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Design and Fabrication of In-Reactor Experiment to Measure Tritium Release and Speciation from $LiAlO_2$ and $LiAlO_2$/Zr Cermets

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Tritium Focus Group Meeting, Germantown, MD

Tritium Production Enterprise: Background

- Tritium is required for US nuclear weapons stockpile
- Tritium has a 12.3 year half-life and must be replenished
- 1988: DOE ceased production of tritium at SRS
- 1988-1992: The US considered the use of dedicated reactors for tritium production
  - Heavy water reactors (HWRs)
  - High temperature gas-cooled reactors (HTGRs)
  - Light water reactors (LWRs)
- 1995-1998: The US considered dual-use facilities
  - Commercial LWRs
  - Accelerators
- 1995: PNNL selected by DOE to be Design Authority for Commercial Light Water Reactor irradiation demonstration

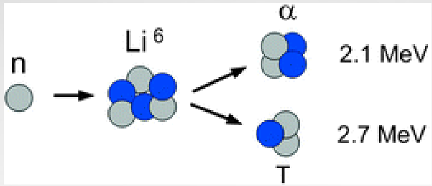
L Reactor at SRS

Tritium Production Enterprise: Background

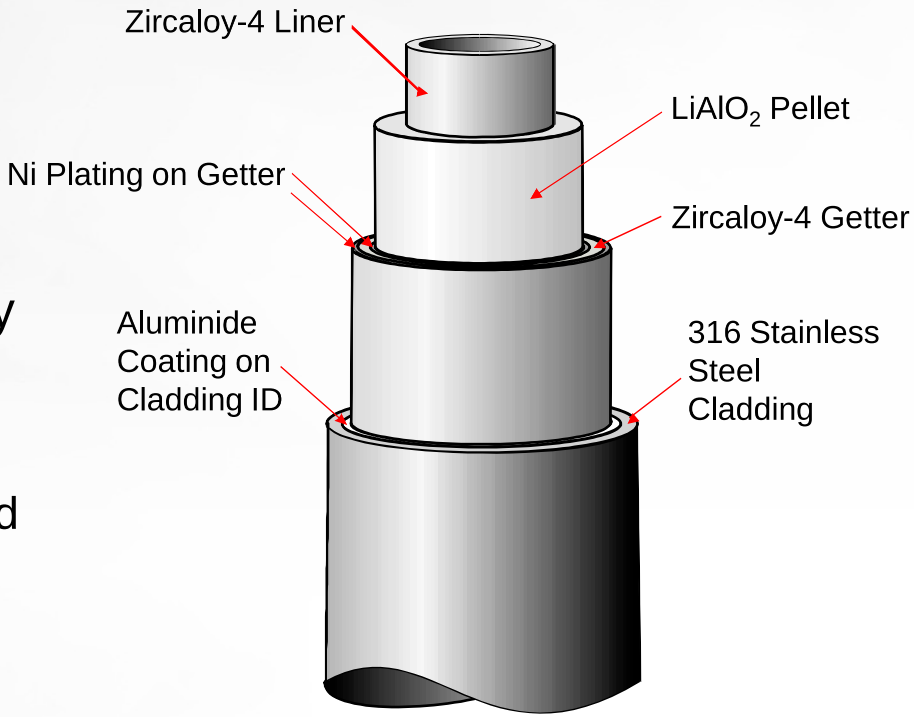
- 1995 – 1997: Lead Test Assembly (32 Tritium-Producing Burnable Absorber Rods, TPBARs) designed and built at PNNL for irradiation in TVA Watts Bar Nuclear Unit 1
- 1999: Post-irradiation examination of LTA
- 2000: The current Commercial Light Water Reactor tritium program was selected by DOE over accelerators for production
- 2001 – 2003: Design and manufacturing scale-up for production TPBARs
- 2003: First production core (240 TPBARs) irradiated at WBN1
- 2005 – 2008: TPBAR design modifications
- 2008: Modified TPBARs (Mark 9.2) first irradiated at WBN1

