



Tritium Transport within the TMIST-3 In-Reactor Experiment

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Overview:

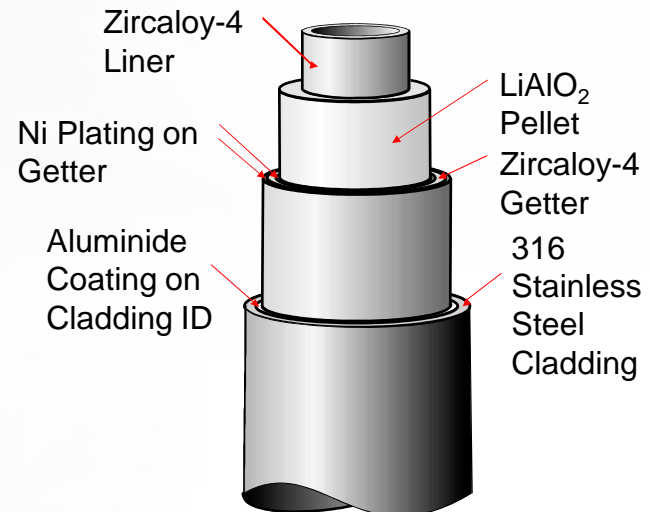
- ▶ Tritium Technology Program
- ▶ Motivation for TMIST-3
 - Modeling Needs
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Tritium Technology Program

- ▶ Supports the Tritium Readiness Subprogram for the National Nuclear Security Administration (NNSA)
 - Tritium production achieved by irradiating tritium-producing burnable absorber rods (TPBARs) in Watts Bar Nuclear Unit-1 (WBN-1)
 - TPBAR performance evaluated by
 - Post-Irradiation Examination (PIE) of select TPBARs
 - Monitoring tritium concentration in the reactor coolant system (RCS)
 - ◆ Observe regulatory requirements for environmental release



Watts Bar Nuclear Plant
Spring City, TN



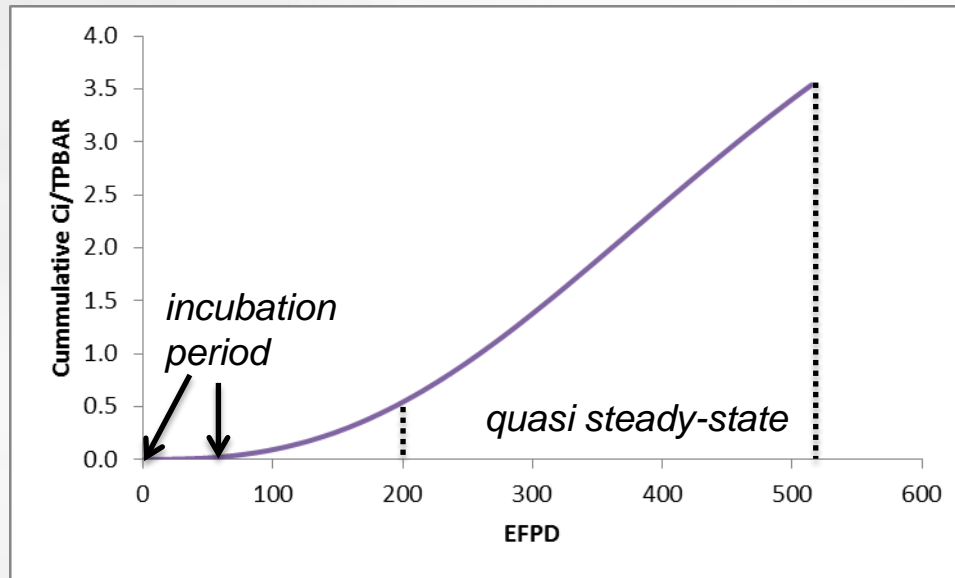
Not to scale

Motivation for TMIST-3

- ▶ Supports TTP Science and Technology (S&T) program
 - Evaluate underlying mechanisms driving TPBAR performance
 - Past TMIST/TMED experiments have evaluated:
 - ◆ Liner oxidation
 - ◆ Cladding permeation
 - ◆ Hydrogen absorption behavior
 - Provide data to improve TPBAR modeling capability
 - Estimate changes in performance due to changes in:
 - ◆ Plant operation
 - ◆ Component manufacture / vendors
 - ◆ TPBAR design
- ▶ Provides in-reactor data for the release and speciation of tritium from γ -lithium aluminate (γ -LiAlO₂) and cermet (γ -LiAlO₂ in Zr) pellets
 - Data collected as a function of time, burnup, and burnup rate
 - Establish an upper limit for pellet burnup
 - Assess release characteristics in context of TPBAR permeation

Modeling Needs

- ▶ Tritium speciation (i.e., T_2 or T_2O)
 - Interaction with adjacent components
- ▶ Kinetics of release
 - Does the apparent incubation period in RCS data result from:
 - Establishment of a driving equilibrium vapor pressure?
 - Tritium retention in pellet?

