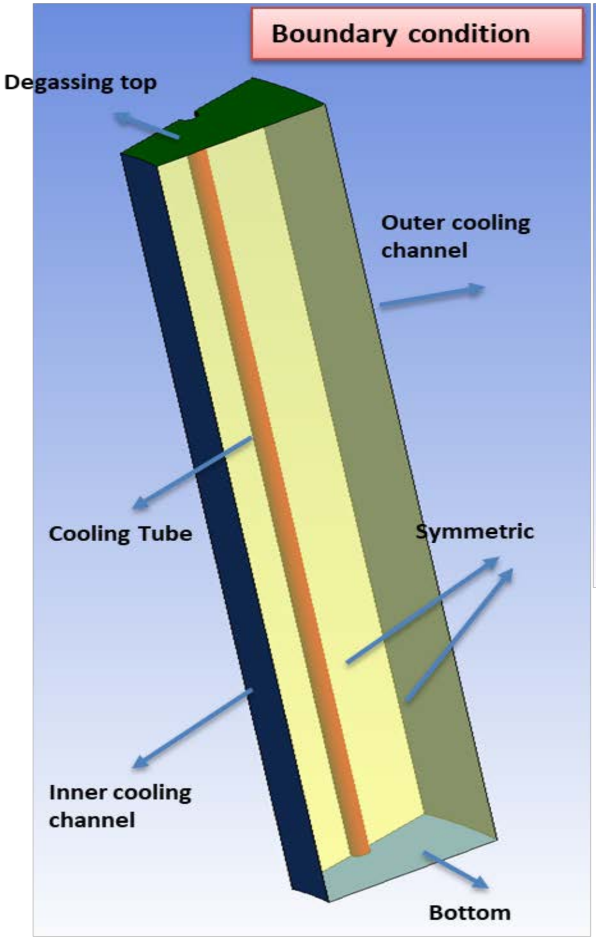
Fission induced power calculation

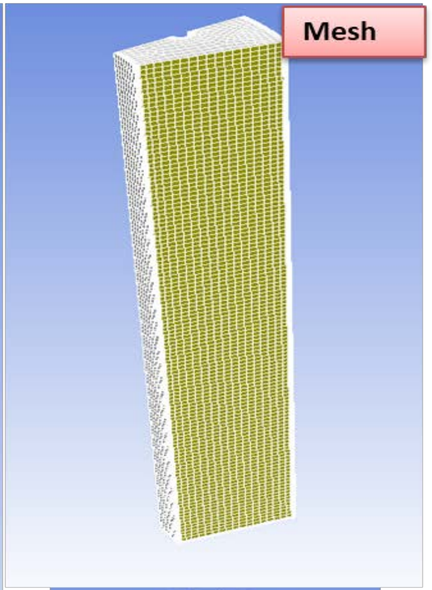
- MCNP6.2. with **ENDF/B-VIII.0**
- K-Code is performed to evaluate the sub-criticality of the system at the cold-start operating condition **(Keff = 0.98)**
- Reference solution for cold-start
  - *- Uranyl sulfate density : 1.85 kg/m3*
  - *- Solution concentration : 140gU/L*
  - *- Temperature : 20C*
- Solution density is function of concentration & temp. (ANL)
- Fission and heating tally are used to evaluate the azimuthally averaged power profile within the fissile solution vessel.

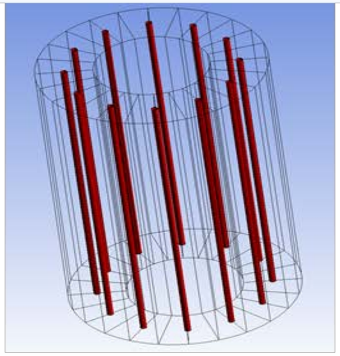
Thermal Hydraulic Solver (Multiphase natural circulation)

→(Temperature and Void profile calculation)

1/24th model







Full model

Multiphase CFD specification

- Fluent17.2 used
- Eulerian based two fluid model approach
- Pseudo transient analysis for steady state calculation

- Hex dominant mesh with prism mesh at the wall (Y+<1)
- URANS turbulence model (SST K-ω)

- Volumetric power (energy) and bubble generation (mass) profiles implemented via UDF script

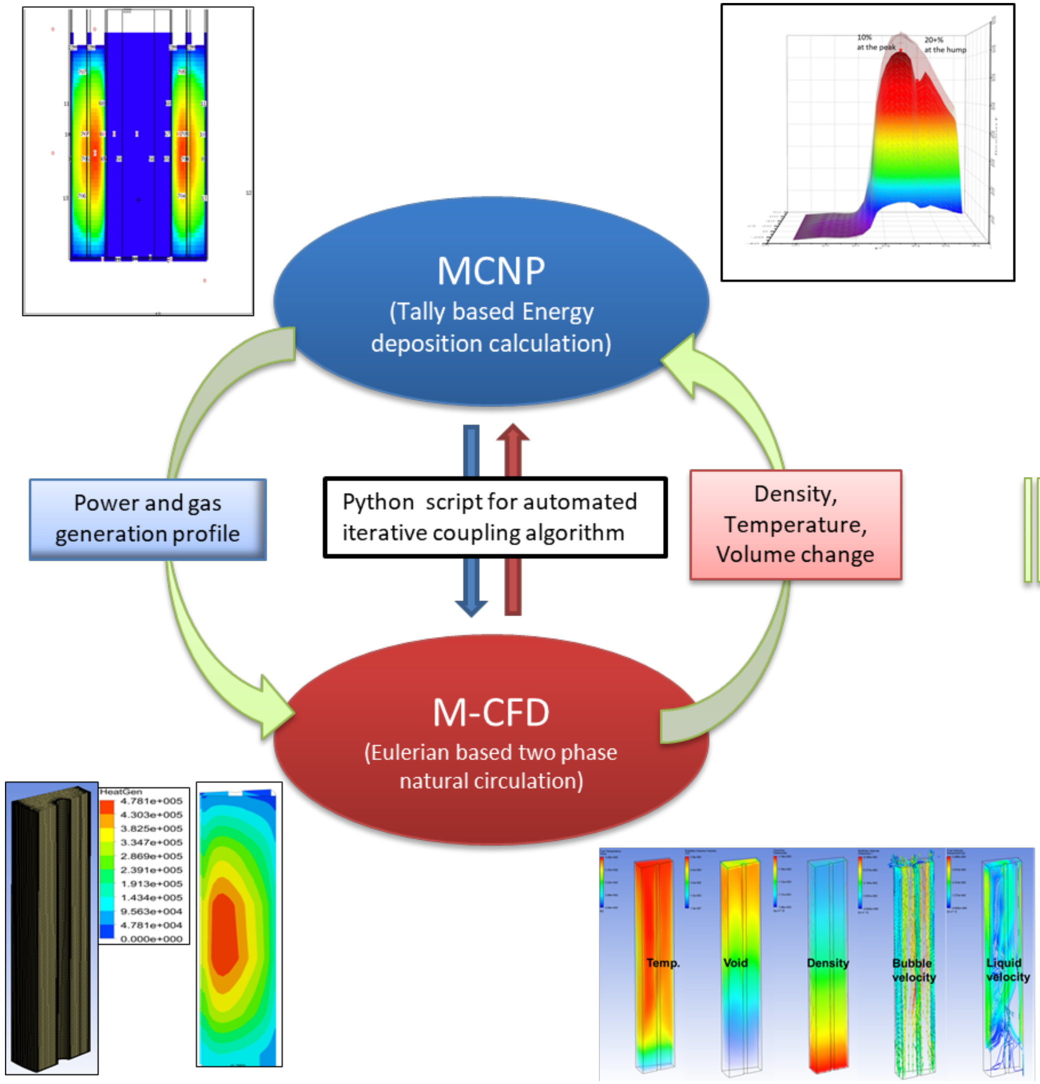
- Convergence check on energy and mass balance
  - Energy in (MCNP) = Energy out (M-CFD)
  - Bubble generation = Bubble loss at degassing surface

- Convective heat transfer coefficients (HTCs) for outer side of cooling surfaces are evaluated based on empirical Gnielinski correlation.

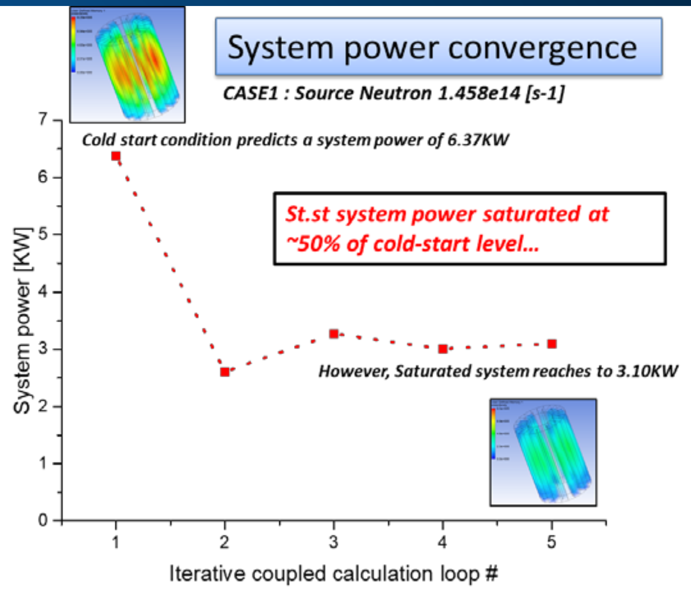
- Multiphase closure model described in back-up slides

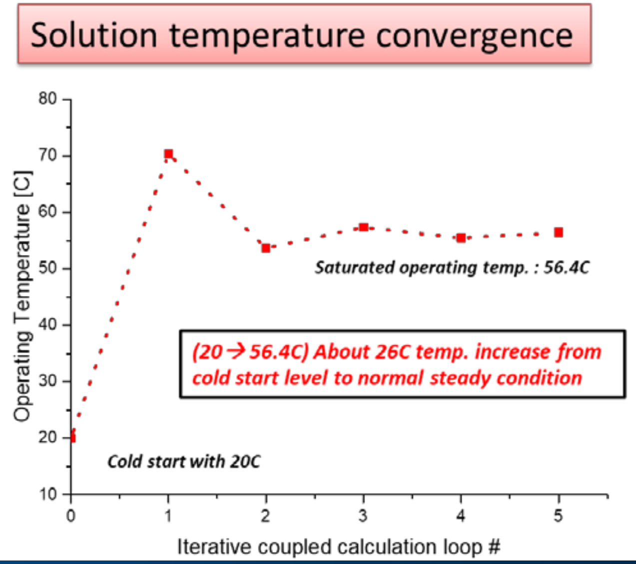
Multi-Physics modeling for fissile solution vessel

→(Power calculated by MCNP ←→ thermal hydraulic calculated by M-CFD)



