

Irradiation Activities at ORNL for Supporting R&D on ^{99}Mo Production

Nesrin Cetiner

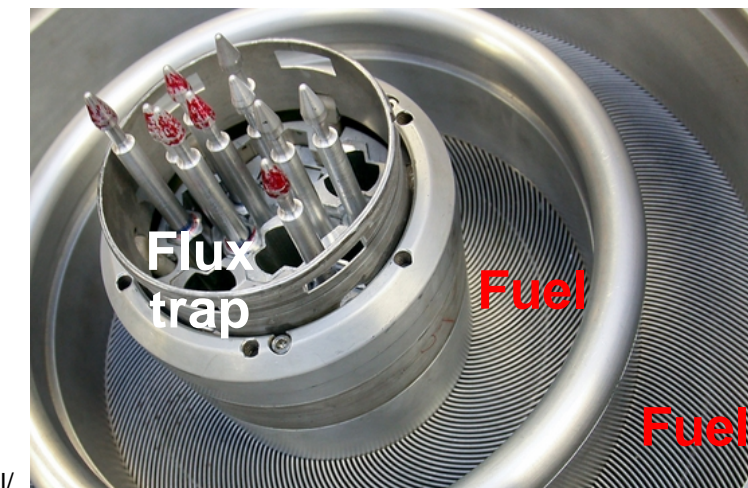
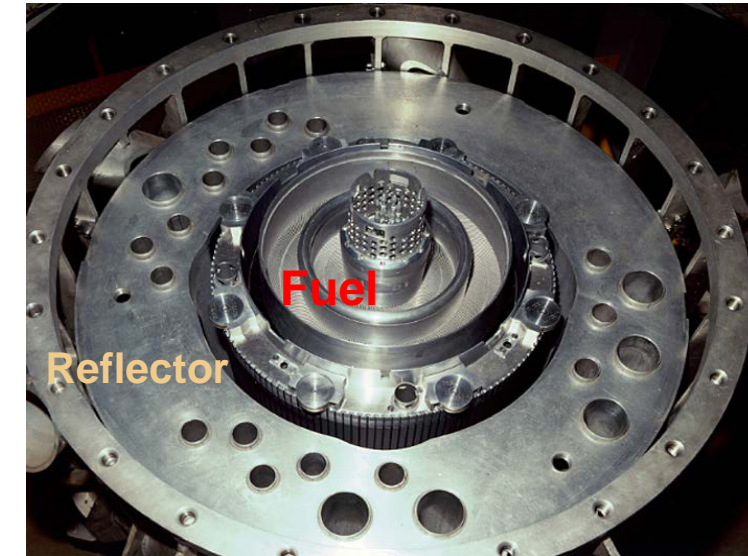
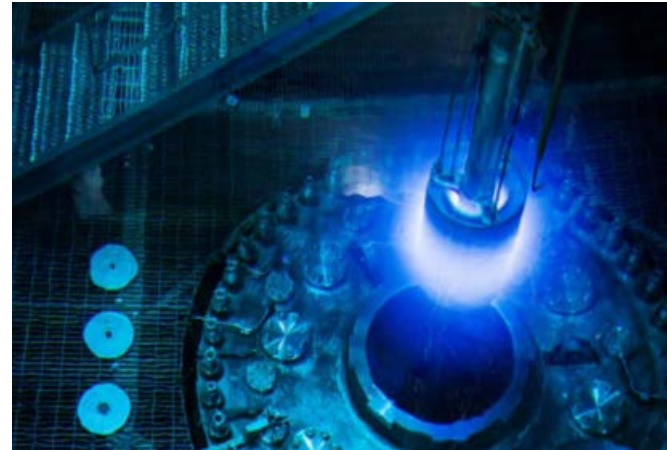
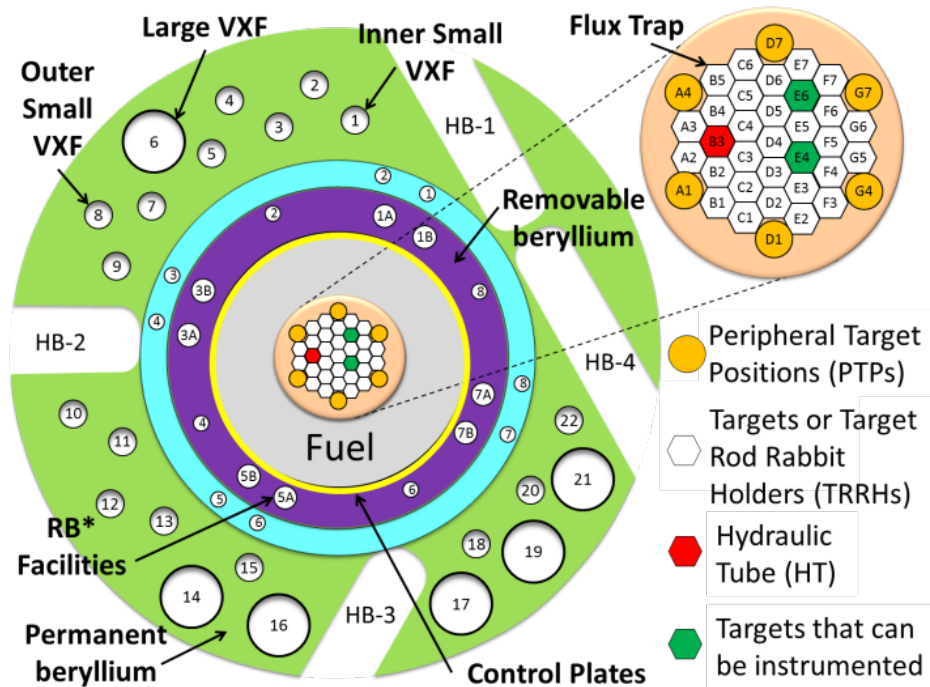
Chris Bryan

2018 Mo-99 Topical Meeting

Sept 23-26, 2018

The High Flux Isotope Reactor (HFIR)

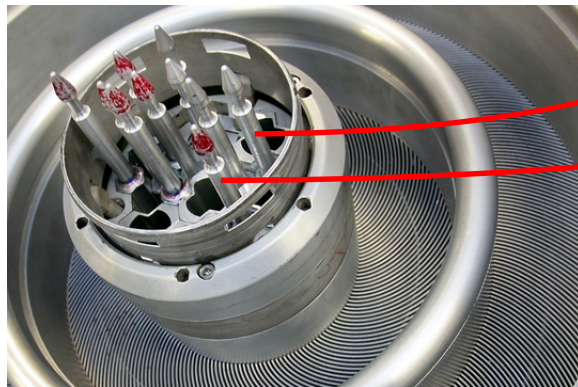
- Pressurized (470 psi), flux-trap type, light water cooled and moderated
- HEU fuel (U_3O_8 dispersed in aluminum) with involute plate geometry
- Operates at a steady state 85 MW
- Experiments in flux trap or beryllium reflector
- 24 days/cycle, 6–7 cycles/year



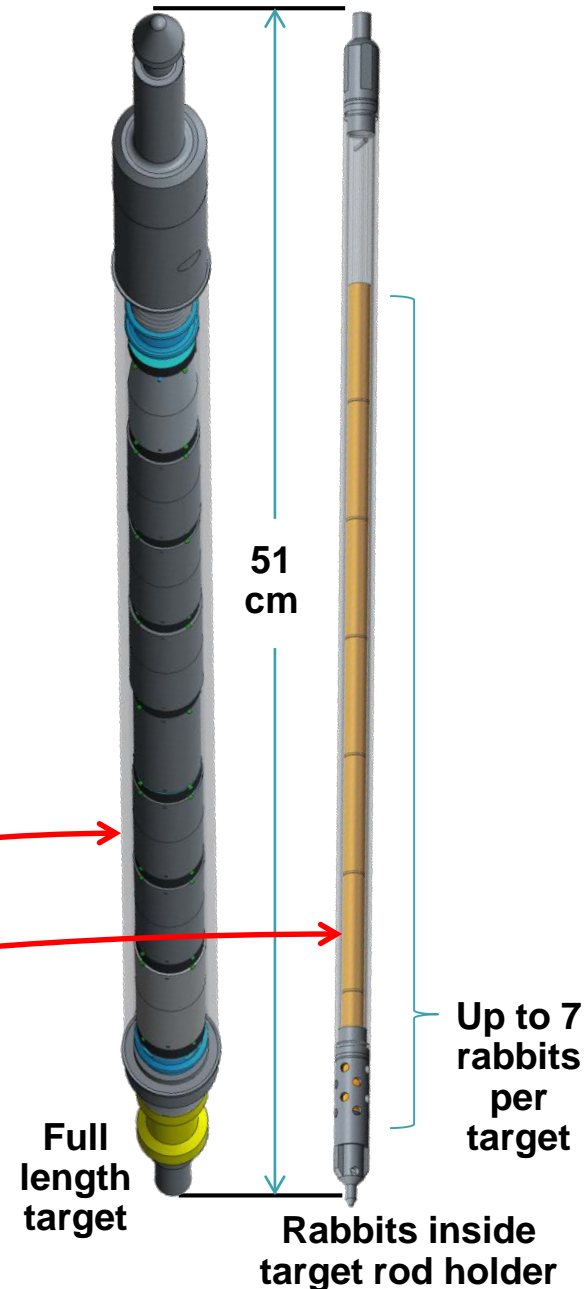
- Thermal flux of 2.1×10^{15} n/cm²/s
- Fast flux of 1.1×10^{15} n/cm²/s