Watts Bar Nuclear Plant
Spring City, TN

Tritium Target Current Technology



- TPBARs replace burnable absorber rods normally used in Westinghouse PWRs (WABAs)
  - WABA reaction:
    - $^{10}B + {}^{1}n_{th} \rightarrow {}^{4}He + {}^{7}Li$
  - TPBAR reaction:
    - $^{6}Li + {}^{1}n_{th} \rightarrow {}^{3}H + {}^{4}He$
- Reactivity worth of TPBARs is slightly greater than WABAs
- Because TPBARs provide reactivity hold-down, they are considered a safety-related component by the NRC
  - All irradiation testing work governed by QA requirements in 10 CFR 50, Appendix B so results can be applied to TPBAR modeling and design



Not to scale

TPBAR Irradiation Performance

- In 2004, during the first production cycle at WBN1, it was determined that TPBAR tritium permeation was higher than predicted by performance models
  - Predicted ≈ 0.5 Ci/TPBAR/cycle
  - Actual ≈ 4 Ci/TPBAR/cycle
- Even 4 Ci/TPBAR/cycle represents only about 0.04% of the tritium produced
- TVA limited the number of TPBARs that could be irradiated because of current license limits on tritium release
- Subsequent irradiations have continued, but quantities are limited to <704 TPBARs/cycle
- An irradiation testing program was implemented in 2006 to provide a scientific basis for improving performance models and providing systematic, long-term TPBAR design evolution

Irradiation Testing Program Objectives

- Overall goal is risk reduction through fundamental understanding of TPBAR performance
  - Accurately explain and predict existing permeation performance
  - Provide confidence in performance predictions to support
    - Operating condition changes
    - Supplier changes
    - Manufacturing process changes
  - Provide basis for evolutionary design changes
- The testing program was tailored to address these objectives in support of the tritium production mission

Irradiation Testing Program

Cladding Permeation
TMIST-2
2006-2012

Getter Performance
TMED-4
2008-2010

Pellet Performance
TMIST-3
2009-2019

Advanced Pellet Mfg
TMED-3
2008-2011

Liner Oxidation
TMIST-1
TMED-1
2006-2010

Data from the Testing Program Has Improved TPBAR Performance Predictions

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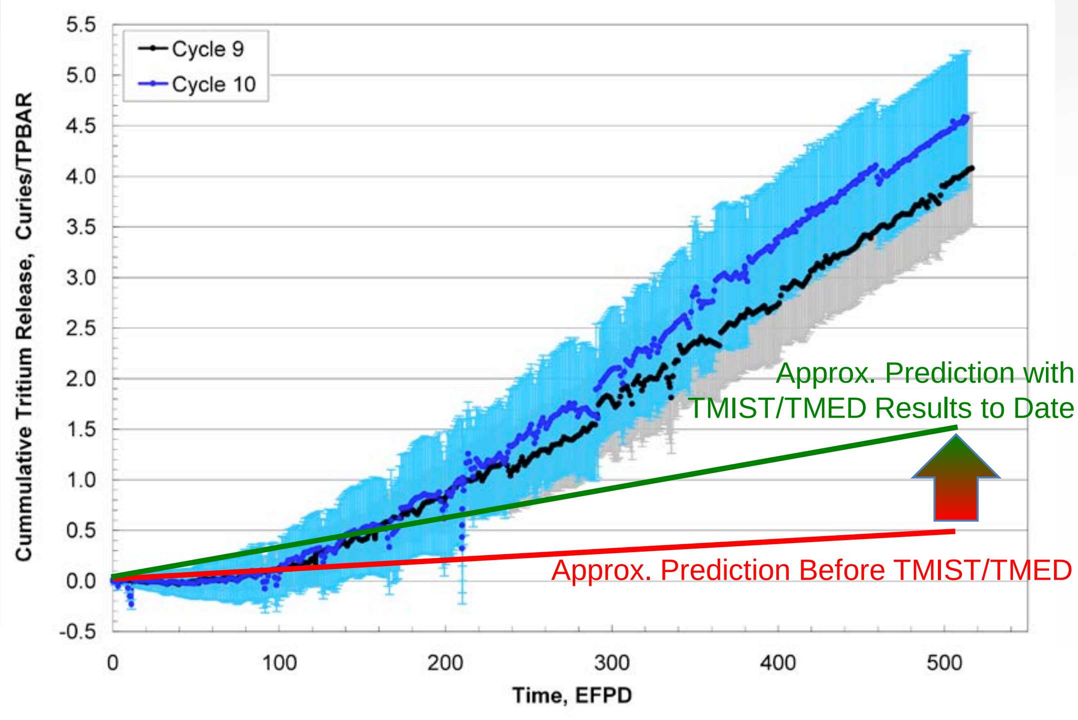
- TROD performance prediction code models updated with data from TMIST-1,TMED-1, TMIST-2, and TMED-4
- Discrepancy between predicted and observed permeation decreased by ~30%
- Time dependence still not correctly modeled
  - Will be improved by TMIST-3 data