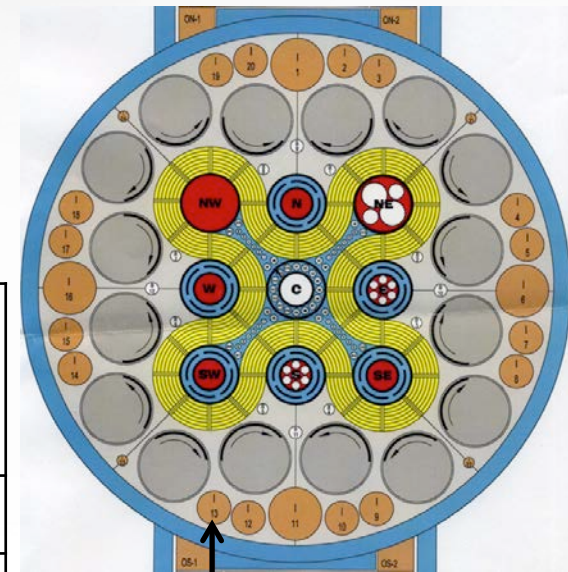


Cycle 9 RCS data

ATR Environment

- ▶ Tritium production achieved by ${}^6\text{Li}(n,\alpha)$ reaction
 - Test position selection based on thermal neutron flux
 - Test matrix requires two test trains
 - Mirrored neutronic and thermal hydraulic conditions desirable
 - Enables data on burnup rate to be acquired
- ▶ Medium I-Hole locations selected
 - I-9 for TMIST-3B
 - I-13 for TMIST-3A

ATR Position	Diameter (cm)	Thermal Flux (n/cm ² -s)	Fast Flux (E>1MeV) (n/cm ² -s)
Small B-Hole	2.22	2.5×10^{14}	8.1×10^{13}
Small I-Hole	3.81	8.4×10^{13}	3.2×10^{12}
Medium I-Hole	8.89	3.4×10^{13}	1.3×10^{12}
Large I-Hole	12.7	1.7×10^{13}	1.3×10^{12}
WBN-1	NA	5.4×10^{13}	2.1×10^{13}



ATR Core Map

Location for the TMIST-3A low-burnup test train (I-13)

TMIST-3 Description: Test Samples

▶ Test specimens

■ Standard TPBAR LiAlO_2 pellets

- 2 μm grain size
- 97-98% TD
- 1 mm wall thickness

■ Large grain LiAlO_2 pellets

- 10 μm grain size

■ Porous LiAlO_2 pellets

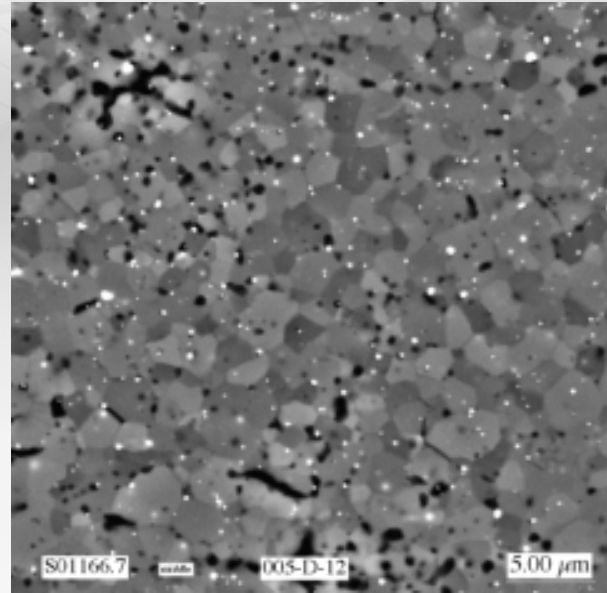
- Small pores (~90% TD)
- Large pores (~85% TD)

■ Thin-wall LiAlO_2 pellets

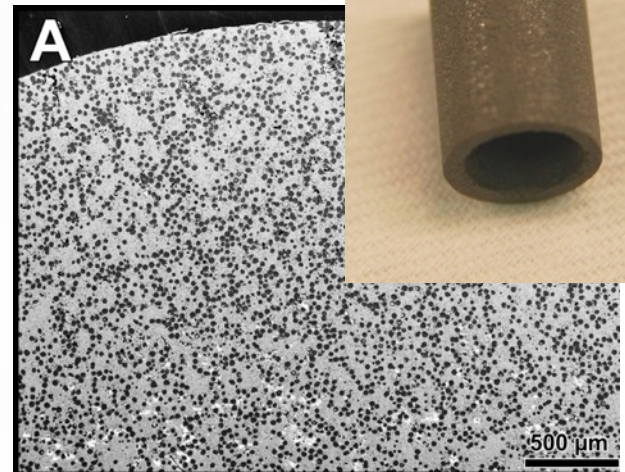
- 0.76 mm wall

■ Cermet pellets

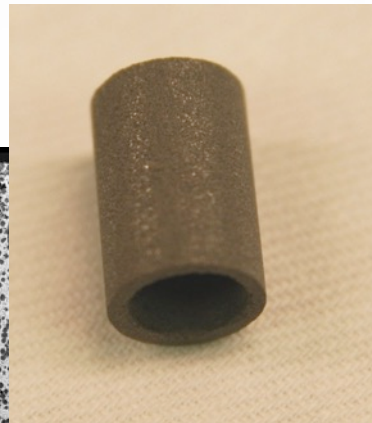
- LiAlO_2 particles in Zr matrix
- Four ceramic particle loadings from 10-40 v/o