Flux Trap Experiments—Rabbits and Targets

- High fast flux: 1.1×10^{15} n/cm²/s
- High thermal flux: 2.1×10^{15} n/cm²/s
- Small diameter: $\varnothing 9.5\text{--}11.3$ mm
- 12.6 dpa/CY (steel)
- ~150 available rabbit positions, none instrumented
- ~15 target positions, 2 instrumented
- Design temperature (60–1200°C)
- Cost: \$30–\$400K

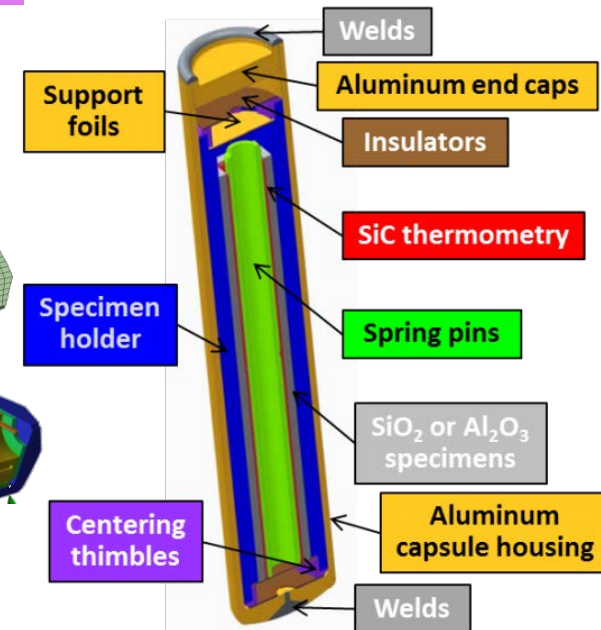
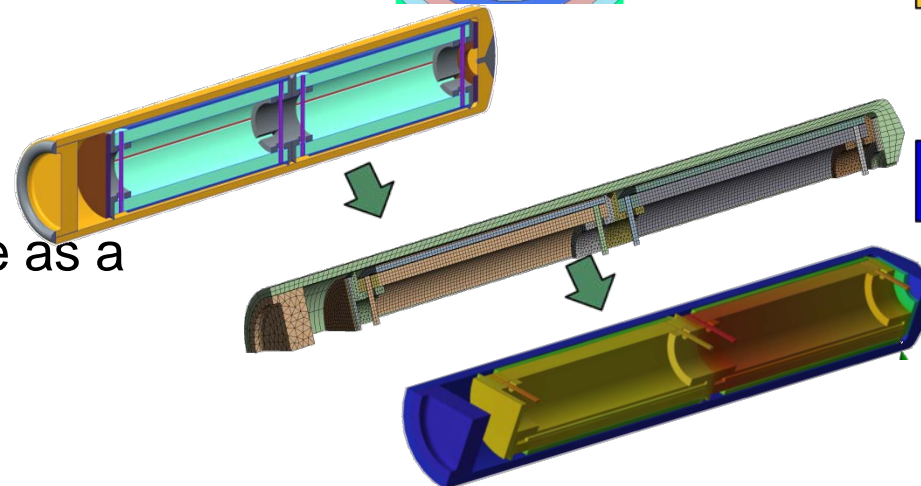
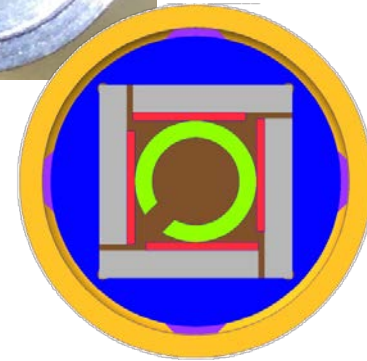
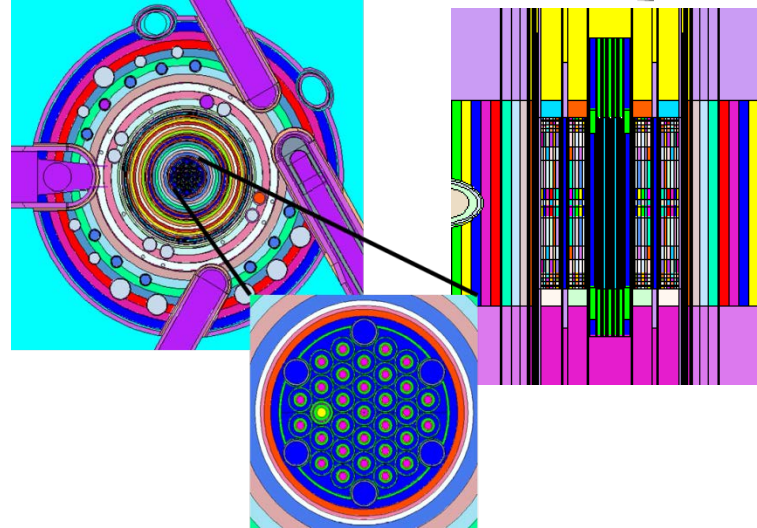
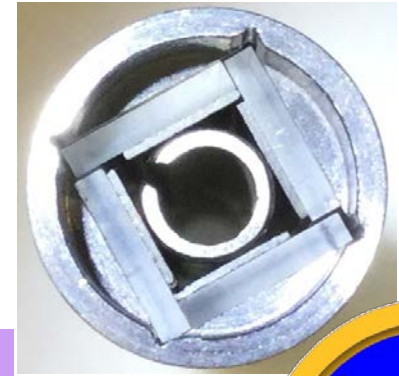


Picture of targets in flux trap: <https://www.comsol.com/blogs/converting-high-flux-isotope-reactor-leu-fuel/>