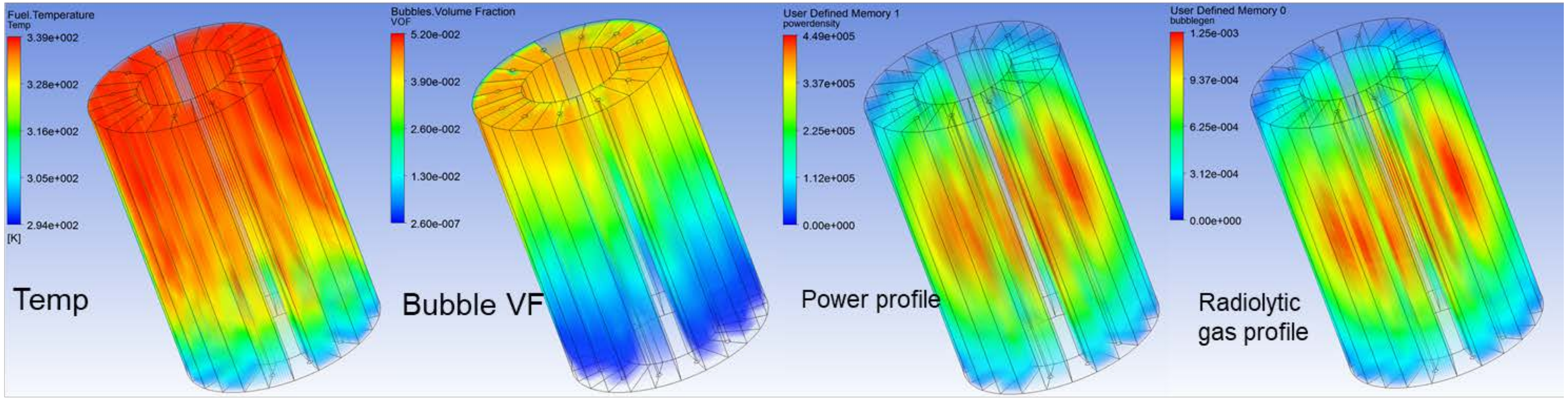
Fission Solution System Analysis





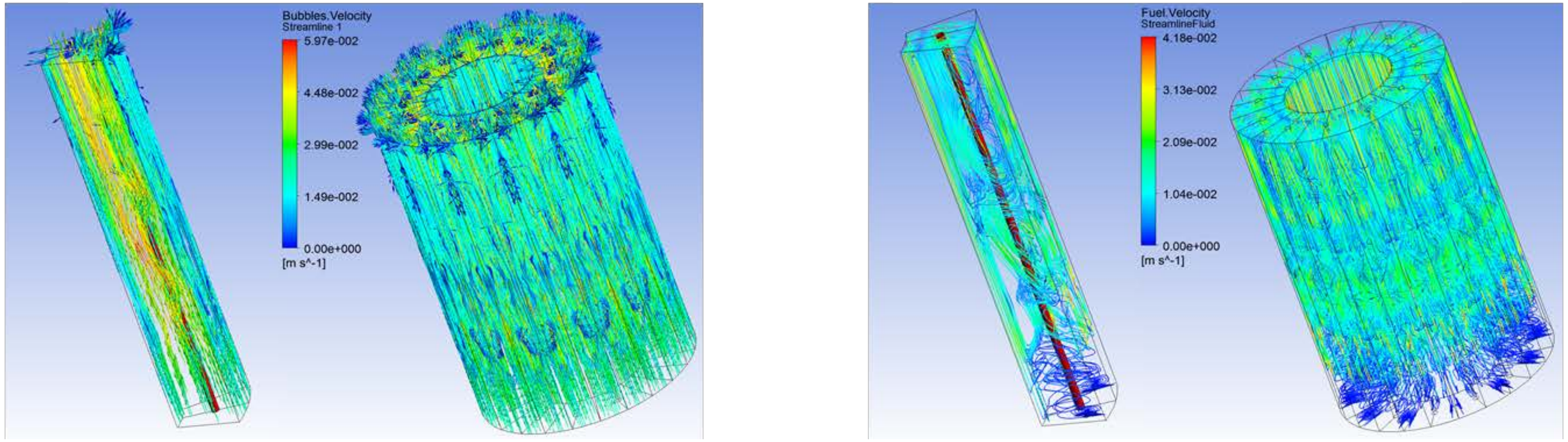
Thermal Hydraulic parameters calculated from M-CFD

→Temp., Void distribution and bubble rising and liquid circulation pattern



Gas phase (i.e. Bubble rising) visualization

Liquid phase natural circulation



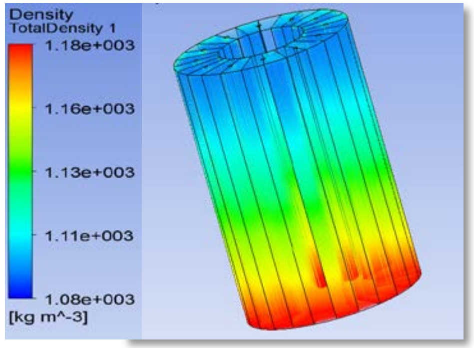
Two-phase fluid dynamic model developed to calculate steady state solution temperature and void profiles for an externally cooled solution vessel

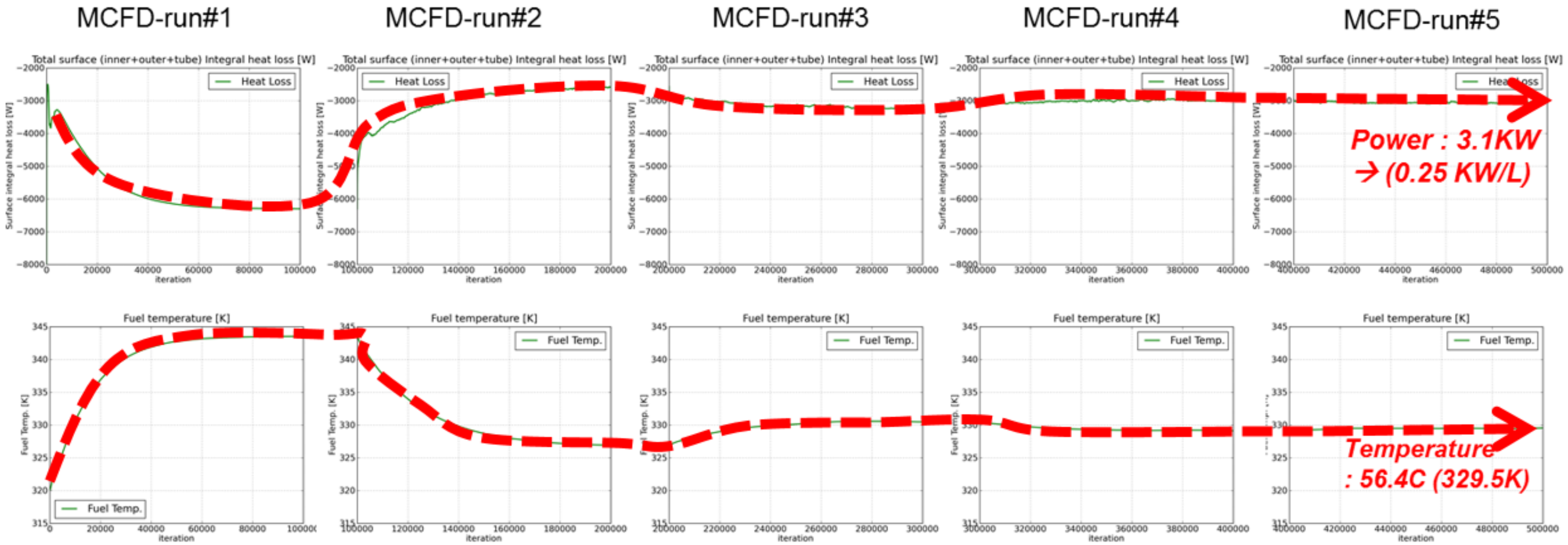
- Solution Density with void profile
  $SD = \rho_{fuel} \times VF_{fuel} + \rho_{gas} \times VF_{gas}$
- Heat transfer by natural convection enhanced by bubble generation
- Non-Uniform volumetric heat and bubble generation profiles
- Temperature-dependent fuel and gas properties
- Results are used to improved HTC for system model (SimApp)

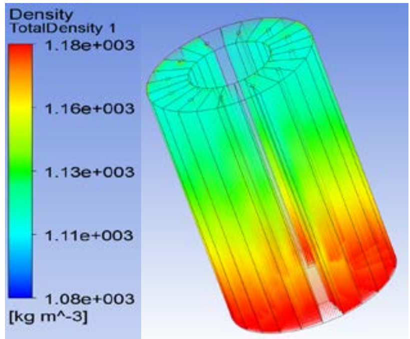
Input & output exchange for coupled calculations

→*Operational parameters (system power, solution temp. & density) are saturated*

CASE-1 (SN=1.458e14s^{-1})	Run#1 (cold start)	Run#2	Run#3	Run#4	Run#5
MCNP ***Cold start condition*** ***(20C, 1185.4 kg/m3)***	Input: 20C, 1185.4 kg/m3 Output: 6.37KW	Input: MCFD-run#1 Output: 2.61KW	Input: MCFD-run#2 Output: 3.27KW	Input: MCFD-run#3 Output: 3.01 KW	Input: MCFD-run#4 Output: 3.10 KW
MCFD Predicted normal operating condition ***(56.4C, 1142.6kg/m^3)***	Input: MCNP-run#1 Output: 70.3C, 1128.6kg/m^3	Input: MCNP-run#2 Output: 53.7C, 1145.1kg/m^3	Input: MCNP-run#3 Output: 57.3C, 1141.8kg/m^3	Input: MCNP-run#4 Output: 55.9C, 1143.0kg/m^3	Input: MCNP-run#5 Output: 56.4C, 1142.6kg/m^3