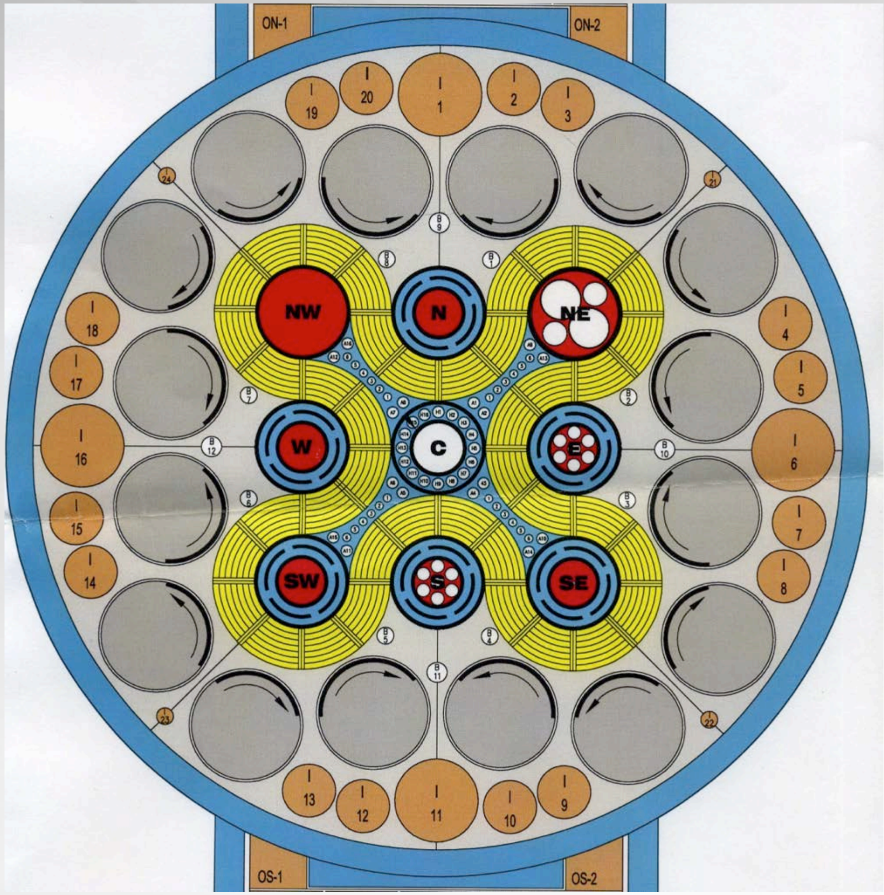
Approx. Prediction with TMIST/TMED Results to Date