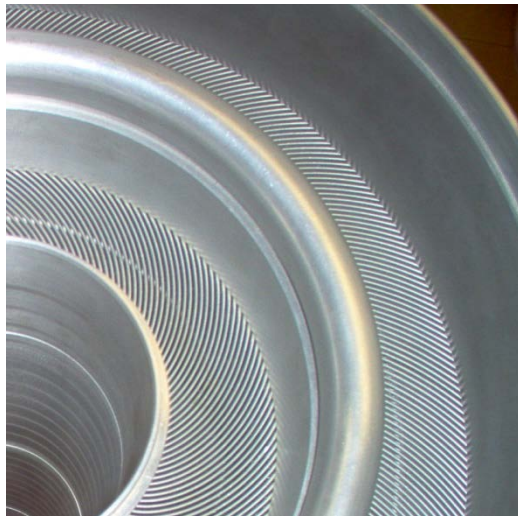


Irradiation Design and Methodology

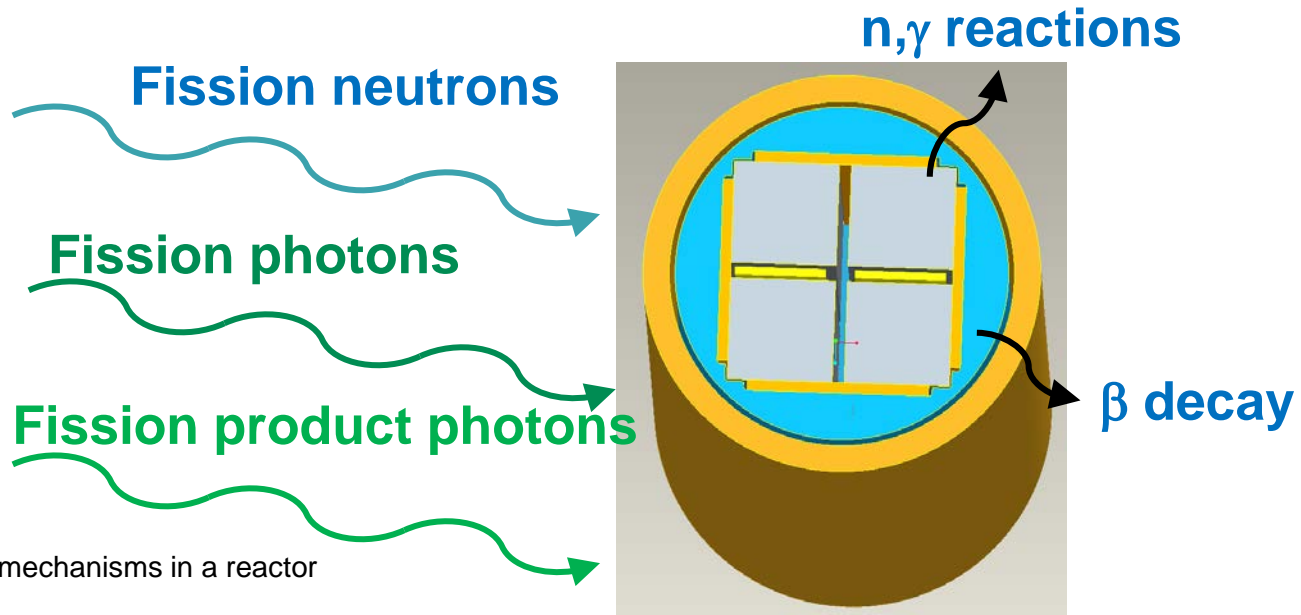
- **Mechanical design (Creo)**
 - Detailed holders with cutouts for specimens and instrumentation
 - Welded and hermetically sealed containment vehicles
- **Neutronics analysis (MCNP + ORIGEN)**
 - Neutron/gamma heating rates
 - Fuel burnup, displacement damage
 - Neutron activation + decay heating
- **Thermal analysis (ANSYS)**
 - Multi-body heat transfer
 - Thermal/mechanical coupling
 - Small gas gaps that open or close as a result of swelling, creep, thermal expansion, etc., used to control temperature



Neutronics Estimate Heat Generation Rates



Visualization of nuclear heat generation mechanisms in a reactor



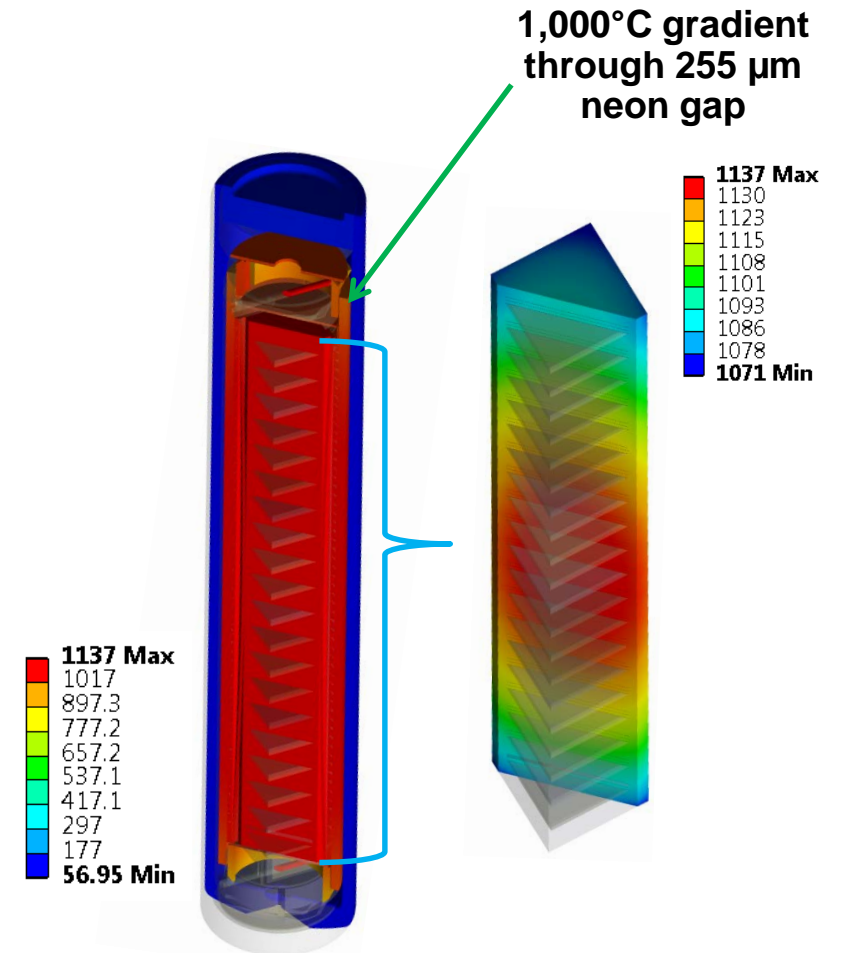
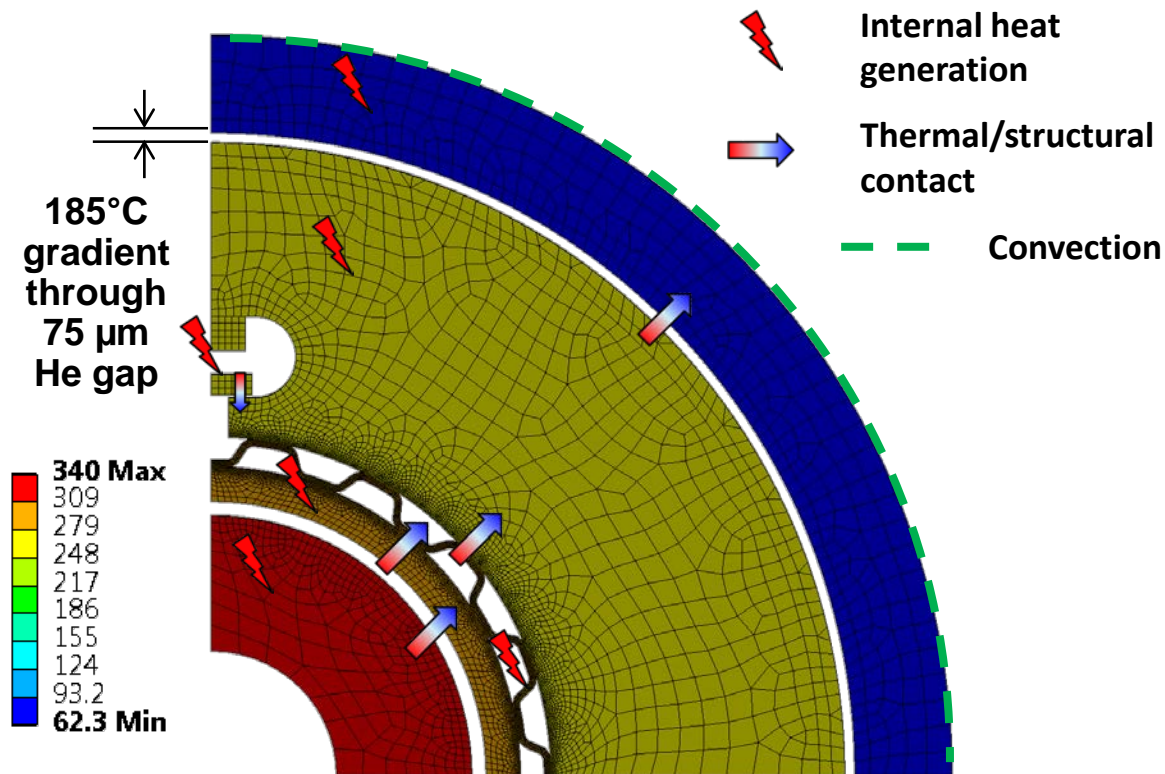
- Heat generation rates comprise more than just gamma heating
- The axial profile is strong but relatively independent of material (0–45%)
- The radial profile in the flux trap is weaker but material dependent (~10–15%)
- The radial profile in the reflector can be large (possibly by orders of magnitude)