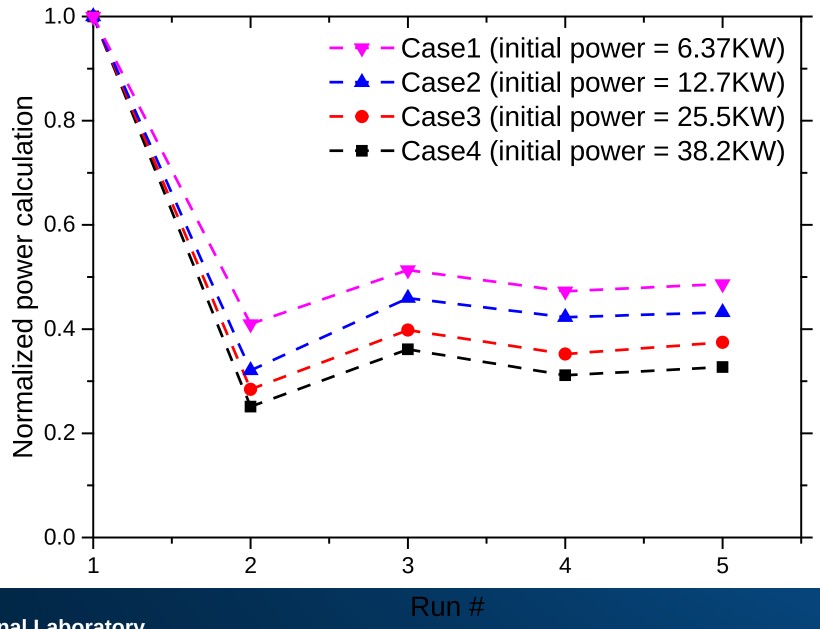


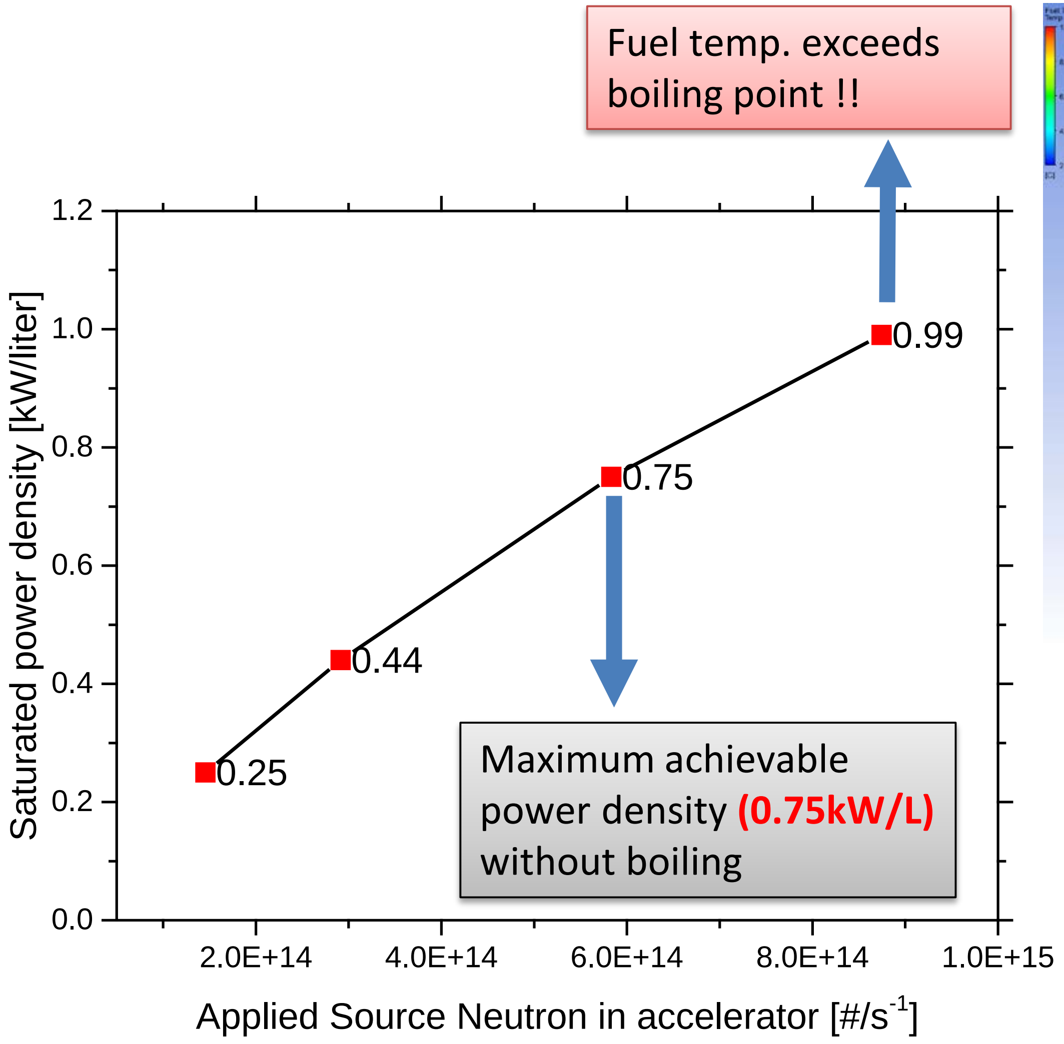
What is the maximum achievable system power density?

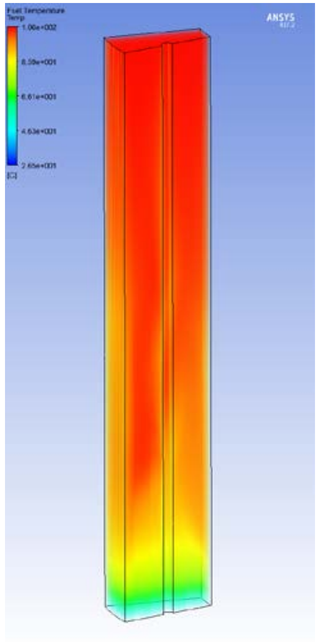
→ Task #1 : Identify Maximum achievable power without boiling

Test matrix for Source Neutron case study

	Source Neutron	Predicted saturated power (initial → saturated)
CASE1	1.458e14 [s^{-1}]	6.37 → 3.10KW **(0.25 kW/L)**
CASE2	2.916e14 [s^{-1}]	12.7 → 5.49KW **(0.44 kW/L)**
CASE3	5.832e14 [s^{-1}]	25.5 → 9.41KW **(0.75 kW/L)**
CASE4	8.745e14 [s^{-1}]	38.2 → 12.5KW **(0.99 kW/L)**







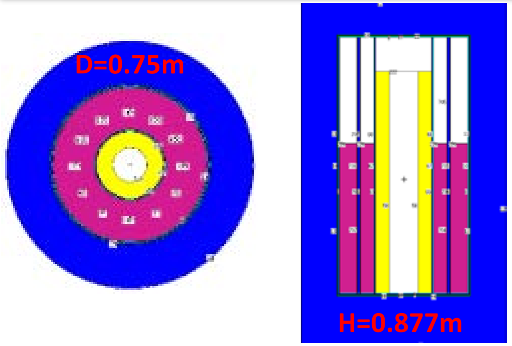
What is a favorable design for enhanced performance and safety?

→Task #2 : Vessel configuration study for optimal design

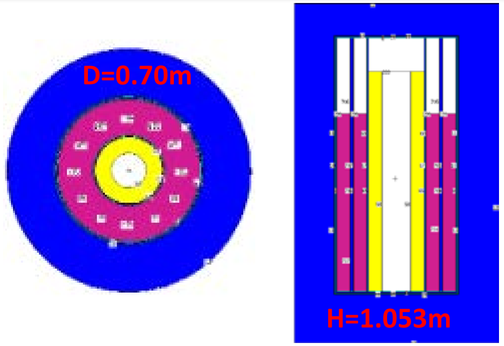
Note identical SN applied
Case3 : 5.83e14 [s^{-1}]

Keff (0.98) and Total solution volume (300L) remain same

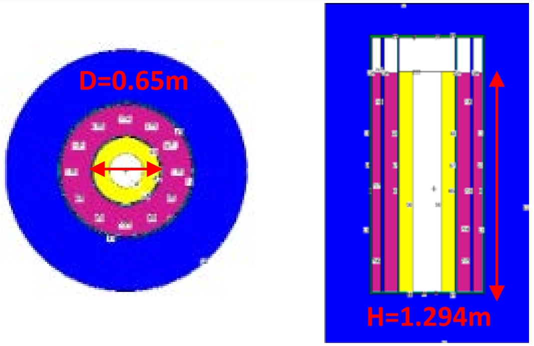
Aspect Ratio (Height/Diameter) = 1.17
Saturated power: 3.00KW/12.9L = 0.2325kW/L
Saturated temperature: 334.55K

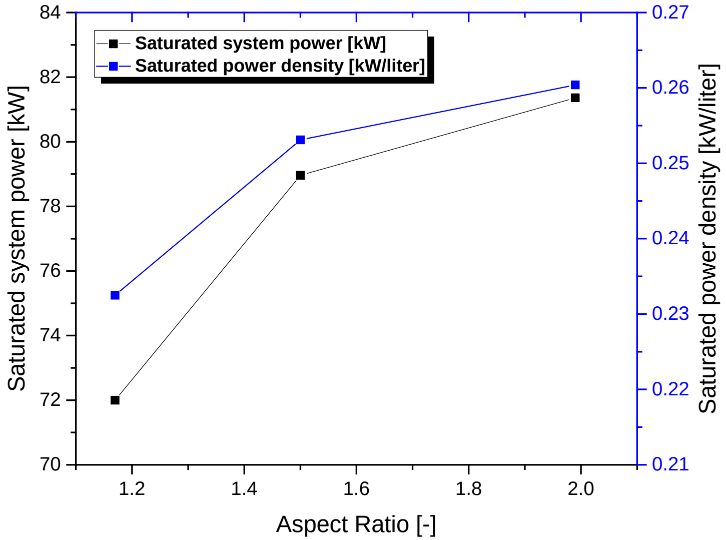


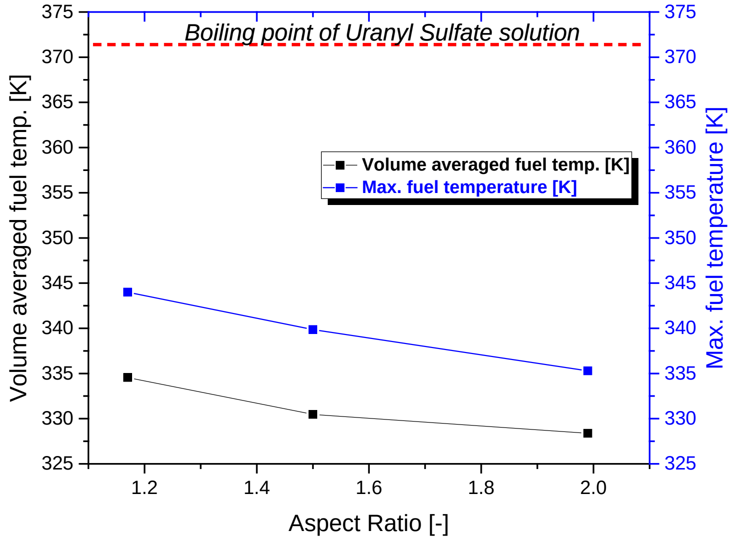
Aspect Ratio (Height/Diameter) = 1.5
Saturated power: 3.29KW/13.0L=0.2531kW/L
Saturated temperature: 330.47K



Aspect Ratio (Height/Diameter) = 1.99
Saturated power: 3.36KW/12.9L=0.2604kW/L
Saturated temperature: 328.38K





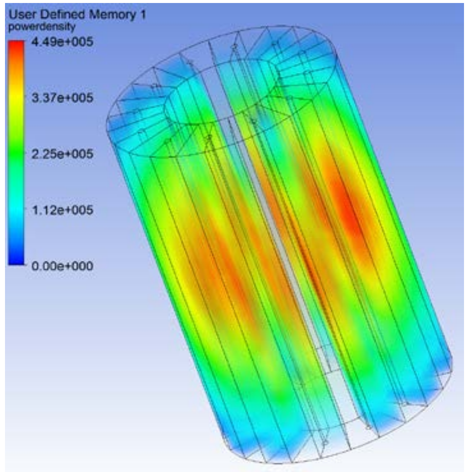


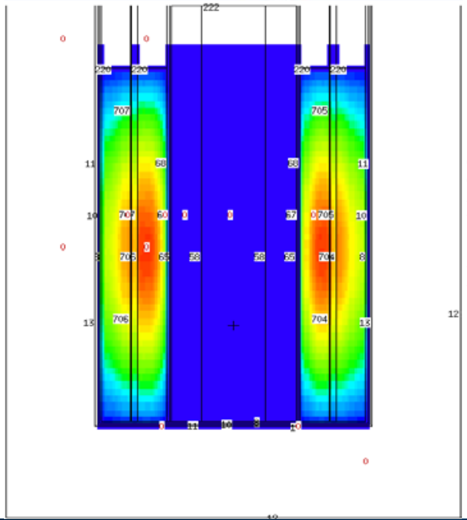
A higher power with lower fuel temperature appears at a slender-shape vessel

What is the most practical coupling method?