Standard LiAlO_2 pellet microstructure

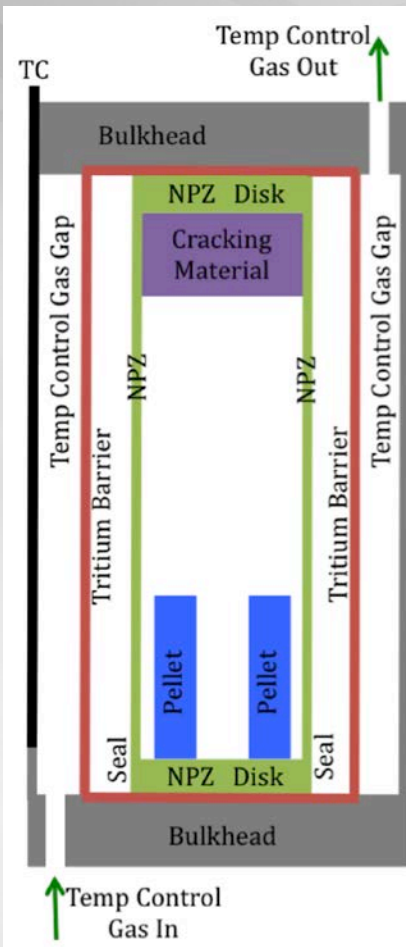


Cermet pellet with 40 v/o LiAlO_2



TMIST-3 Description: Capsule Design

Closed Capsule



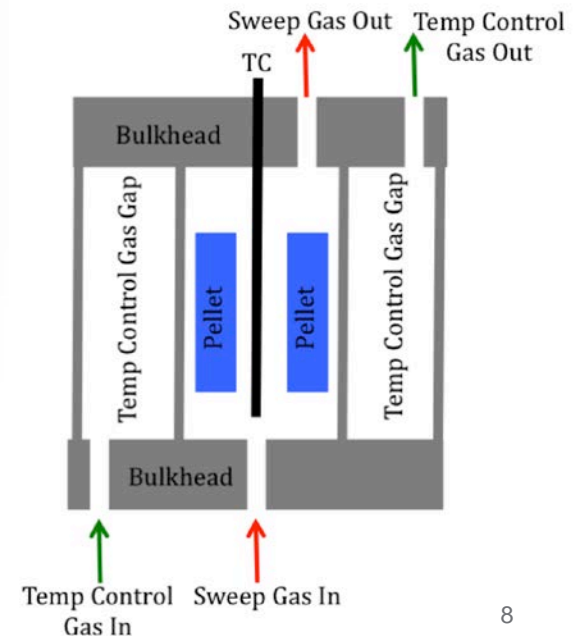
▶ Closed capsules

- Used for speciation measurements and pellet integrity/retention tests
- Tritium released from pellets as T_2 and T_2O is spatially segregated and gettered in-situ
- Speciation data inferred from post-irradiation examination tritium assays

▶ Flow-through capsules

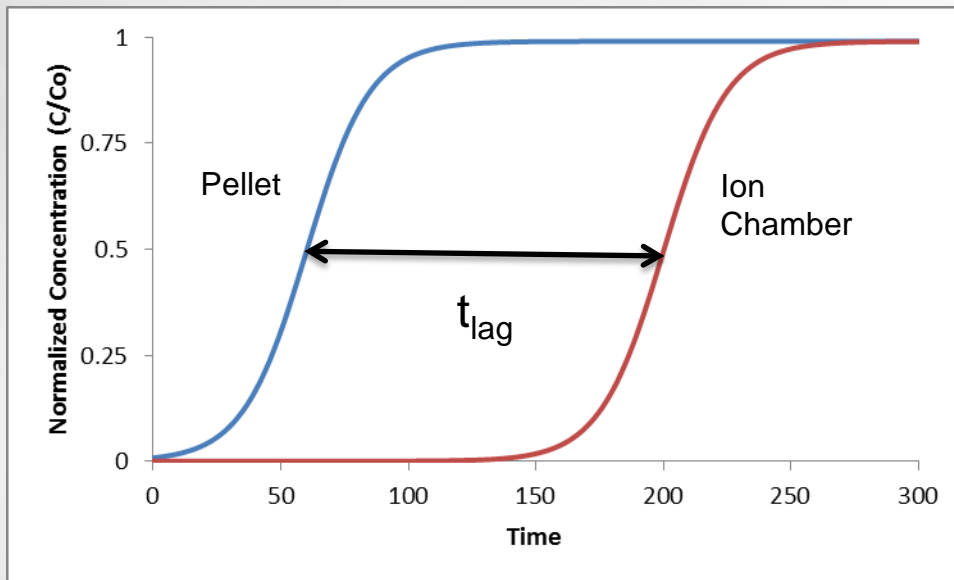
- Used for time, burnup, burnup rate, and temperature dependent tritium release measurements
- Tritium released from pellets is carried to ex-reactor measurement system for analysis
- Total tritium measurement only

Flow-Through Capsule



Tritium Transport

- ▶ Sweep gas from flow-through capsules transports tritium to measurement system outside of reactor (ex-reactor)
 - Ion chambers continuously monitor tritium concentration
 - Sweep gas proceeds from ion chambers to oxidizer columns bubblers prior to getter beds and exhaust
 - Results from ion chamber will be used to reveal tritium release kinetics