Relative contributions to the total heat generation rate (W/g)

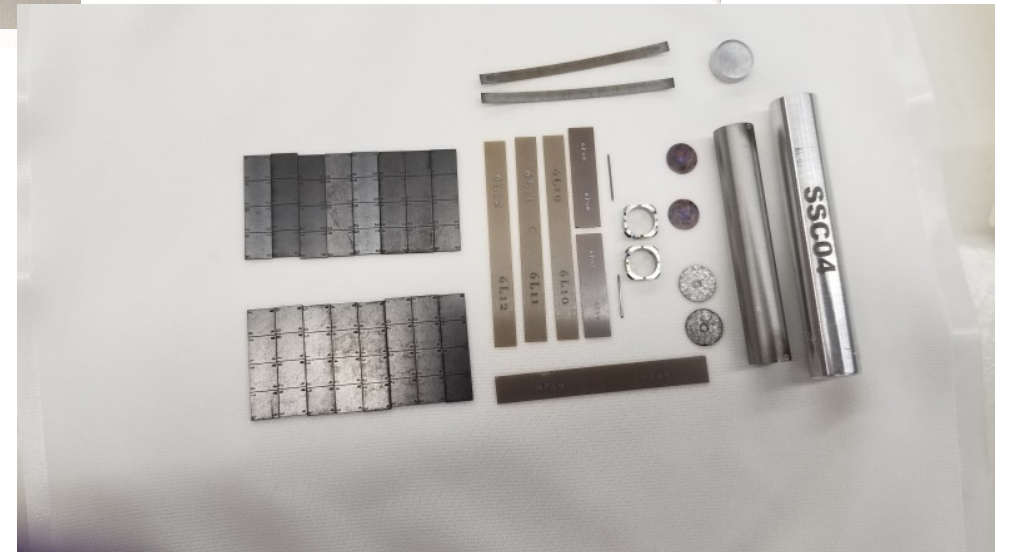
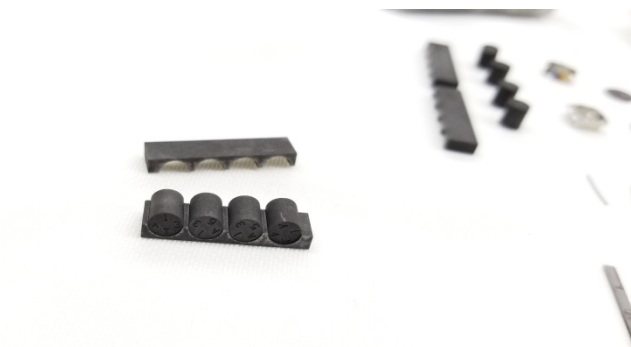
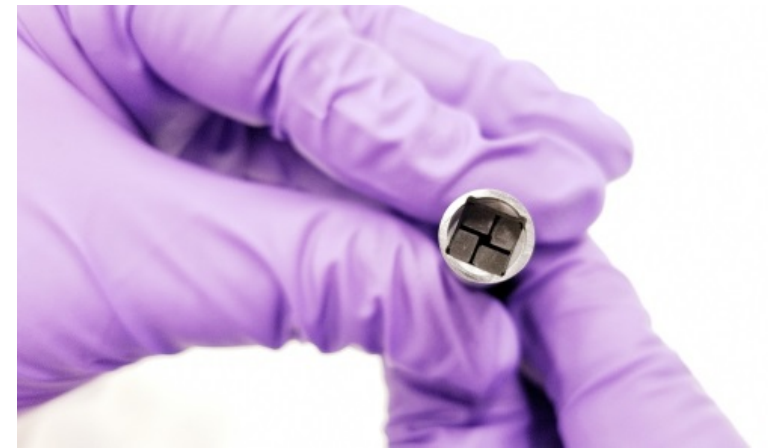
	Vanadium	Graphite
Core fission neutrons	0.3 (1%)	3.3 (11%)
Core fission photons	22.5 (48%)	17.7 (59%)
Core fission product photons	11.8 (25%)	8.8 (30%)
Local β decay	12.1 (26%)	—

Thermal Modeling

- **Extremely high power density in HFIR core**
 - High neutron and gamma heating rates: 30 W/g to >100 W/g in flux trap
 - Large spatial gradients in temperature and dose without proper design
- **Temperature controlled by varying fill gas conductivity and size of gas gaps**
 - Fill gas is usually He, Ne, Ar, or a mixture of these gases
 - Gaps on the order of micrometer to millimeter
 - Thermal/structural coupling to account for thermal expansion and/or swelling



Experiment Assembly



Materials Irradiations for SHINE Medical Technologies

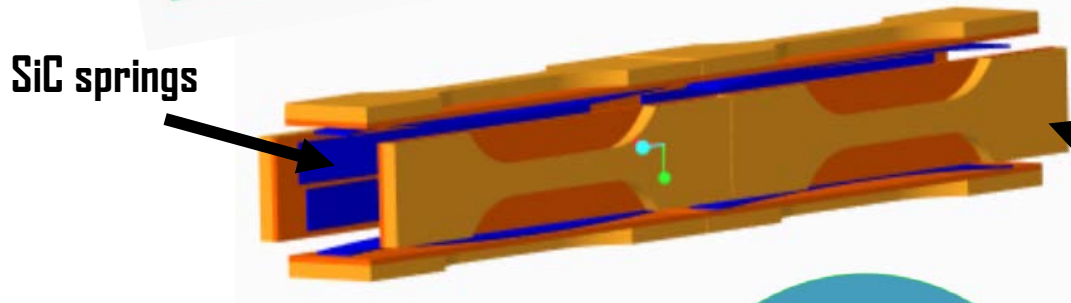
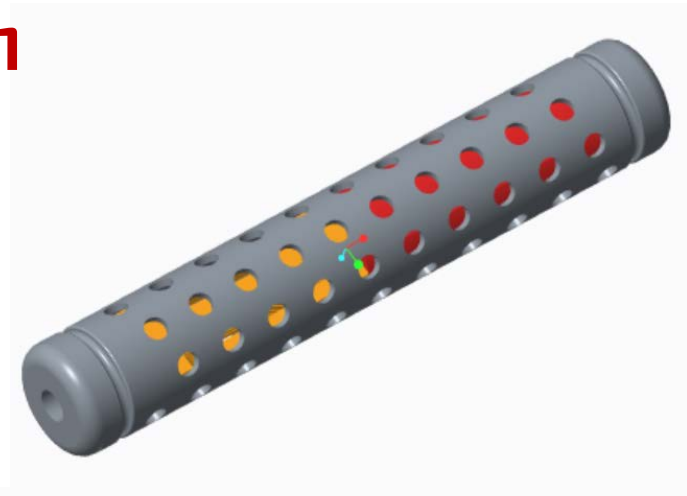
- SHINE Medical Technologies is developing a accelerator-driven subcritical assembly for the domestic production of molybdenum-99 and other isotopes for medical diagnosis and treatments
- The subcritical design has a number of materials challenges that are unique and require evaluation
- To support this effort, study of Zircaloy-4 and hydrided Zircaloy-4 under different experimental conditions, including neutron irradiation at temperatures of 60°C and 100°C, has been initiated