→Task #3 : Incremental coupling methodology development & demonstration

Method-I
- Single-cell approach
- Constant Height

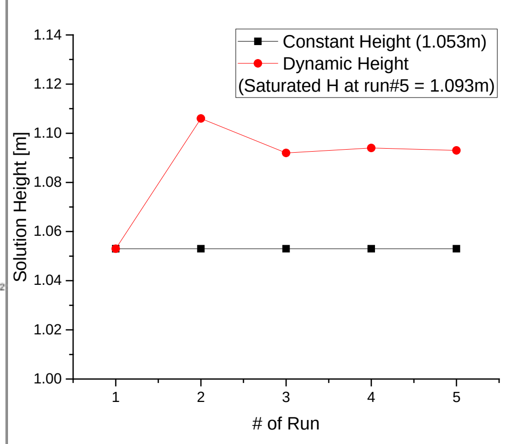




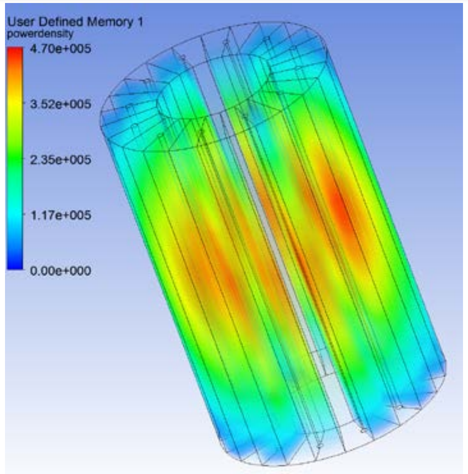
Fissile mass balance concept

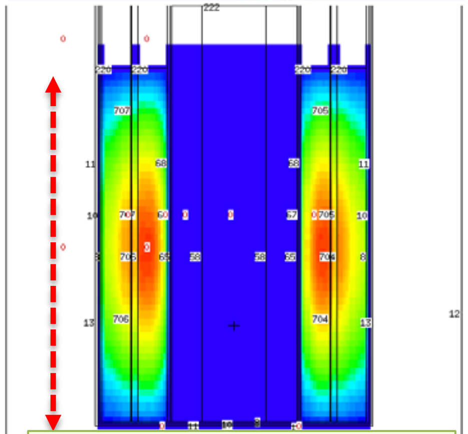
$$V_1\rho_1 = V_2\rho_2 \ \ (mass\ conservation)$$
$$\therefore H_2 = H_1\frac{\rho_1}{\rho_2}$$

Solution height change



Method-II
- Single-cell approach
- **Height adjustment**

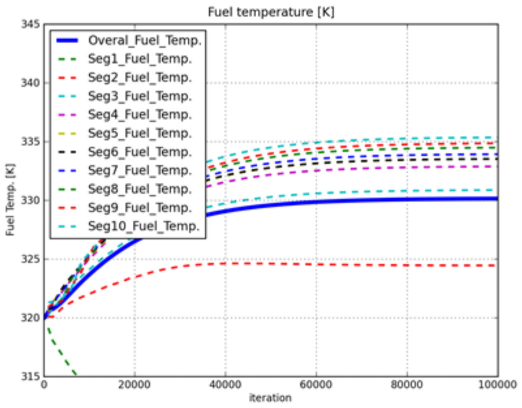




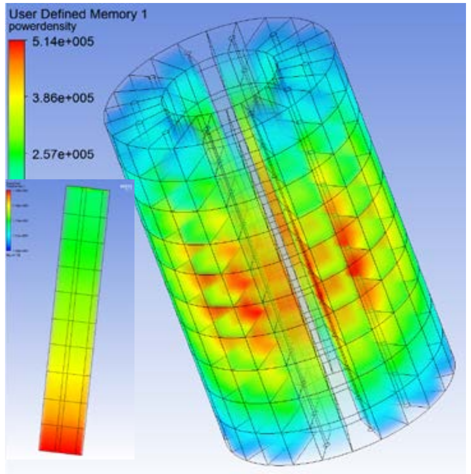
Results:
- Power/ height change

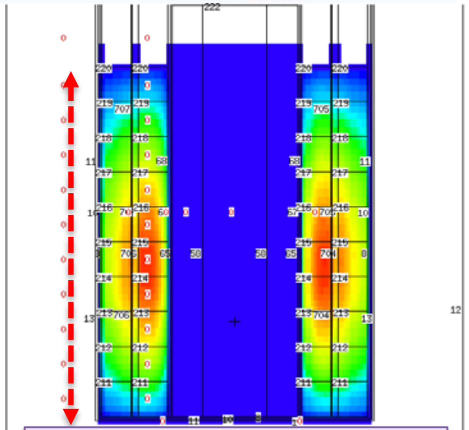
Heterogeneous density

Local solution temp.



Method-III
- **Multi-cell approach**
- **Height adjustment**



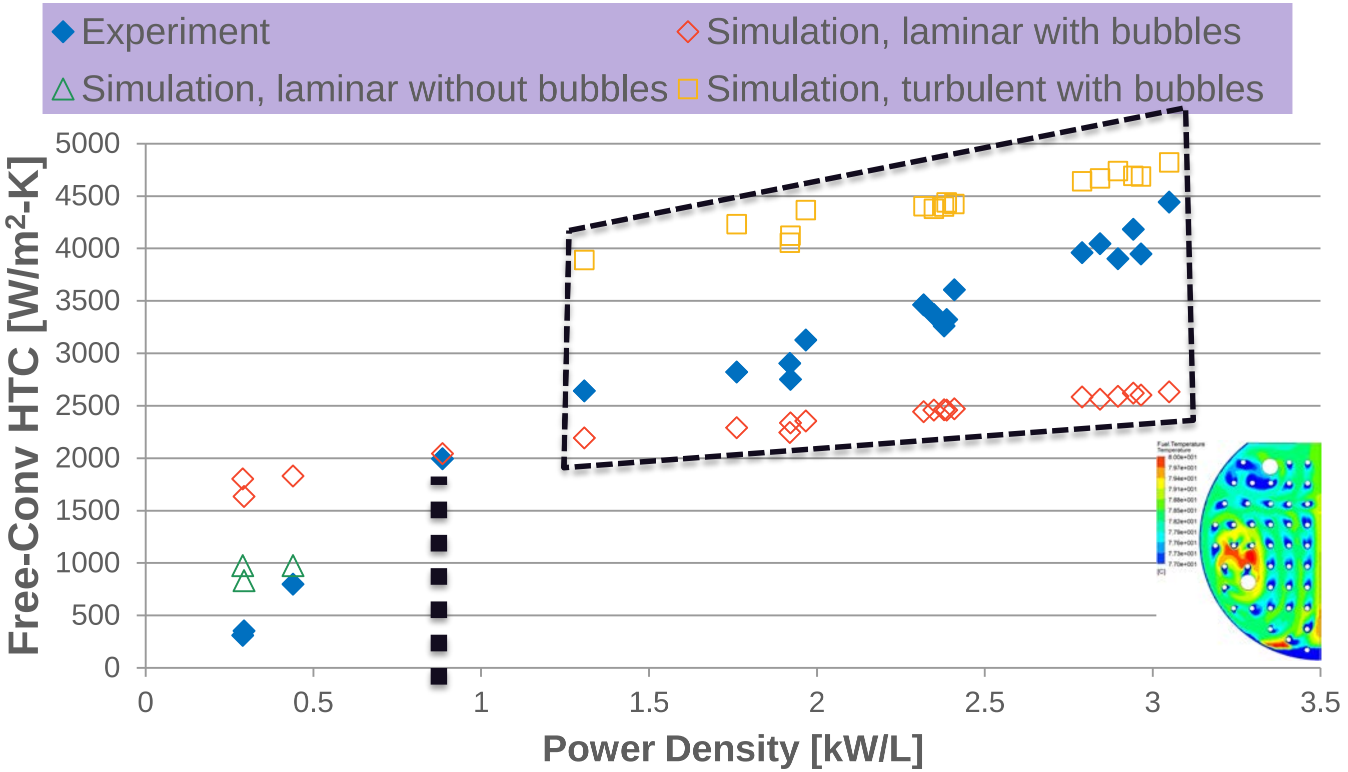


Results:
- Slight power shift

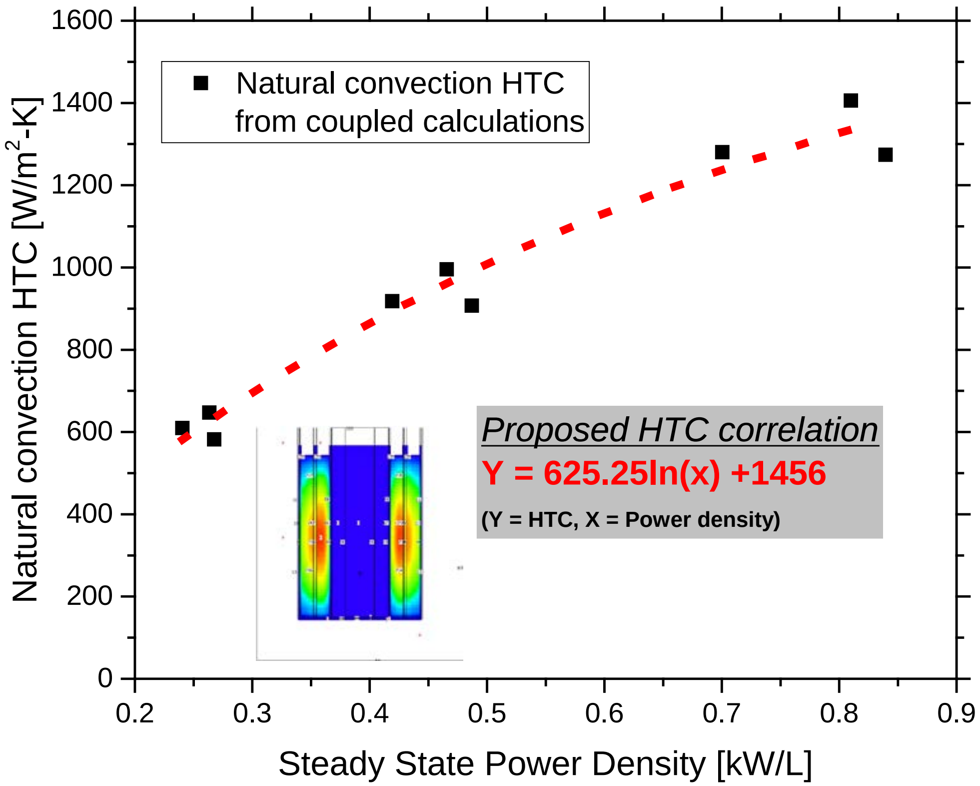
What is the most challenge in fissile solution TH modeling?