Approx. Prediction Before TMIST/TMED

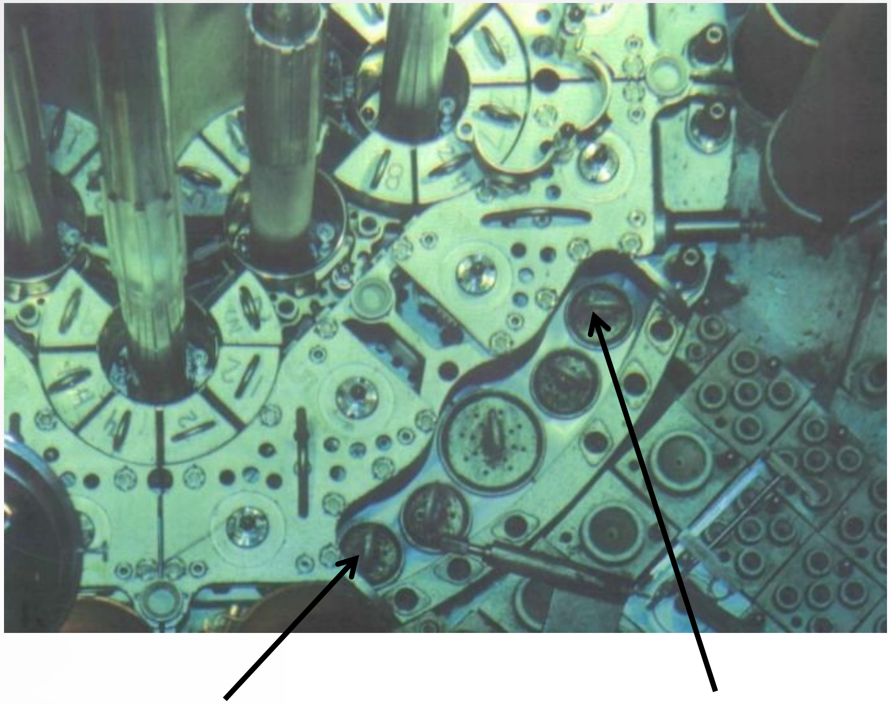
Pellet Performance Irradiation Experiment TMIST-3

- Data from TMIST-3 will
  - Explain time dependence of pellet tritium release and its relationship to TPBAR permeation
  - Evaluate the speciation of tritium release as a function of burnup, burnup rate, and time (T_2O versus T_2)
  - Define relationships between pellet burnup, burnup rate, and tritium release to help define an acceptable TPBAR operational envelope
  - Improve fundamental understanding of pellet microstructure and its effects on performance
  - Provide a better definition of the pellet burnup limit
  - Determine whether modifications to the pellets could improve TPBAR performance
    - Increased tritium retention
    - Increased TPBAR void volume

ATR Irradiation Positions TMIST-3



ATR Core Map

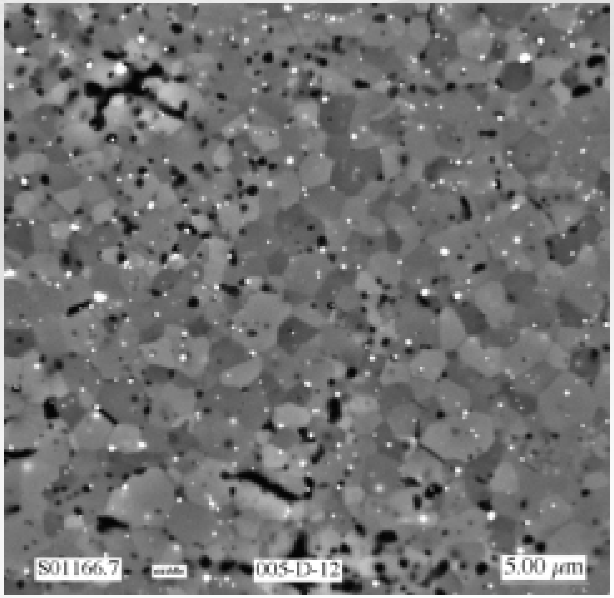


Location for the TMIST-3A low-burnup test train (I-13)