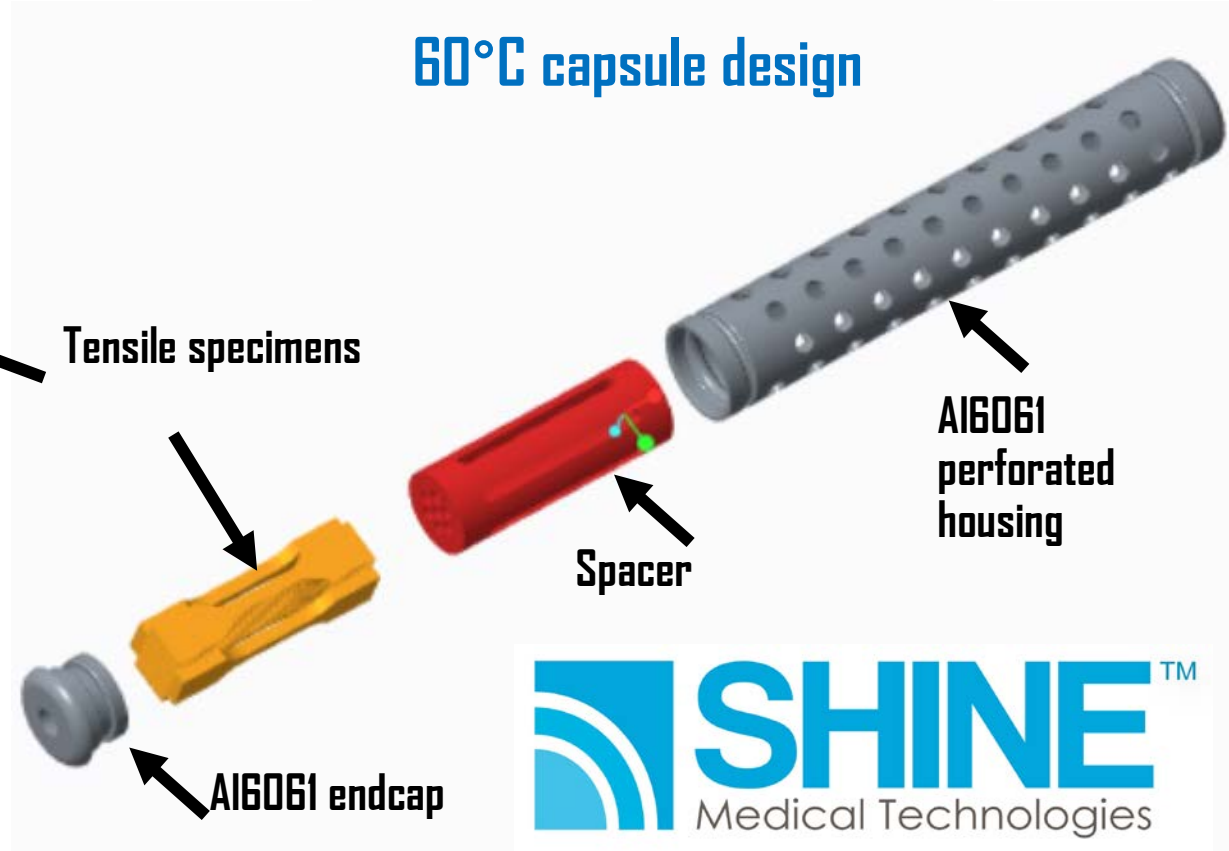
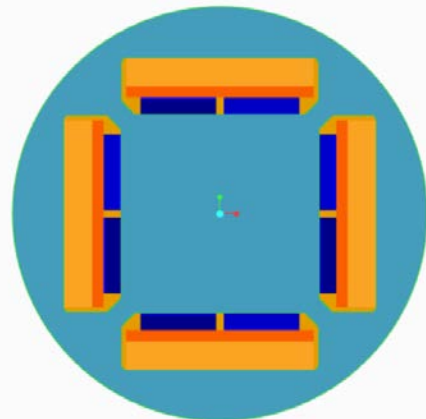
Irradiation Plan

Material Type	Design Temperature	Hydrogen Level	Desired Fluence (n/cm ²)	Irradiation Facility at HFIR	Irradiation Duration	Capsule Design Requirements
Zircaloy-4 Base Metal	60°C	<ul style="list-style-type: none"> 0 ppm 250 ppm 500 ppm 	1.00e20	HT-5	~1.3 days	Perforated housing, in contact with reactor coolant (~53°C)
			1.00e21	HT-5	~13 days	
Zircaloy-4 TIG Welded	100°C	<ul style="list-style-type: none"> 0 ppm 250 ppm 500 ppm 	1.00e20	HT-5	~1.3 days	Sealed capsule, filled with mixed gas
			1.00e21	HT-5	~13 days	

Capsule Mechanical Design

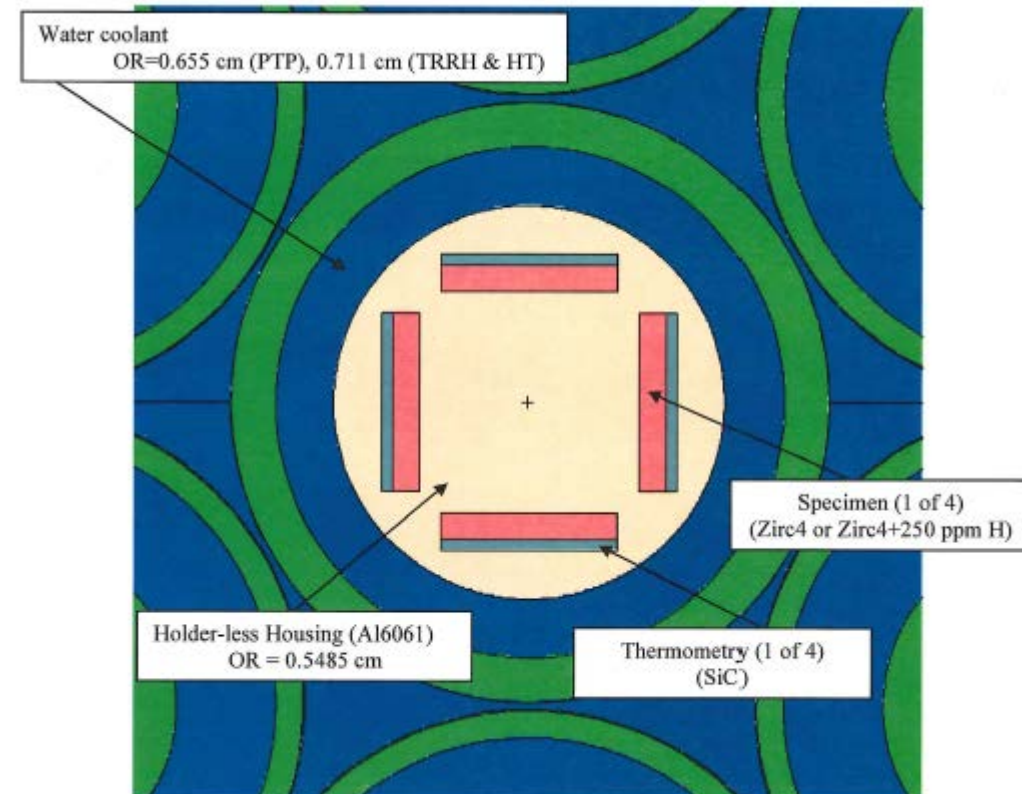


100°C capsule design



Heat Generation Rate Calculations

- The MCNP code is used to provide a flux spectrum and to estimate the prompt fission and fission product decay portions of the total heat
- The SCALE code system is used to estimate the decay heat portion of the total heat
- Nuclear heating in HFIR is dominated by photon absorption in most materials
 - Heat generation due to photon absorption is dependent on the atomic number of the absorbing element and the magnitude and spectrum of the capture gammas released as a result of neutron absorption
- Peak heat generation rates of Zircaloy-4, SiC, and Al6061 are calculated for the hydraulic tube irradiation facility in the HFIR flux trap



- In the absence of very strong absorbers, the flux in the flux trap is generally flat and radially symmetric

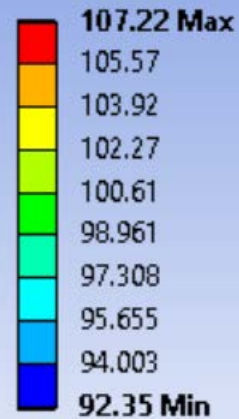
Case	BOC			EOC			Design		
	PTP	TRRH	HT	PTP	TRRH	HT	PTP	TRRH	HT
Zirc4*/ Al-6061	55.6	54.7	53.4	51.3	49.9	48.8	53.4	52.3	51.1
FeCrAl/Molybdenum	35.2	34.4	33.2	33.6	32.6	32.0	34.4	33.5	32.6
Inconel600/Molybdenum	37.7	36.5	35.9	35.7	34.8	34.0	36.7	35.7	34.9
Zirc4/Molybdenum	47.4	46.2	44.4	44.8	44.0	42.5	46.1	45.1	43.4

*Results for Zirc4 and Zirc4 + 250ppm H in the explicit model are statistically equivalent.