→Million Dollar questions: 1)What is the adequate bubble (Radiolytic Gas) size?
2)Bubble induced turbulence mixing effect?

Supo Natural convective HTC calculation by Alex



Natural convective HTC estimation for SHINE



- ***Less than 1kW/L power density region, simulations over-predict the HTC value by a factor of 2~3***
- ***High power density, simulations with different turbulence model provide a window of HTC***
- ***Turbulence mixing and bubble size need to be further investigated for high fidelity calculation***

Mid-year funding R&D activities

1. Nek5000 assessment for natural circulation simulation capability

2. Alternative generic fission solution design with e-beam induced photonuclear reaction

3. ARGUS paper review

Background: What is Nek5000?

❖ **Nek5000 is a fast and scalable high-order CFD solver simulating unsteady incompressible flow and conjugate heat transfer [1]**

- Spectral element method (SEM) based **open source code developed by ANL**
- https://nek5000.mcs.anl.gov/
- Selected as a high-resolution TH tool for **advanced nuclear system modeling in NEAMS**
- https://neams.inl.gov/SitePages/Home.aspx

Figure 2: SEM grid with data, solution, and geometry in terms of a 7^{th} order polynomial

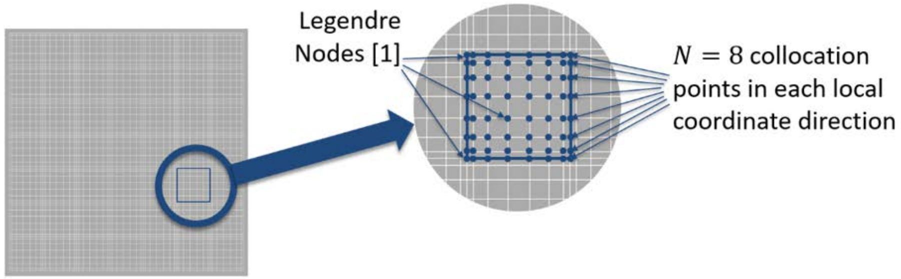


Figure 1: 7x7 (E=49) spectral elements

[1] NEK5000 user manual Version 17.0 Dec 17, 2017. ANL

- Used in **Exascale Computing Project (ECP)** for Coupled Neutronic and CFD simulation regarding Small Modular Reactor design
- https://www.exascaleproject.org/project/exasmr-coupled-monte-carlo-neutronics-and-fluid-flow-simulation-of-small-modular-reactors/

❖ **Accomplishes spatial discretization using Gauss-Legendre quadrature.**

- Elemental data is expanded in terms of Gauss-Lobatto-Legendre (GLL) points, whose weights are based on an $(N-1)^{th}$ order Legendre polynomial.

❖ **Decreases error exponentially by increasing polynomial order.**

- Alternately, traditional FEM decreases error algebraically as the number of elements increases.
- **A key characteristic of the spectral element method is this exponential convergence on the exact solution.**

Test problem for Nek5000 assessment

→ *initial assessment : Single phase Natural Circulation in a box*

Figure 3: Spectral Element Mesh

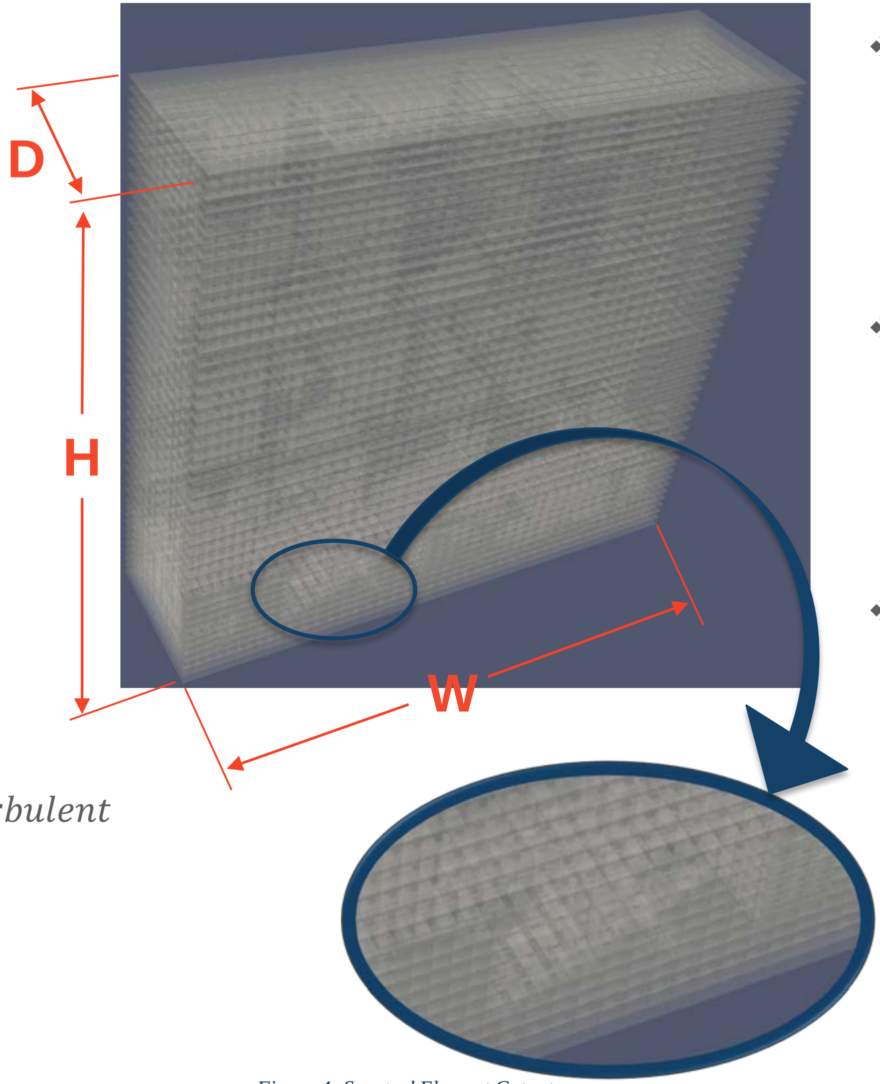


Figure 4: Spectral Element Cutout

❖Conditions:

- Initial
  - $T_0 = 10\ ℃$
- Boundary
  - No-slip Velocity at Walls
  - Heat Flux (q'')
    - $q''_{x_{min}} = -1.0E5x + 1.8E4$
    - $q''_{x_{max}} = -1.0E5x$
  - Thermally Insulated elsewhere
- Boussinesq Approximation
  - $ffy = g\rho(1 - \beta(T - T_0))$
- Rayleigh number
  - $Ra = \frac{g\beta}{\nu\alpha}(T_s - T_\infty)x^3 \sim 7.69 \times 10^7 \therefore turbulent$
- Prandtl number
  - $\mathrm{Pr} = \frac{c_p\mu}{k} = \frac{\nu}{\alpha} \sim 9.5$

❖Geometry:

- Dimensions:
  - Height = Width = $0.18\ m$
  - Depth = $0.05\ m$

❖Mesh

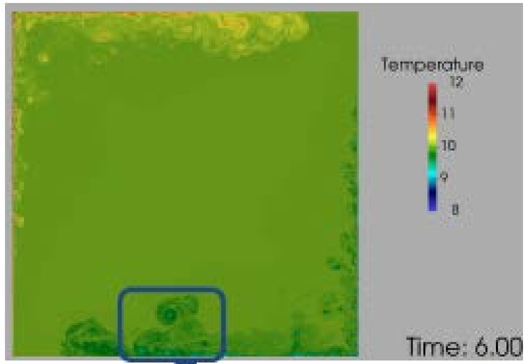
- $EN^3 = 38.72 \times 10^6$ DOF
  - Spectral Elements = $E = 38{,}720$
  - 9^{th}order polynomial = $N = 10$

❖Miscellaneous

- $\Delta t = 0.02$
- CFL = 2.0
- 3^{rd}order Backward Difference Scheme

Preliminary result of Natural Circulation using Nek5000

Fig. 9. q''_L thermal development at t = 6 seconds



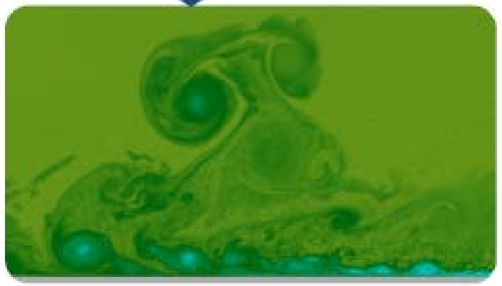
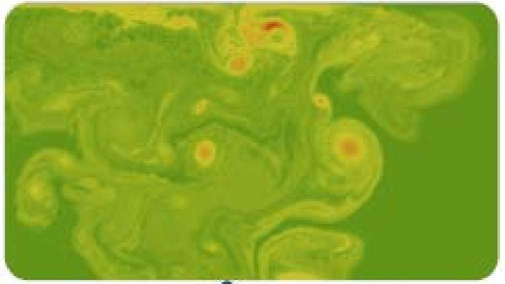