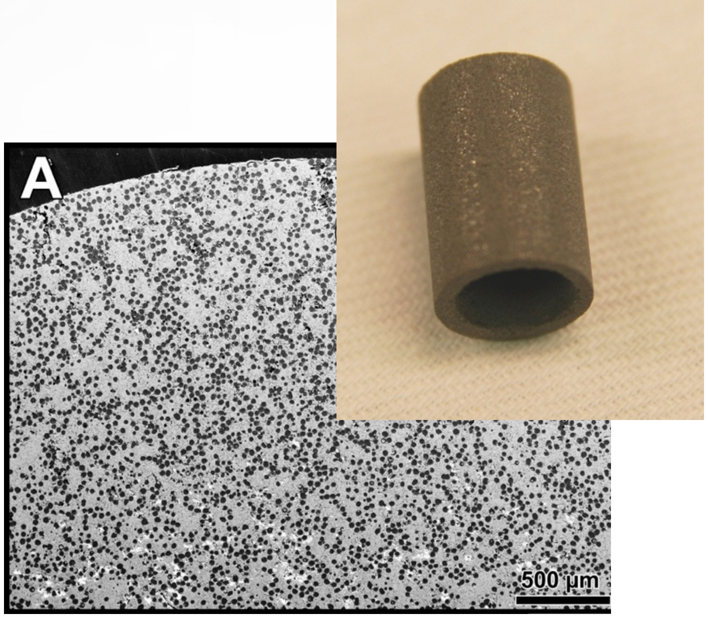
Location for the TMIST-3B high-burnup test train (I-9)

Test Specimens
TMIST-3

- 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



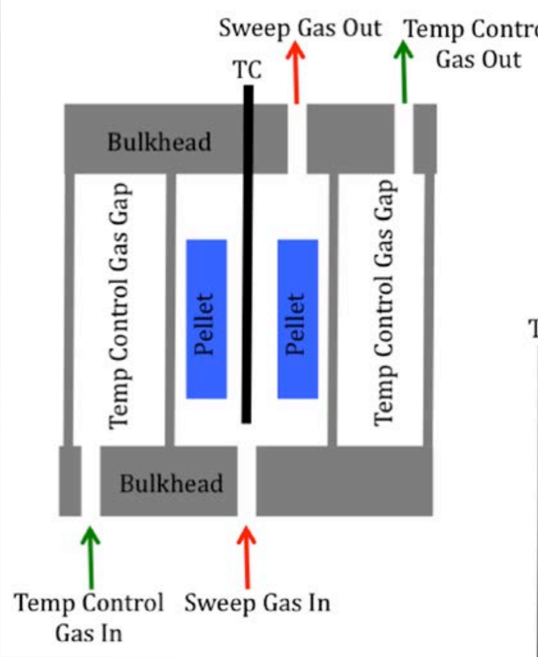
Standard $LiAlO_2$ pellet microstructure



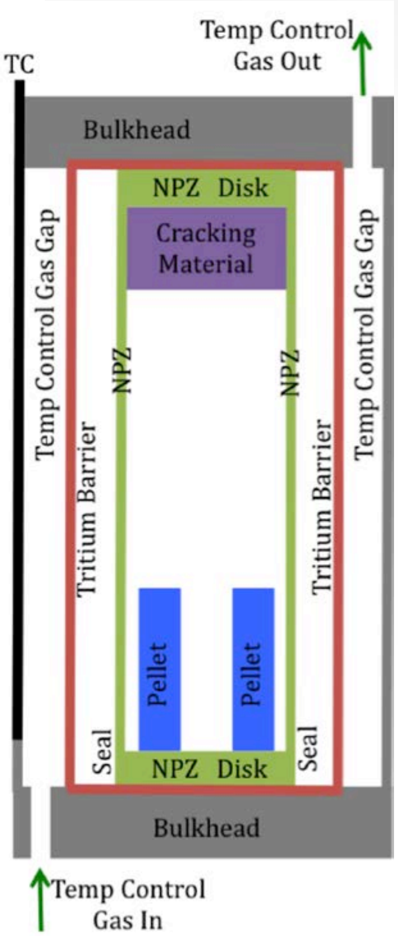
Cermet pellet with 40 v/o LiAlO2

Capsule Design
TMIST-3

- 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
- 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