Thermal Analysis Results of 100°C Capsule

F: Steady-State Thermal

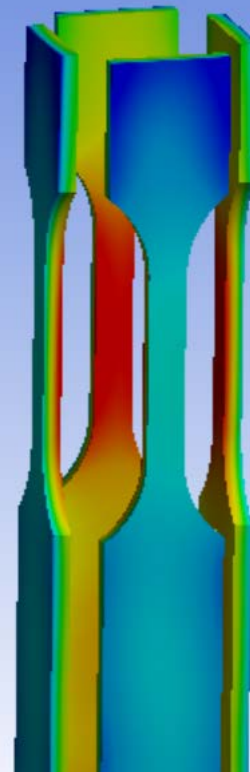
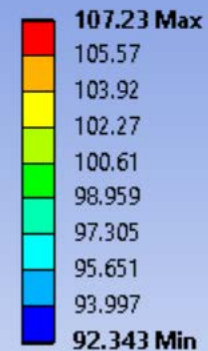
Tensile
Type: Temperature
Unit: °C
Time: 1
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- 68% helium, 32% argon mixed gas
- ΔT across the gauge section is $\sim 10^\circ\text{C}$
- Zircaloy-4 thermal conductivity $13.4\text{W/m}\cdot\text{C}$
- Zircaloy-4 density 6550 kg/m^3

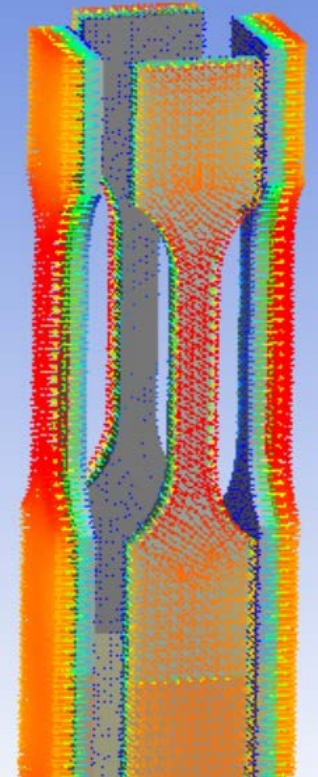
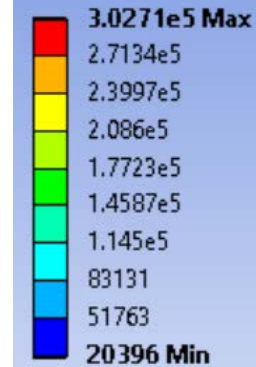
F: Steady-State Thermal

Temperature 3
Type: Temperature
Unit: °C
Time: 1



F: Steady-State Thermal

Total Heat Flux 2
Type: Total Heat Flux
Unit: W/m^2
Time: 1



Total heat flux

Irradiation Design and Analysis for Coqui Pharmaceutical

▪ Neutronics calculations

- Scoping calculations to determine the proper enrichment
- Heat generation rates
- Fission gas inventory
- Burnup, decay heat
- Depletion
- Activity, isotopic composition of the target
- Specific activity
- Mo-99 yield

• Safety basis calculations

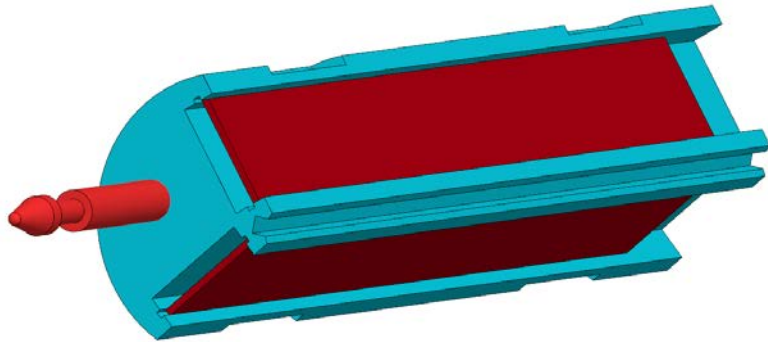
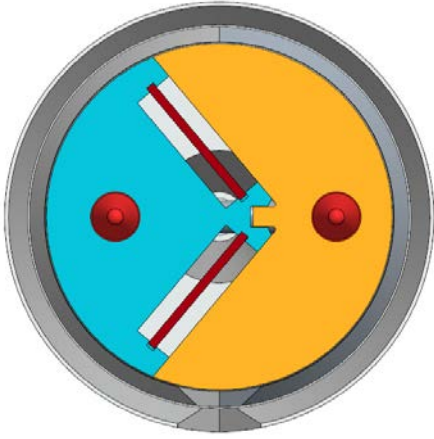
- Steady-state normal operation—at a full power of 87.6 MW
- Steady-state with flow blockage —at full power and 50% of the full flow
- Steady-state at 130% power (110.5 MW) and 100% flow
- Loss of offsite power (LOOP)
- Small break loss-of-coolant accident (SBLOCA)

• Preliminary design of the irradiation rig

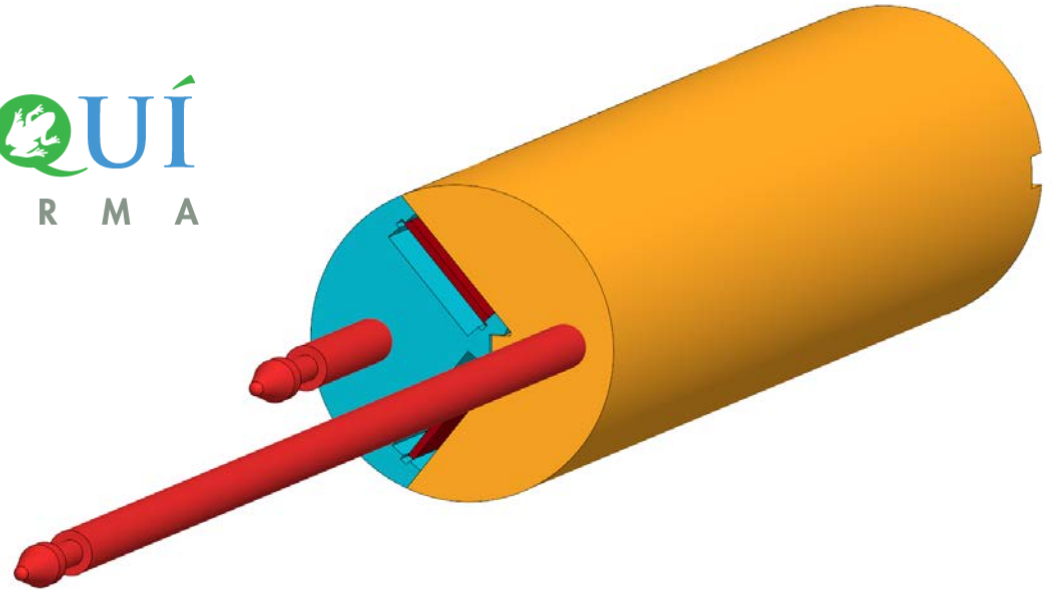
- Flow calculation, determining orifice for a proper pressure drop
- Flow test
- Thermal and structural analysis