Fig. 10. Cutout of q''_L BC

Fig. 12. Cutout of q''_C BC



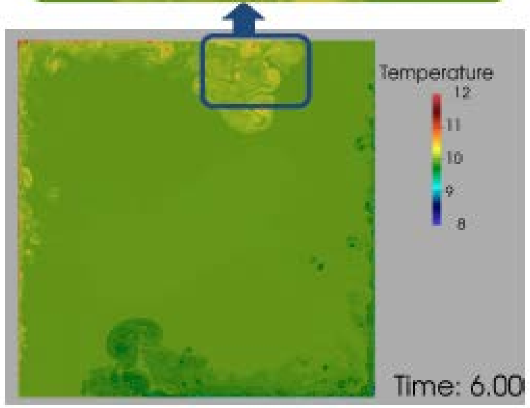
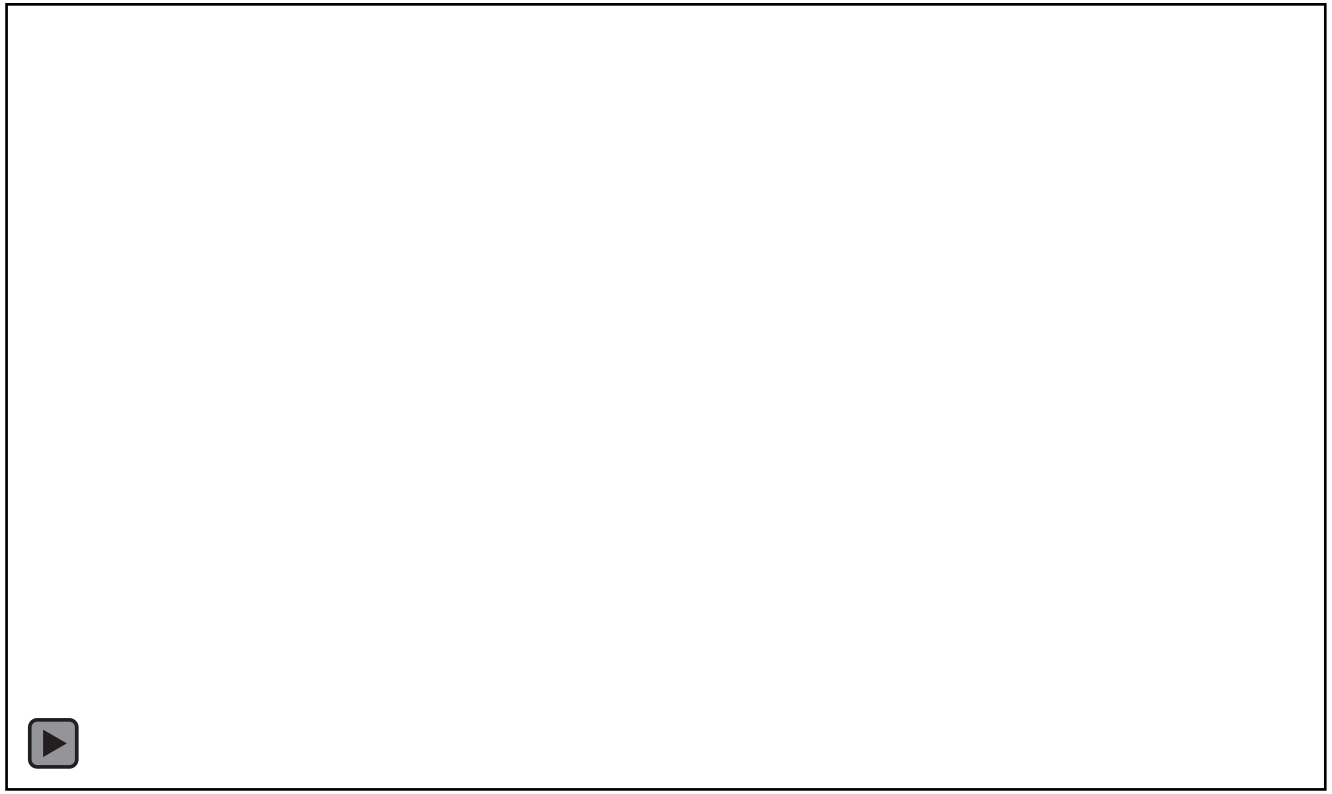


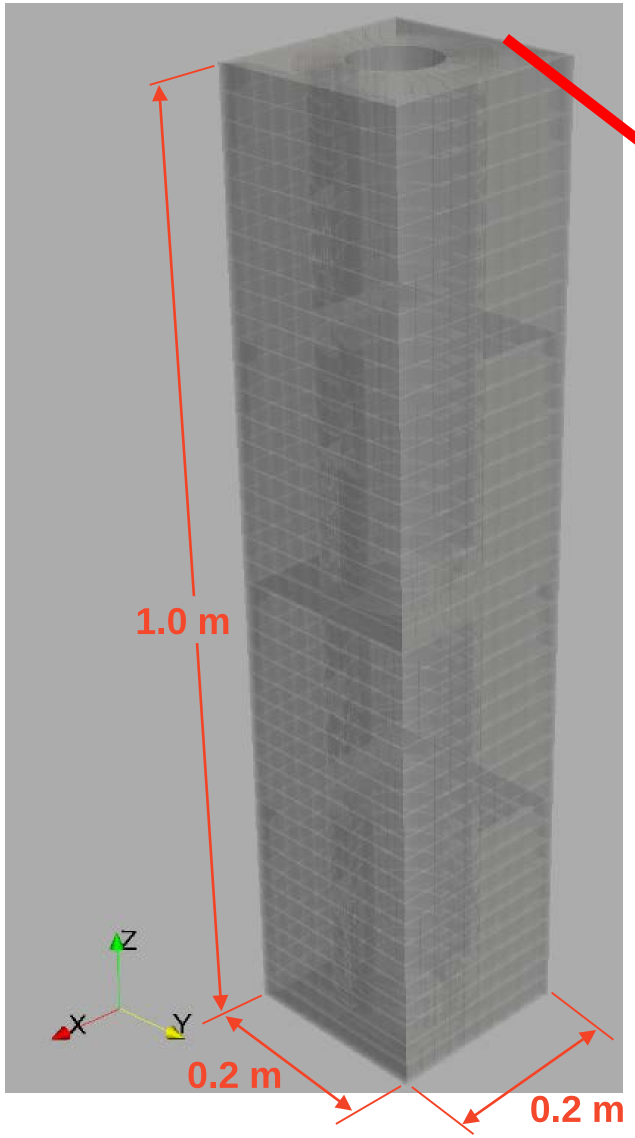
Fig. 11. q''_C thermal development at t = 6 seconds

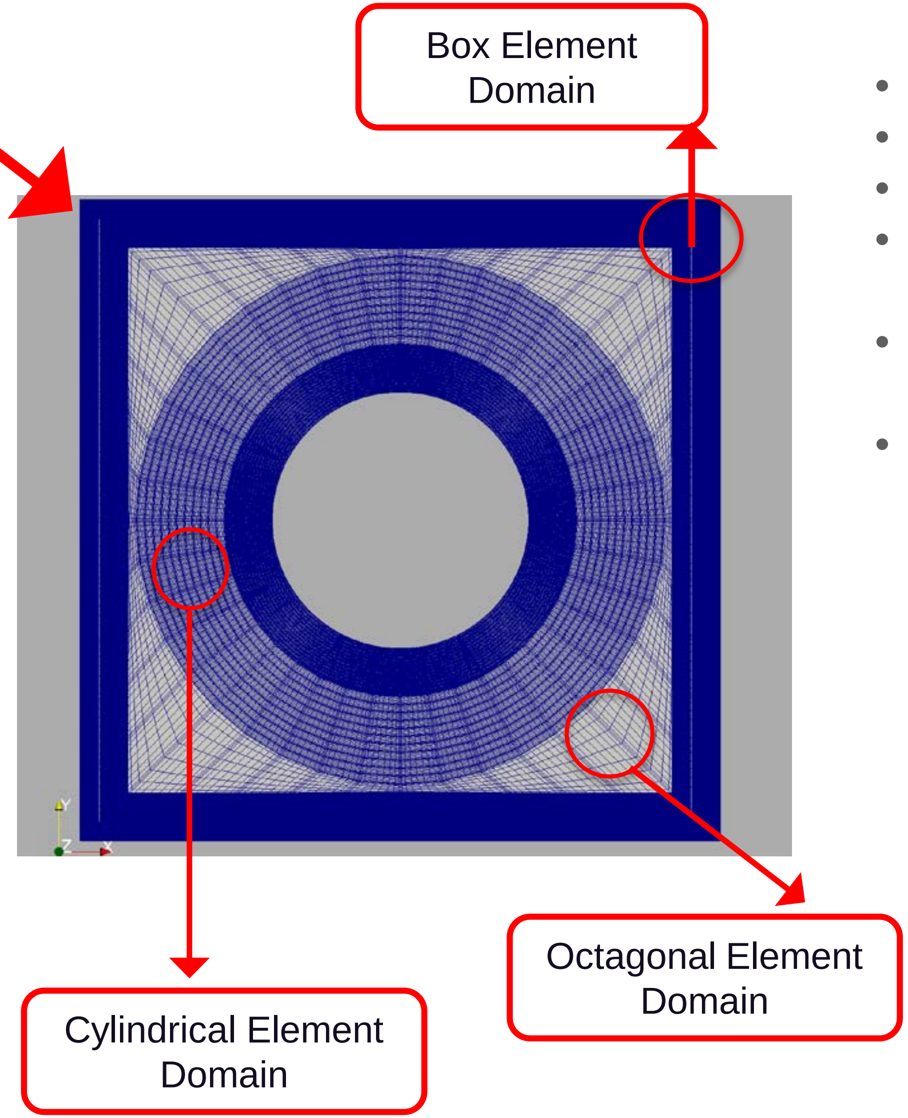


- Temperature evolving behavior in a Natural circulation condition (This simulation is a DNS level calculation)
- Small size thermal eddy is captured using Nek5000

- Positive heat flux (left) and Negative heat flux(right) and no volumetric heat generation are applied for initial assessment in a rectangular box
- 3D transient natural convection with temperature contour

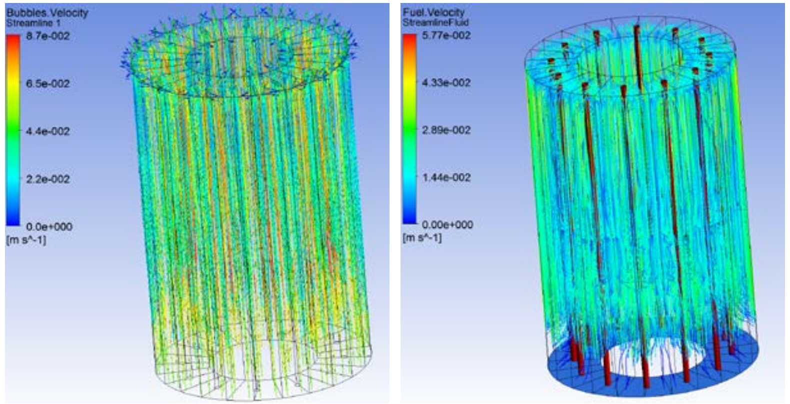
Segmented solution reactor mesh model (1/12th) with Nek5000