- ▶ A time lag, t_{lag} , is expected between pellet release and ion chamber measurement
 - Ideally, t_{lag} results only from the transit time between pellet and ion chamber
 - In reality, tube wall interactions can significantly impact ion chamber response

Ideal IC response relative to step change in pellet release

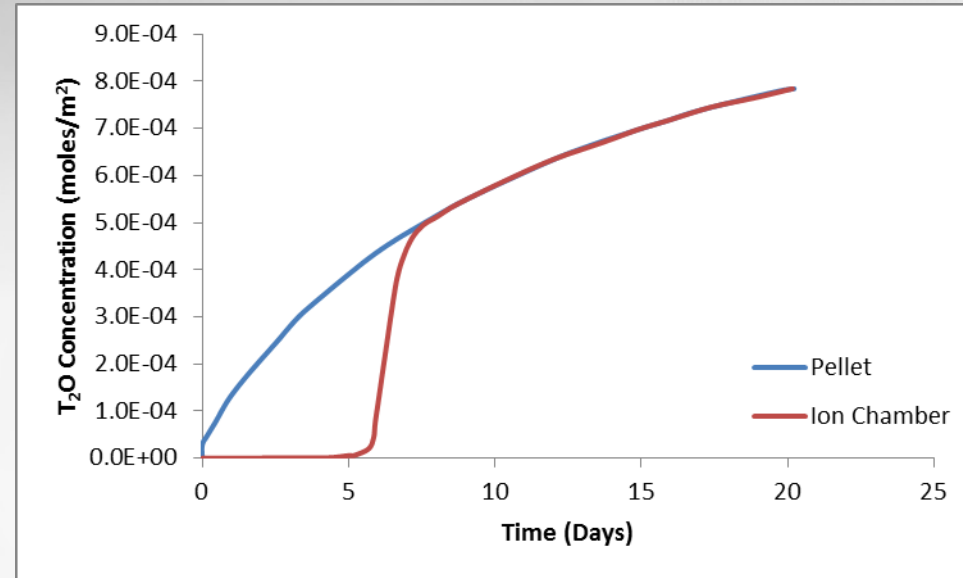


Initial Transport Model

- ▶ Tube wall interactions with T_2/T_2O laden gas were modeled to estimate ion chamber response time
 - Majority of pellet release is expected to be T_2O
- ▶ Multiple processes can act concurrently to impact tritium transport
 - Adsorption
 - Physisorption
 - ◆ Gas atoms or molecules are loosely bound on surface
 - ◆ Sensitive to partial pressure and to temperature
 - Chemisorption
 - ◆ Gas atoms or molecules are strongly bonded to surface
 - ◆ Sensitive to temperature and insensitive to pressure below threshold value
 - Isotopic exchange
 - Tritium and protium can “swap” or freely exchange with one another
 - Assumed to occur between physisorbed and chemisorbed inventories

Initial Transport Model

- ▶ Model assumptions
 - 100' of 1/8" Cu tubing at 150°F
 - T₂O introduced at inlet at 30 sccm
 - Mass transfer coefficient taken from T. Shiraishi et al. for water adsorption/desorption on surface
 - Output generated by WATER code developed at INL



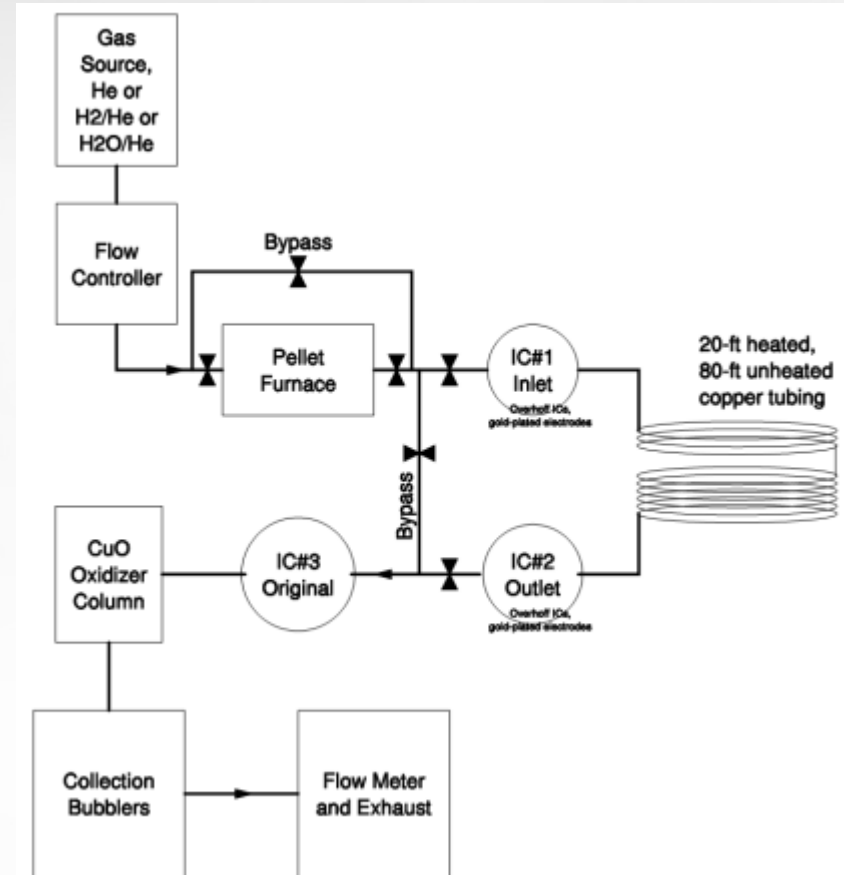
- ▶ Predictions indicate slow ion chamber response times
 - t_{lag} of 1 week or more can be expected, depending on release rate estimates
 - Slow response times expected to mask details of pellet release behavior
- ▶ Recommend filling physisorbed and chemisorbed inventories with a pre-test T₂O flush prior to test