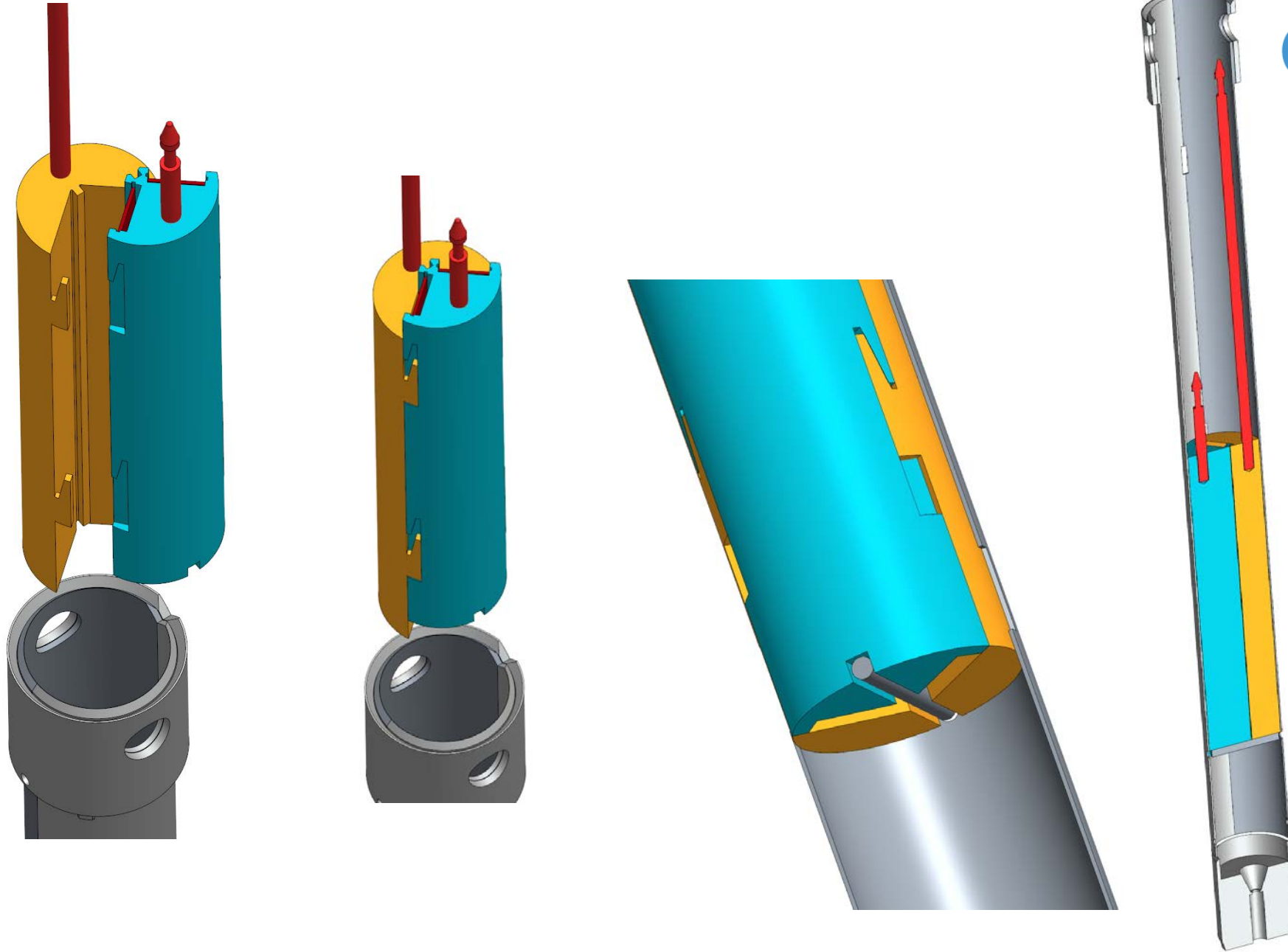
Preliminary Design of the LVXF Irradiation Rig



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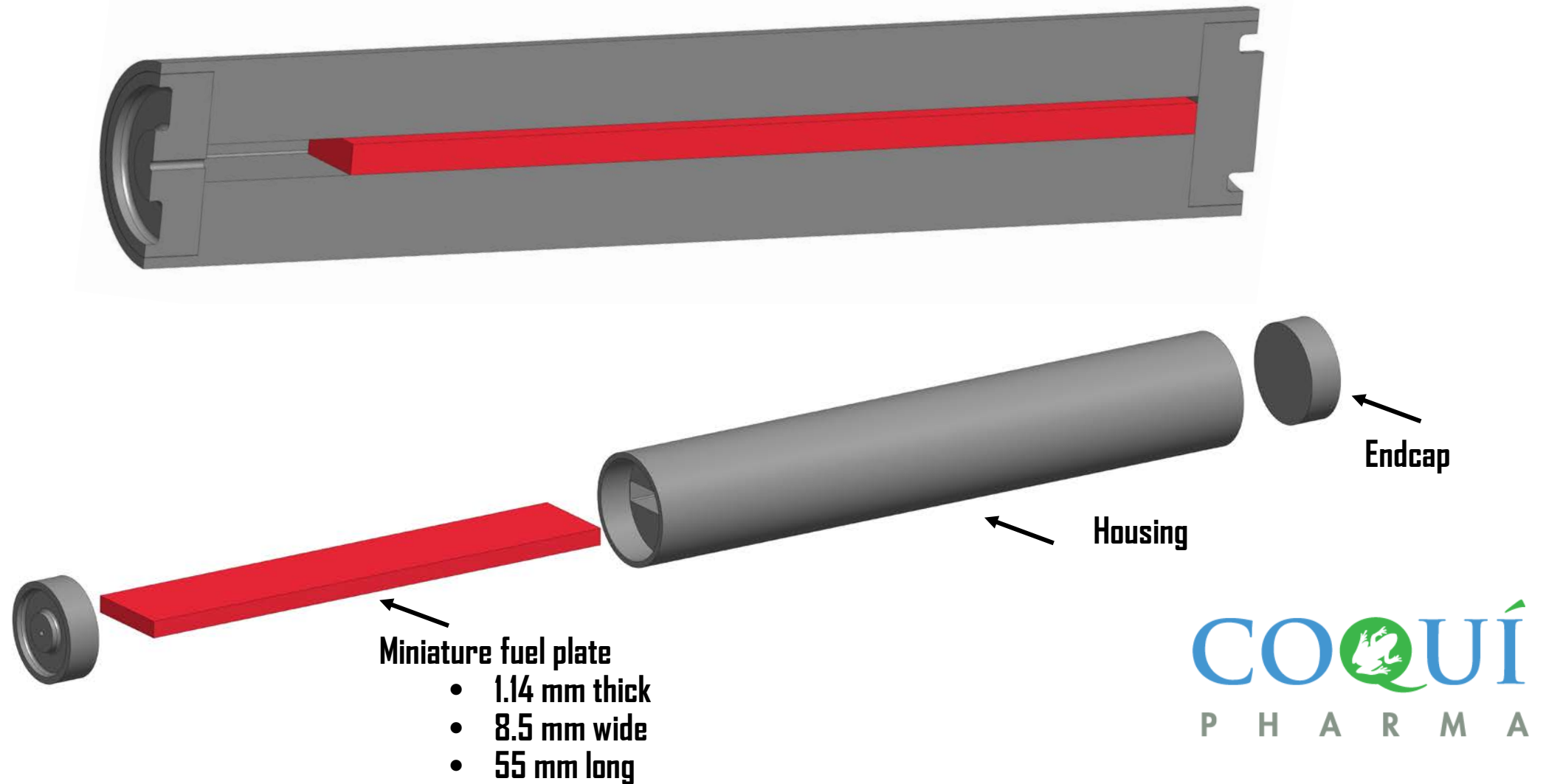