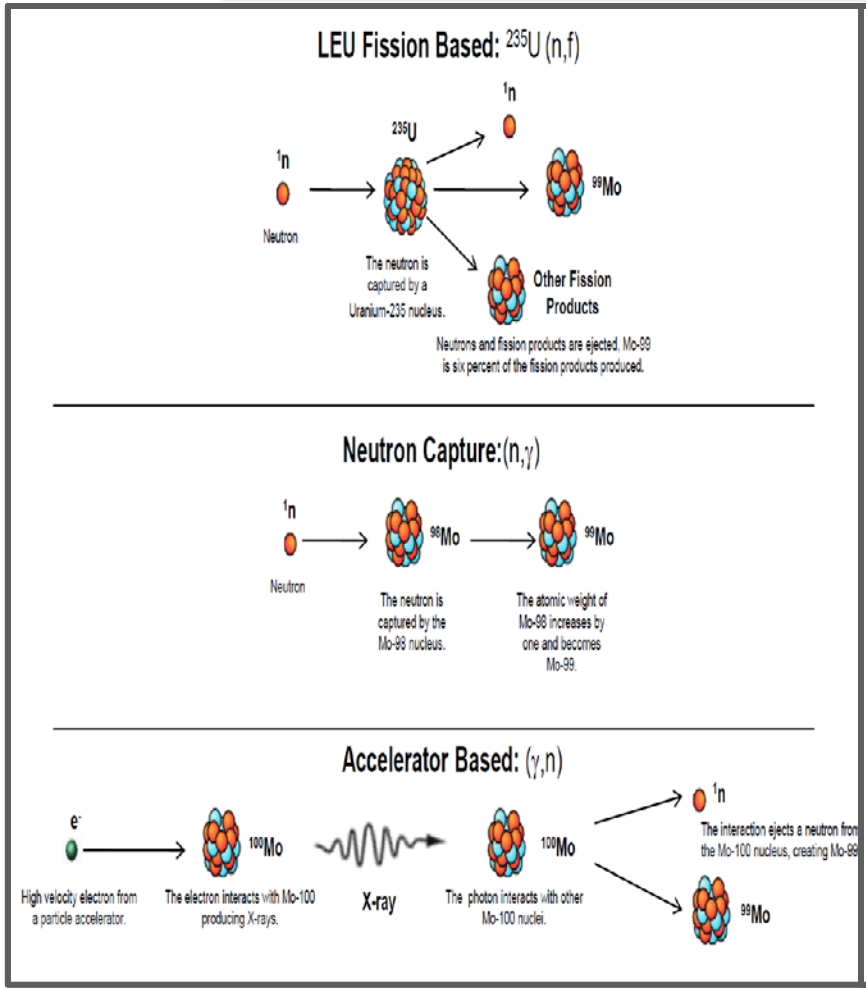
- DNS based Single-phase natural circulation
- 1/12th segmented model with no bubble
- Uniform heat generation rate
- Cooling surfaces
  - A tube and inner/outer channels
- HTC calculation and benchmark with existing CFD result
- Perform a feasibility study of Nek5000 for Moly production application

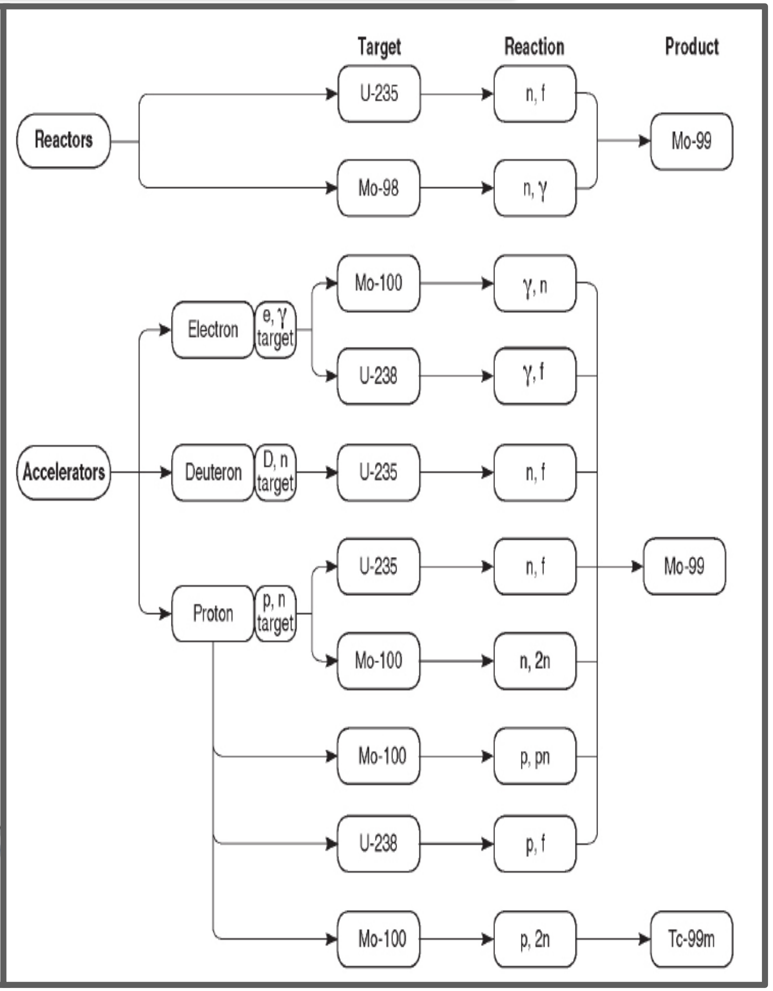
A full scale TH modeling for a generic solution vessel system



Accelerator based photonuclear (γ,n) reaction for Mo99 production

Mo99 production methods using reactors and accelerators





Figures taken from “Moly-99 for medical imaging” (2016) The National Academies Press
https://www.ncbi.nlm.nih.gov/books/NBK396169/pdf/Bookshelf_NBK396169.pdf

Traditional Mo99 production method

Three Mo99 production methodologies
- LEU fission based
- Neutron Capture
- Accelerator based

Two approaches for system design
- Reactor based system
- Accelerator based system

Alternative Mo99 production method

→ Accelerator + Fissile solution system

Accelerators used as **neutron generators**
- DT accelerator or DD
  - 14Mev mono energy source of N
- E-beam accelerator
  - Bremsstrahlung X-ray
  - A broad energy spectrum of N

Horizontal fissile solution design with e-beam accelerator

→*A generic system design for initial assessment*

Initial specifications for a "generic" system design

Neutron source and Vessel configuration

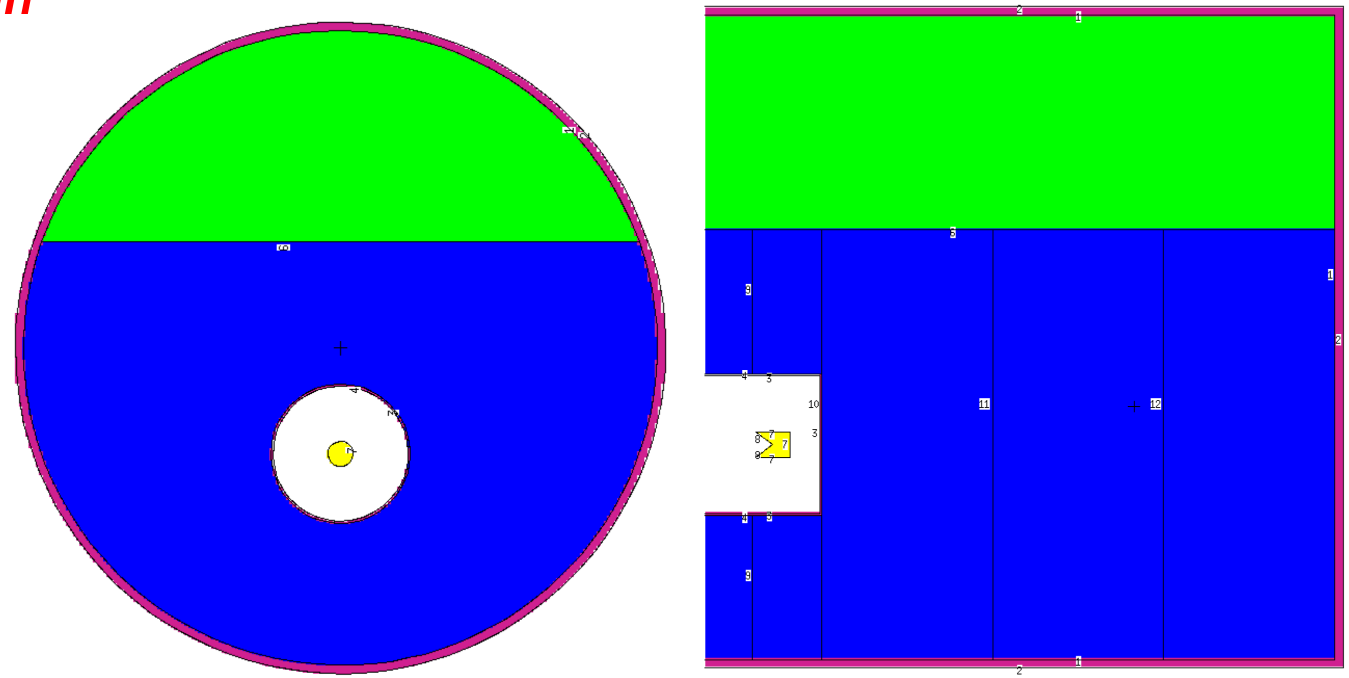
- E-beam accelerator (40 MeV, 100kW)
- Horizontal accelerator orientation
- **Natural U for photonuclear reaction (yellow)**
- Uranyl nitrate/ Uranyl sulfate solution both tested (blue)
- Radiolytic gas stripping by inert gas (green)

Cooling configuration

- No cooling channel in the solution
- External cooling by forced convection

Note that further study is required to identify the optimal baseline system using MCNP and thermal hydraulic calculation

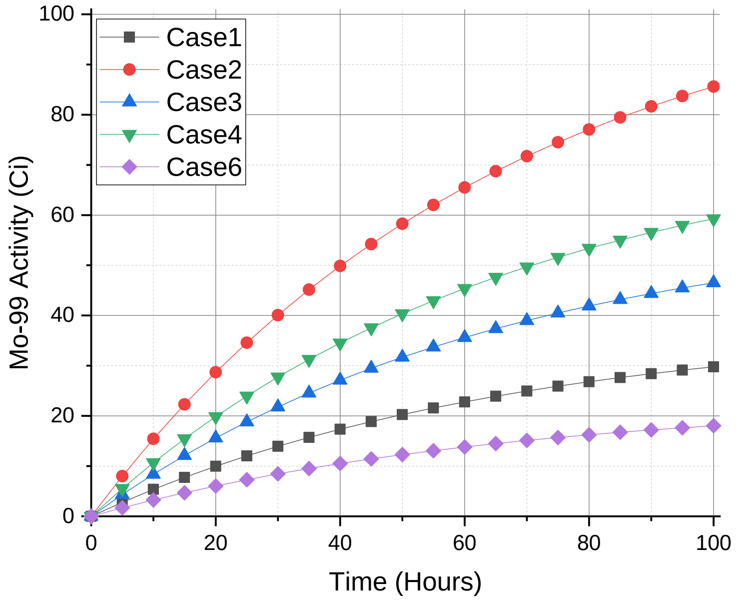
Side views of a proposed system



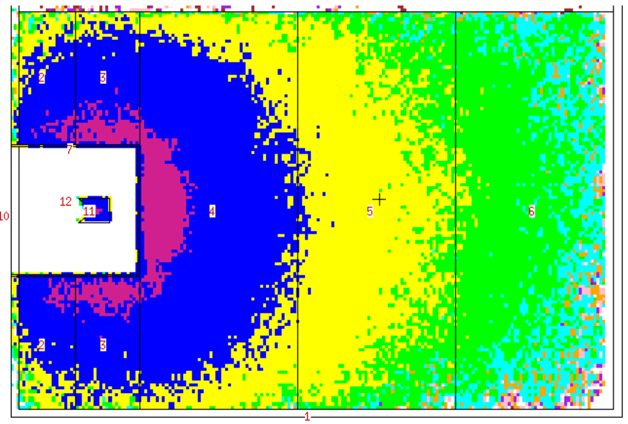
- *Initial system design is based on a discussion with GMIS*
- *A kick-off meeting was held on Aug. 21*

Preliminary results for initial system assessment

→ *Various scenarios study with different solution conditions*



Mo-99 activity calculation over 100 hrs. of irradiation with various solution conditions at accelerator specification 1