T. Shiraishi, S. Odoi, M. Nishikawa, "Adsorption and Desorption Behavior of Tritiated Water on the Piping Materials," *Journal of Nuclear Science and Technology* v.34 [7] p.687-694 (1997)

M. Nishikawa, T. Takeishi, Y. Kawamura, Y. Takagi, Y. Matsumoto, "Tritium Balance in the Piping System of a Fusion Reactor," *Fusion Technology* v.21 p. 878-882 (1992)

Initial Transport Experiment

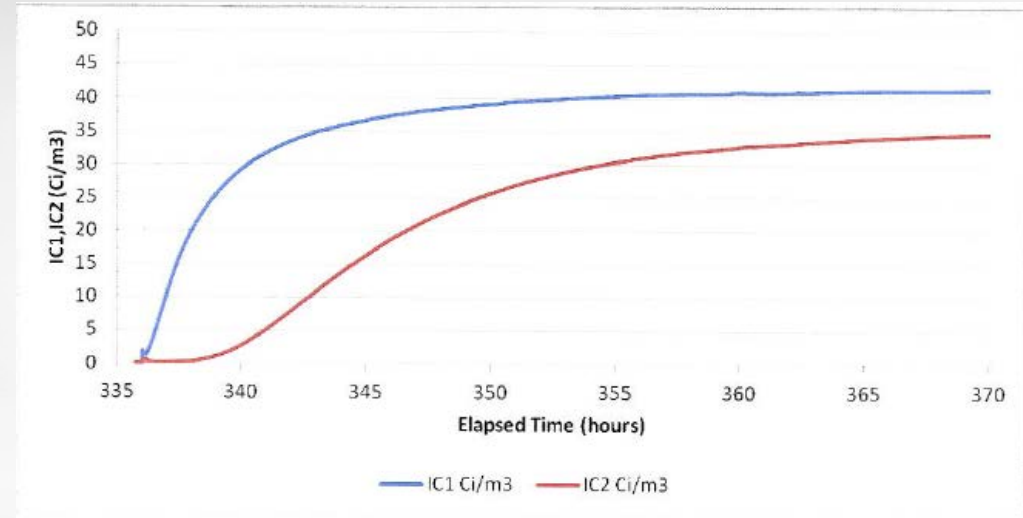
- ▶ Loop test setup in laboratory to generate data for model comparison
 - Irradiated γ -LiAlO₂ pellet used as source (T₂O)
 - 30 sccm UHP He
 - 100' 1/8" Cu tubing (20' at 150°F)
- ▶ Tests conducted to compare ion chamber performance and cleanup method
 - Ion chambers 1 & 2 use wire electrode design to minimize moisture retention
 - Ion chamber 3 is an older design with perforated stainless steel electrodes susceptible to moisture retention
 - Cleaning methods included
 - Dry He purge
 - Heating
 - Isotopic swamping (H₂, H₂O)



Initial Transport Experiment

▶ Ion chamber response (IC1 vs IC2) after source (T_2O) is released

- IC2 is shifted and modified relative to IC1
- t_{lag} is greater than time required for gas transit only
 - Consistent with tube wall interaction
 - Shape of IC2 response suggests slower mass transfer coefficient and smaller inventory
 - Final difference attributed to calibration uncertainty



▶ Response time is on the order of hours (< 1day) as opposed to a week or longer

- Suggests TMIST-3 pellet release events can be resolved reasonably well without a need for T_2O pre-flush

Initial Transport Experiment

▶ Overhoff Ion Chamber

- Designed to minimize T_2O retention
- Electrodes gold-plated
- Wire electrode design, virtual wall
- Triple electrode design
- Bakeable to at least 150C
- Used at PNNL and JET



Initial Transport Experiment

► Summary of cleaning operations

Ion Chamber Clean-up Method #	Method Description	Order of Effectiveness
1	Dry He Purge	Least Effective
2	Dry He purge with heat (80-130°C)	Slightly more effective
3	Isotopic Swamping with H ₂	More effective
4	Isotopic Swamping with H ₂ O	Most effective
5	Isotopic swamping with heat	No difference from 4

- Isotopic swamping with H₂O was easily the most effective clean-up method evaluated during this test
 - Rapid return to baseline observed on all three ion chambers

Summary and Future Work

- ▶ Tube wall interactions impact transport from source to ion chamber
 - Initial model indicated significant modifications to the lag time (several days to several weeks) and shape of the detector response
 - Test results indicate shorter lag times (< 24 hours) can be expected
 - Indicates TMIST-3 pellet release characteristics can be satisfactorily resolved
 - Eliminates need for pre-test flush with T₂O
- ▶ Ion chamber clean-up can be readily achieved by flushing with water vapor (H₂O)
 - Rapid return to baseline by removing absorbed tritium from ion chamber
 - Improved ion chamber designs exhibit less tritium build-up through absorption, but still benefit from periodic cleanup during long tests
- ▶ Additional work required to reconcile modeling efforts with test results
 - Evaluate assumptions and mass transfer coefficients in existing model
 - Implement new code developed to evaluate transport in TMIST-3