Preliminary Design of the LVXF Irradiation Rig



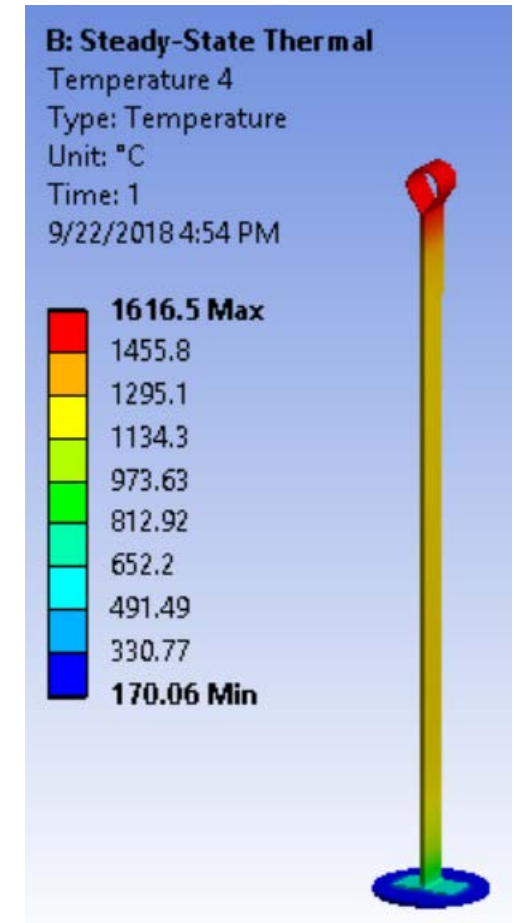
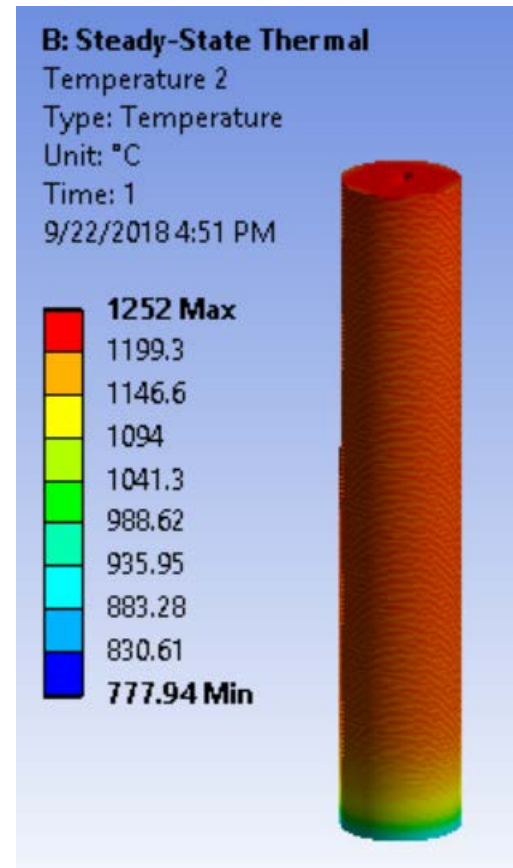
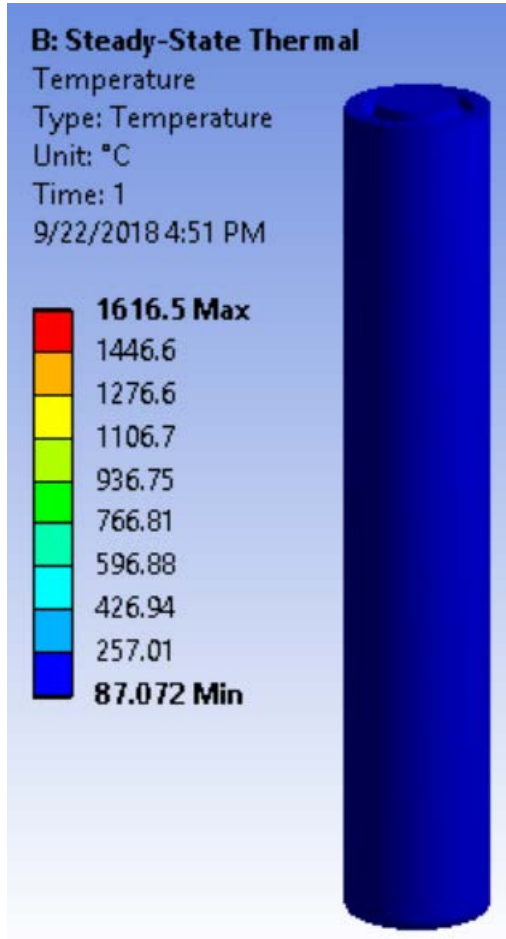
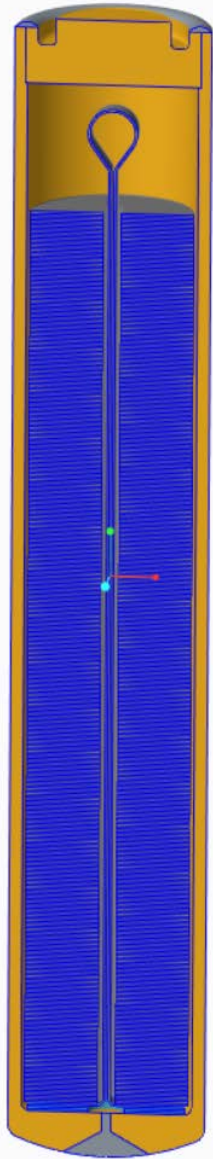
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Miniature Target Test Capsule for Hydraulic tube Irradiations



Irradiations for BWXT

- ~24 g moly disks
- ~1700 W total capsule heat load
- Max temp 1616°C under the Limiting Condition Scenario (LCS) at 130% power



Questions?

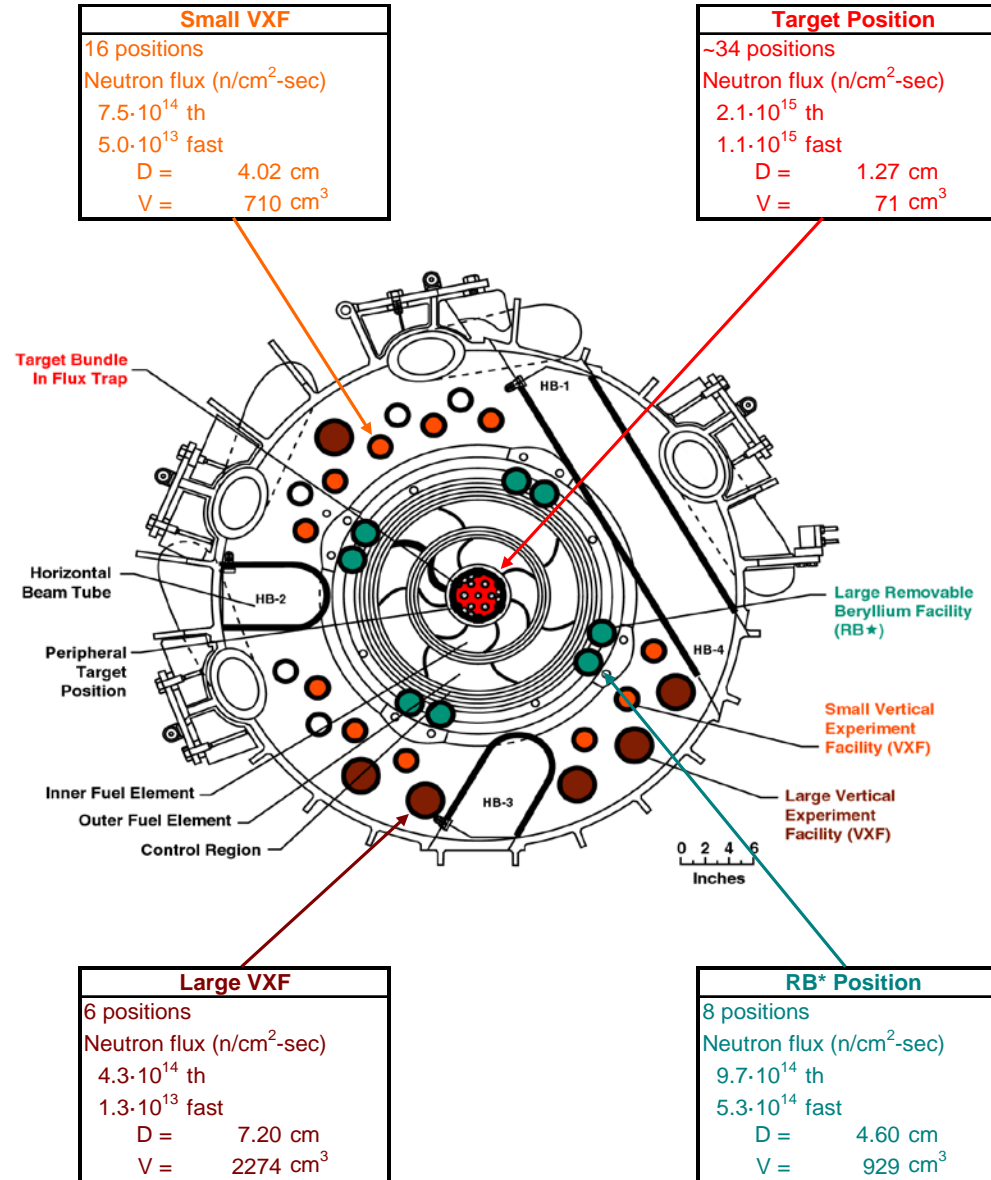
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HFIR Experiment Locations



NOTE: Facility diameters and volumes represent total available. Actual specimen volumes will be less.