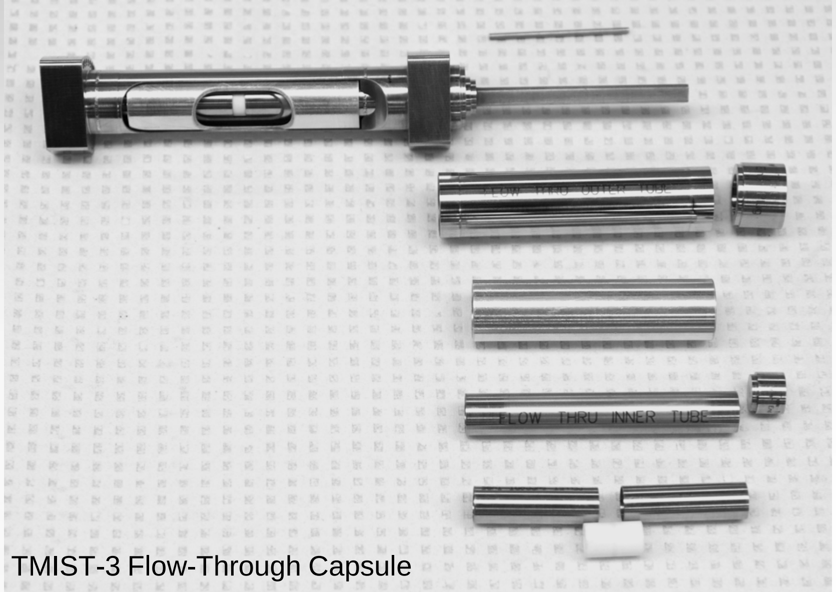
Flow-Through Capsule



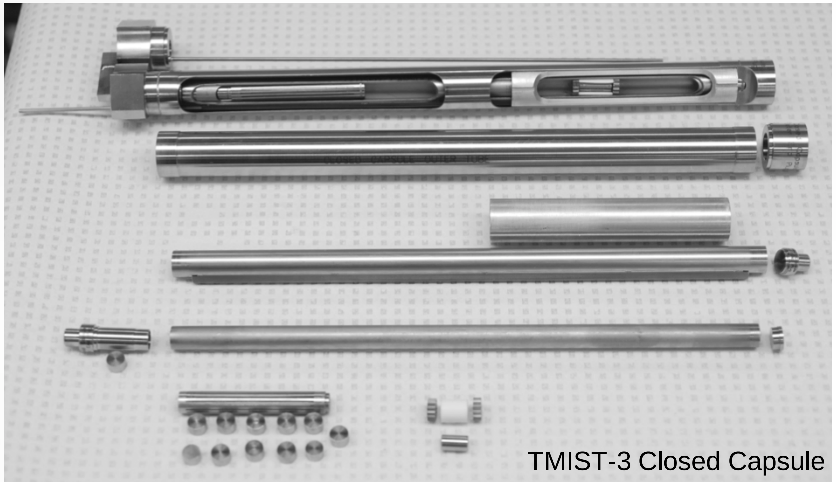
Closed Capsule

Test Train Design
TMIST-3

- Two test trains
  - TMIST-3A – Irradiate for ~1.5 yr
  - TMIST-3B – Irradiate for ~2.5 yr
- Two capsule types in each test train (41 total)
  - Flow-through – 15 total
  - Closed – 26 total
- All capsules have active He-Ne temperature control gas
  - One capsule designed to operate over a wide temperature range to evaluate temperature effects
- Flow-through capsules have He sweep gas to remove tritium for ex-reactor sampling
- 106 total leads for both test trains