Acknowledgments

- ▶ Glen R. Longhurst
 - Idaho National Laboratory/ Southern Utah University
 - Model development and WATER code simulation
- ▶ David L. Baldwin
 - Pacific Northwest National Laboratory
 - Test loop design and testing at Radiological Processing Laboratory

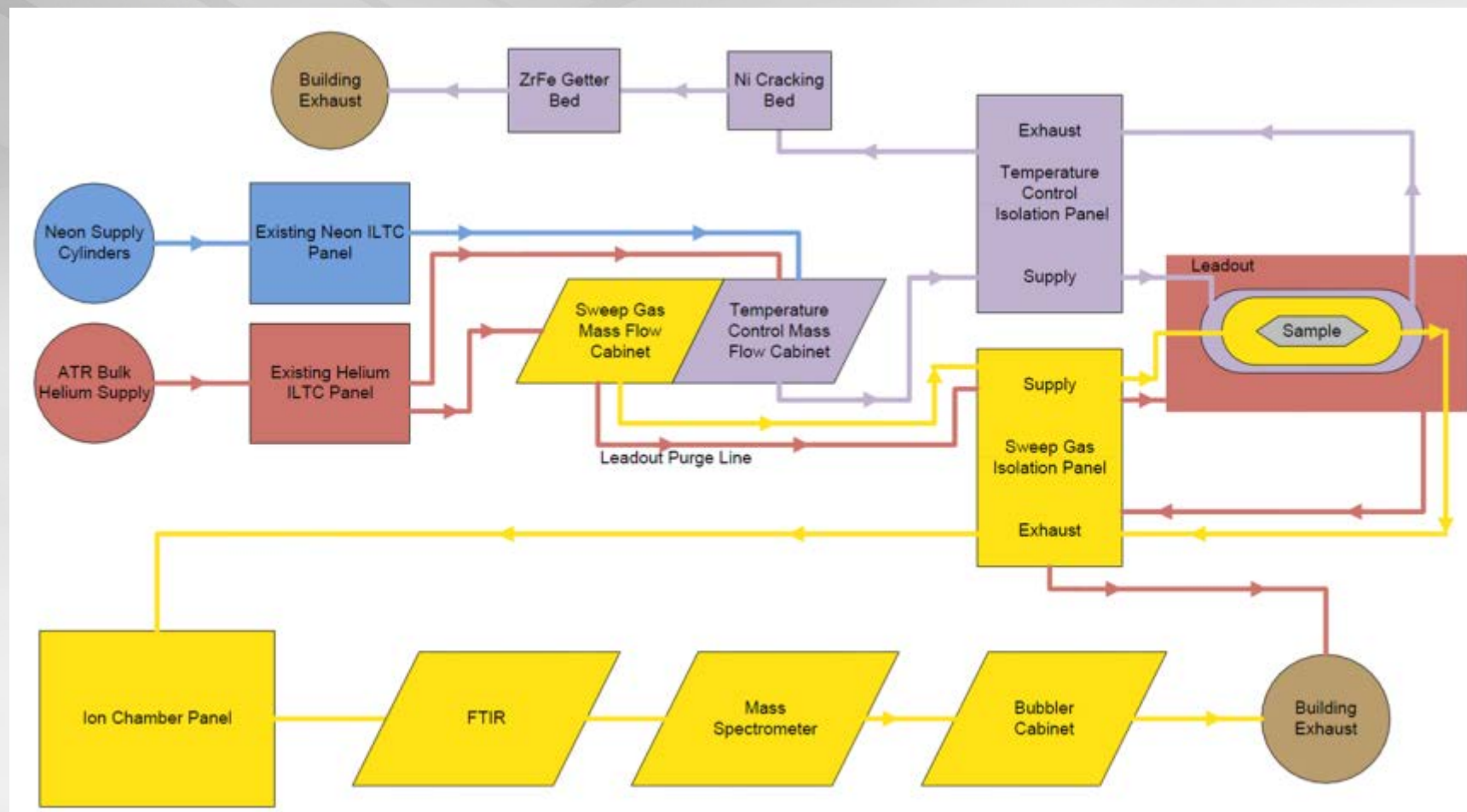
BACKUP SLIDES



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Layout of TMIST-3 gas flow system



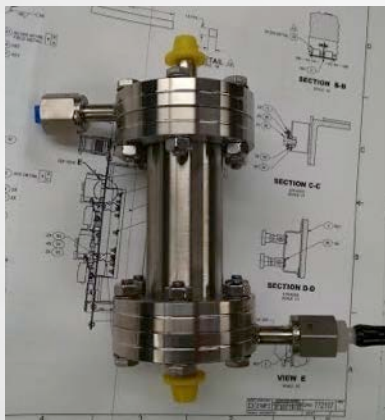
Components within TMIST-3 gas flow system