Fission rate calculation with MCNP6.2

Accelerator specification 1 *(40Mev & 100kW)*

	Uranyl type	U235%	UO2(NO3)2 Concentration	solution base	Sol. Den. [g/cc]	Keff	Mo99@100hrs	
Case1	UO2(NO3)2	Nat. Uranium	1.015mole/liter(400g/L)	H2O	1.31	0.1981	30	Curies
Case2	UO2(NO3)2	Nat. Uranium	1.015mole/liter(400g/L)	D2O	1.31	0.4163	85	Curies
Case3	UO2(NO3)2	Nat. Uranium	1.523mole/liter(600g/L)	H2O	1.47	0.2607	46	Curies
Case4	UO2(NO3)2	Nat. Uranium	2.03mole/liter(800g/L)	H2O	1.64	0.3217	60	Curies
Case5	UO2(NO3)2	3%_Enriched	2.03mole/liter(800g/L)	H2O	1.64	0.8591	724	Curies
Case6	UO2SO4	Nat. Uranium	0.58mole/liter(215g/L)	H2O	1.185	0.12	18	Curies

Test 1 : H_2O vs D_2O effect (Case 1&2)

- Heavy water scenario leads to high production due to low absorption X-section (better thermalizing)
- Heavy water produce Tritium gas which is not favorable for off-gas system design

Test 2 : $UO_2(NO_3)_2$ concentration effect (Case 1, 3, &4)

- Higher concentration produces linearly more Mo-99 activity

Test 3 : Enrichment effect (Case 4 &5)

- 3% enriched solution make the solution system still subcritical (K_{eff} = 0.85)

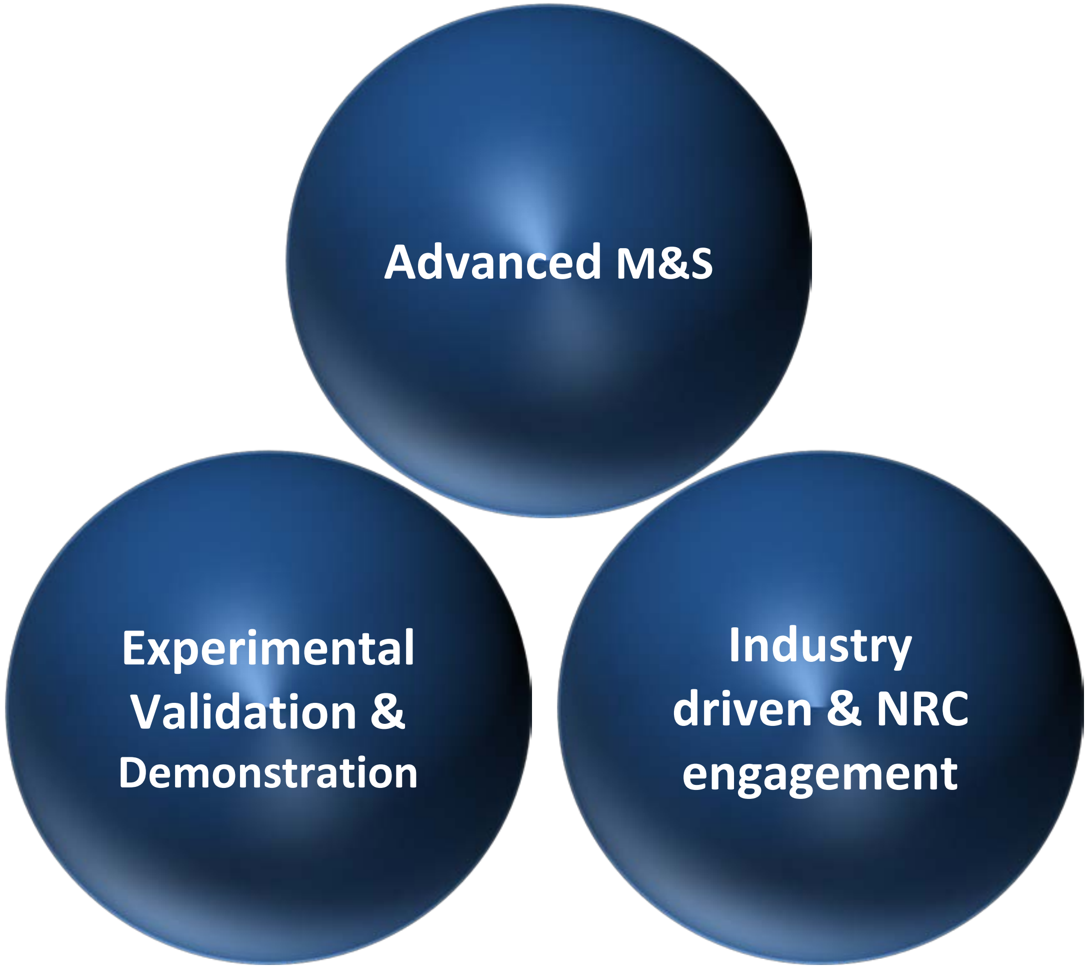
Test 4 : Uranyl Nitrate vs Uranyl Sulfate effect (Case 1&6)

- Observed that no big difference in production at the similar concentration, but Nitrate has higher solubility limit

Accelerator specification 2 *(35Mev & 10kW)*

Case11	UO2(NO3)2	Nat. Uranium	1.015mole/liter(400g/L)	H2O	1.31	0.1981	3.4	Curies
Case12	UO2(NO3)2	Nat. Uranium	1.015mole/liter(400g/L)	D2O	1.31	0.4163	9.7	Curies
Case13	UO2(NO3)2	Nat. Uranium	1.523mole/liter(600g/L)	H2O	1.47	0.2607	5.2	Curies
Case14	UO2(NO3)2	Nat. Uranium	2.03mole/liter(800g/L)	H2O	1.64	0.3217	6.8	Curies
Case15	UO2(NO3)2	3%_Enriched	2.03mole/liter(800g/L)	H2O	1.64	0.8591	82.5	Curies
Case16	UO2SO4	Nat. Uranium	0.58mole/liter(215g/L)	H2O	1.185	0.12	2.1	Curies

A Success path for Nuclear Technology development



Thanks for your attention
Q&A

S. Jun Kim (LANL)

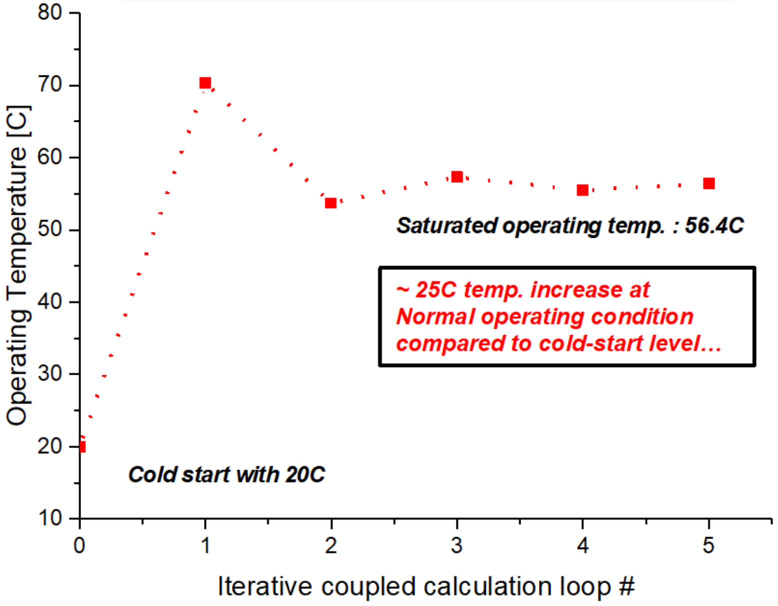
Back-up slides…

Back-up slides…

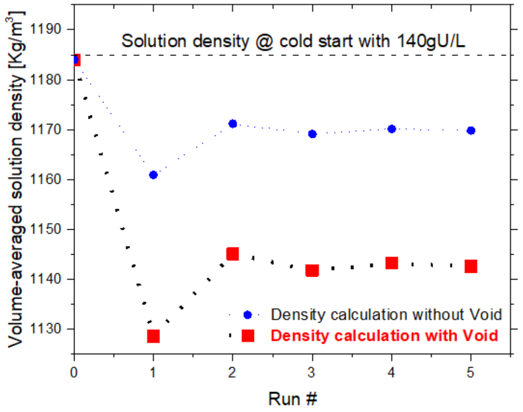
Prediction of operational solution thermal hydraulics

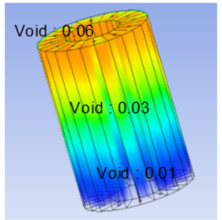
(Solution temperature and solution density with void evaluation)

Solution temperature convergence

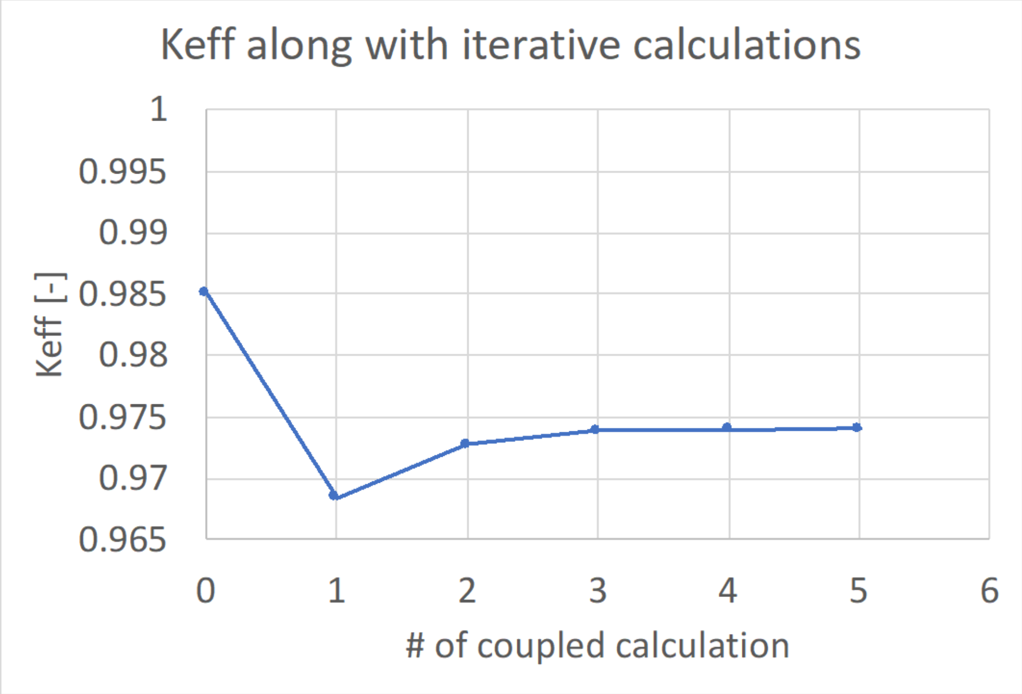


Density convergence





Accurate void profile calculation in MCFD results in realistic density and power calculation in fissile solution vessel



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