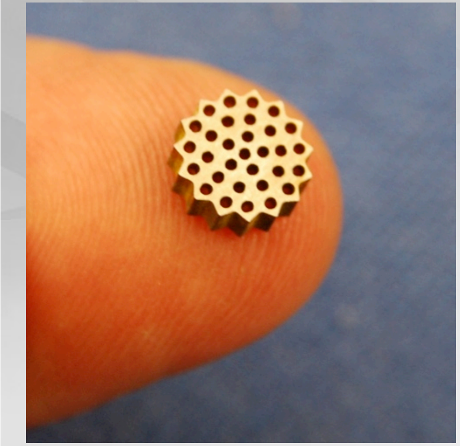


TMIST-3 Flow-Through Capsule

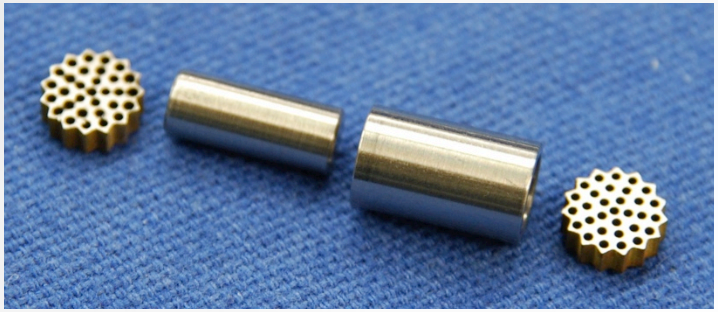


TMIST-3 Closed Capsule

Capsule Fabrication and Assembly TMIST-3

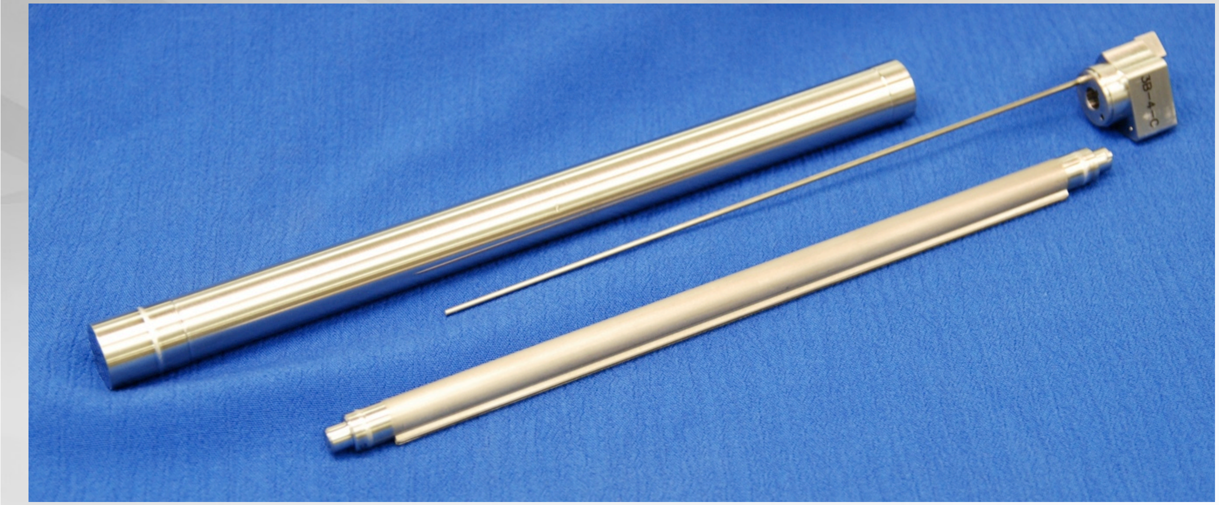


Some capsule components were challenging to machine, such as this Ni200 spacer designed to allow gas flow through and around the pellet



Double closed capsule assembly undergoing fit-up inspection after laser tack welding the end plugs, but before final end plug crossover closure welding via electron beam and TIG

Capsule Fabrication and Assembly TMIST-3



Closed capsule showing completed inner capsule with electron beam closure welds and t/c guide tube attached via laser weld, outer capsule with bottom closure weld, and end plug with temperature control gas inlet flow tube attached via laser weld

Completed flow-through capsules with and without Al6061 heat sinks

Path Forward
TMIST-3

- Test train assembly proceeding at INL
- TMIST-3A test train scheduled for insertion in ATR in spring 2014
  - Before ATR core internals changeout
- TMIST-3B test train scheduled for insertion in ATR in fall 2016
  - After ATR core internals changeout
- Post-irradiation examination to be completed at PNNL
  - Optical, scanning, transmission electron microscopy with EDS/WDS
  - XRD for phase identification
  - He pycnometry, Hg porosimetry
  - $^{6}Li/^{7}Li$ isotopic analysis and flux wire dosimetry to confirm burnup
  - Closed capsule puncture, pressure measurement, gas analysis
  - Retained ^{3}H, ^{3}He, ^{4}He, O assays
  - ^{3}H assays in closed capsule getter and cracker components
- Comparison of data to production TPBARs irradiated in WBN1
- Improvement of pellet and TPBAR performance models