Ametek ProLine Quadrupole Mass Spec



Tyne 10cc Ion Chambers



Temperature Control Gas Mass Flow Controller Cabinet

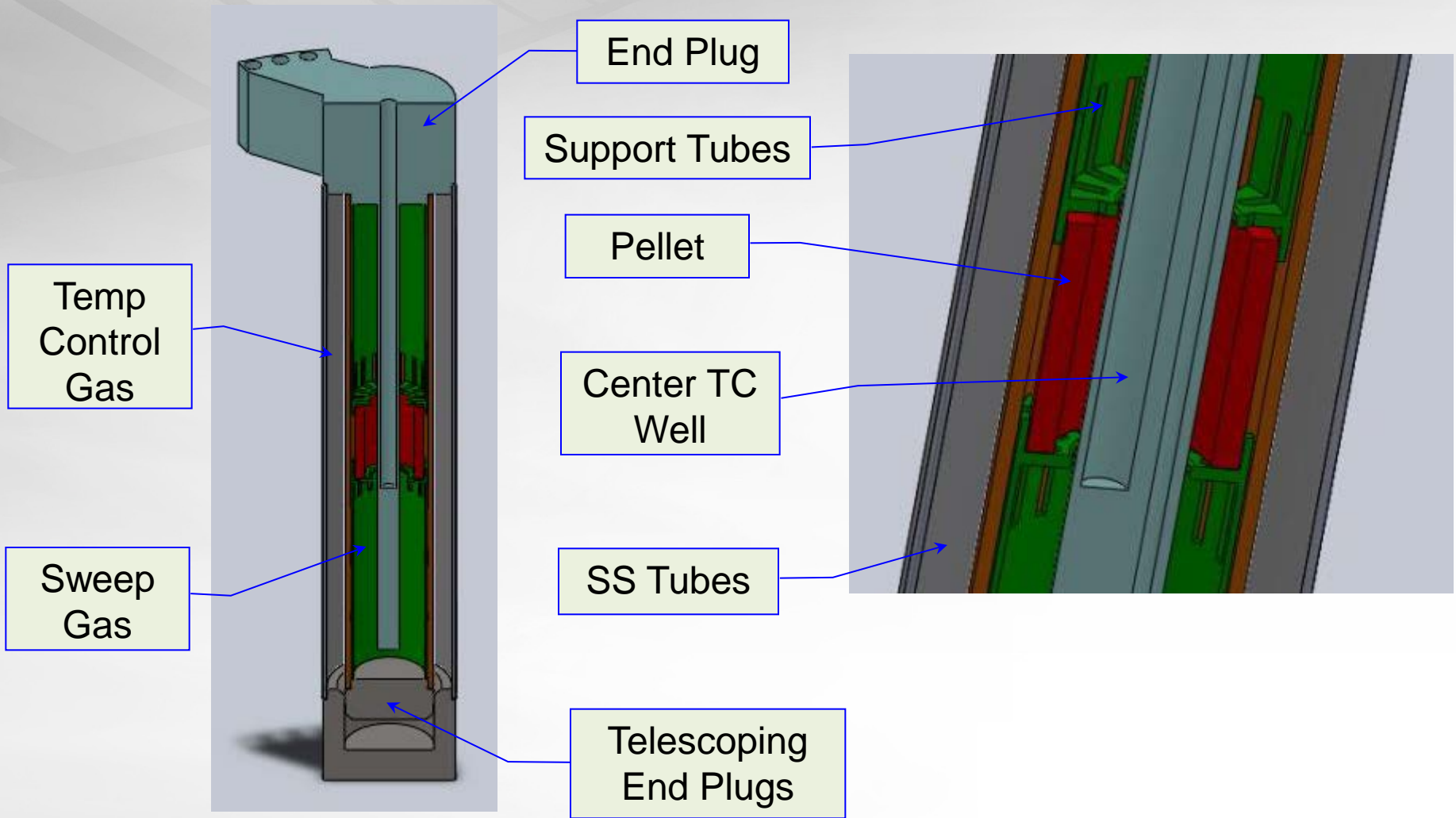
Getter Bed Cabinet

Flow Through Capsule - Overview

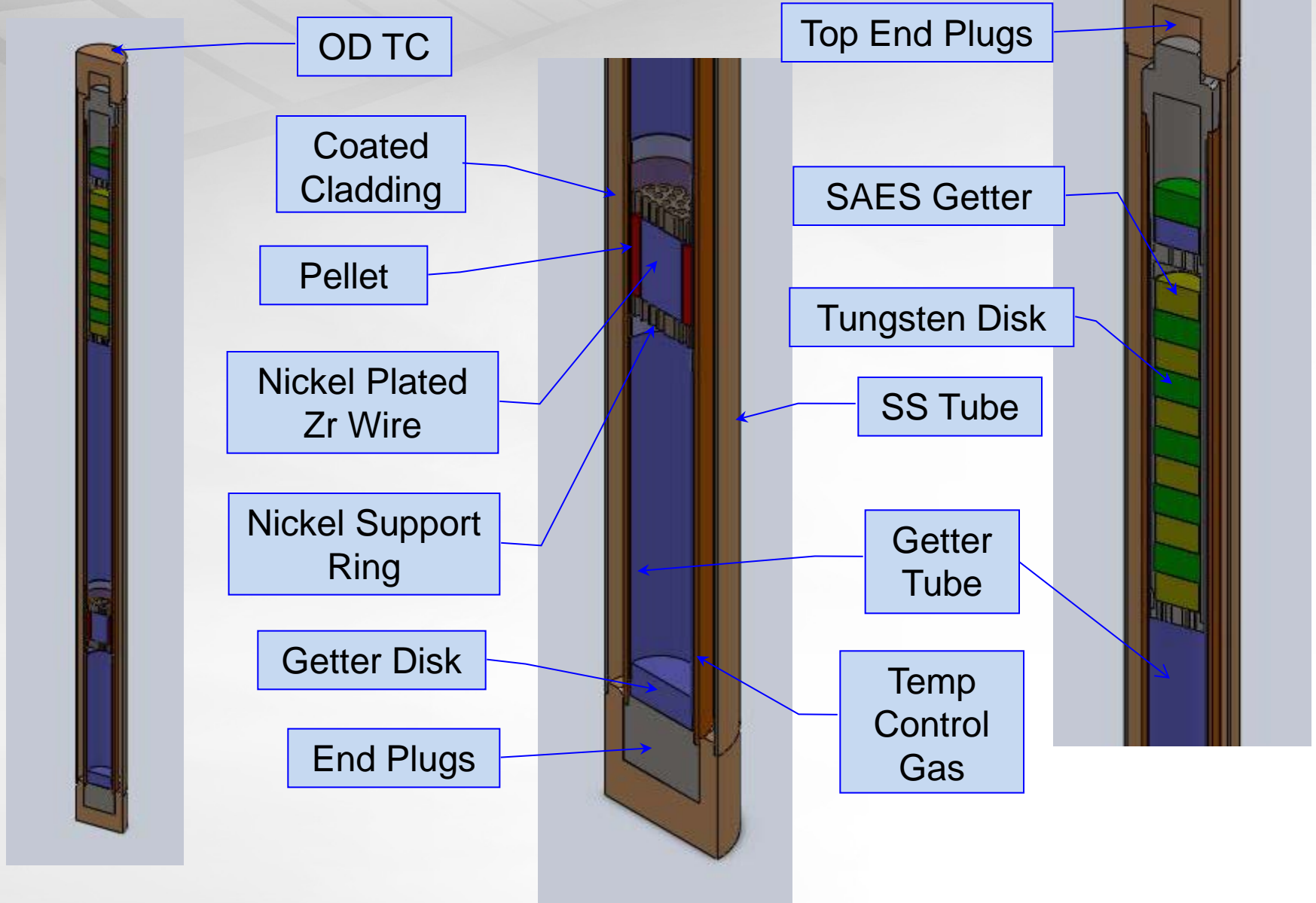


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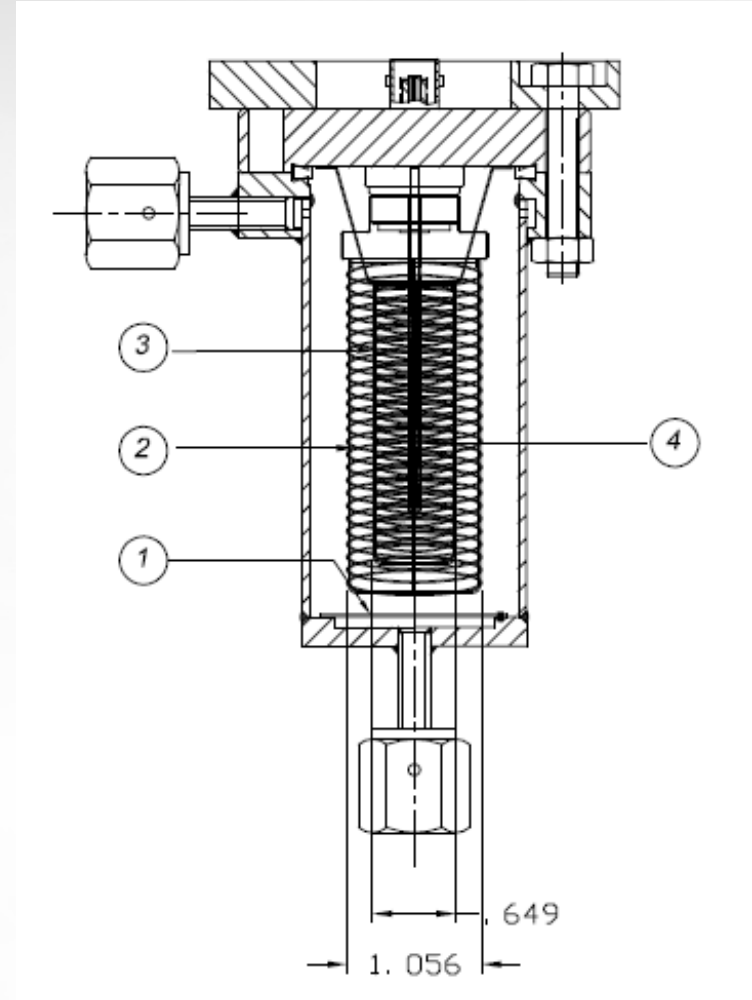


Closed Capsule - Overview



Initial Transport Experiment

► Tyne Ion Chamber



1. *DIFFUSER*, mesh, electropolished, guide the gas flow evenly;
2. *PRECIPITATOR*, wire, -100V to the chassis ground, creat electrical field;
3. *GROUND CASE*, wire, 0V to the chassis ground;
4. *COLLECTOR*, stainless rod, -100V+offset created by the ion.