LiAlO$_2$ Tritium Release Data from the First Two Cycles of the TMIST-3A Irradiation Experiment

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PACIFIC NORTHWEST NATIONAL LABORATORY
RICHLAND, WA
MAY 9, 2017

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Overview

► TMIST-3 Pellet Performance Irradiation Experiment
  ■ Motivation for TMIST experiments
    ◆ Enhance predictive capability to accurately explain existing permeation performance
    ◆ Mitigate programmatic risk due to changes in vendors, fabrication methods, and reactor operation
    ◆ Provide basis for evolutionary design changes
  ■ Objectives of TMIST-3
    ◆ Determine speciation of tritium released from pellet (T$_2$ vs. T$_2$O)
    ◆ Evaluate tritium release kinetics

► TMIST-3 Experiment Description
  ■ Pellet designs
  ■ Test capsule designs
  ■ Test train designs
  ■ Irradiation environment
  ■ Ex-reactor measurement system

► Preliminary Results from Cycle 160A and 160B
  ■ Cycle descriptions
  ■ Thermal model predictions
  ■ In-situ measurements

► Summary of Preliminary Conclusions
TMIST-3: Experiment Description

**Pellet Designs**
- **Standard TPBAR LiAlO₂ pellets**
  - 2 µm grain size
  - 97-98% TD
  - 1 mm wall thickness
- **Large grain LiAlO₂ pellets**
  - 10 µm grain size
- **Porous LiAlO₂ pellets**
  - Small pores (~90% TD)
  - Large pores (~85% TD)
- **Thin-wall LiAlO₂ pellets**
  - 0.76 mm wall
- **Cermet pellets**
  - LiAlO₂ particles in Zr matrix
  - Four ceramic particle loadings from 10-40 v/o

**Capsule Designs – Closed and Flow-through**
- **Closed capsules**
  - Used for speciation measurements and pellet integrity/retention tests
  - Tritium released from pellets as T₂ and T₂O is spatially segregated and gettered in-situ
  - Speciation data inferred from post-irradiation examination tritium assays
TMIST-3: Experiment Description

- Capsule Designs – Closed and Flow-through
  - Flow-through capsule
    - 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](image)

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TMIST-3: Experiment Description

- Lead Out
  - Top of Core
    - 57 Total Lines
    - 22 Temperature Control Gas Lines (11 Mass Flow Controls)
    - 18 Sweep Gas Lines (9 Mass Flow Controls)
    - 16 Thermocouples
      - Top & Bottom Banking
      - Center Banking
  - Back & Front
    - 654 Pellet Samples

![Diagram](image)

Note: Crossed lines are not junctions.
TMIST-3: Experiment Description

ATR Core Map

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

ATR Position | Diameter (in) | Thermal Flux (n/cm²·s) | Fast Flux (E>1MeV) (n/cm²·s) |
---|---|---|---|
Medium 1 Hole | 3.5 | 3.4x10¹³ | 1.3x10¹² |
WBN-1 | NA | 5.4x10¹⁴ | 2.1x10¹⁵ |
TMIST-3: Experiment Description

Cycle 160A
TMIST-3: Preliminary Results

Cycle 160A Reactor Operation

- September 16 to November 8, 2016
  - 53 calendar days at ~ 20 MW
  - ~48 EFPD at 23 MW

![Graph showing reactor operation](image)

TMIST-3: Preliminary Results

Cycle 160A Reactor Operation

- Nominal capsule thermal performance (standard 140 GVR closed capsule)

![Graph showing capsule performance](image)
Temperature control gas mixtures adjusted to increase neon concentration to offset lower than anticipated lobe power.

1A and 3A setpoints revised because original setpoints were not achievable at 100% Ne.
- 1A lowered from 617°F to 607°F
- 3A lowered from 695°F to 598°F

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<th>TC ID</th>
<th>Setpoint (°F)</th>
<th>Average (°F)</th>
<th>STDEV (°F)</th>
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Flow Rate Data

- Initial: He (sccm) 30, Ne (sccm) 0
- Actual: He (sccm) 30, Ne (sccm) 30

Acceptability of the test train thermal performance:

- Majority of capsules achieved thermocouple readings within specified range.
  - Lower lobe power required temperature control gas adjustment to be Ne-rich for most capsule.
  - Setpoints were revised for 2 capsules that were unable to reach the specified temperature range despite being set to 100% Ne flow.

- Test position (I-13) on periphery of core.
  - Challenging position to model flux profile.
    - Comparisons between flux wire measurements and initial model predictions revealed significant disparity.
    - Increased uncertainty in reactor physics models which impacts thermal performance of the mechanical design.

Despite revising two capsule setpoints, the overall thermal performance of the test train design provided satisfactory temperature control.
Tritium detected in gas phase as reactor brought to $N_f = 1$
- No significant delay/incubation period prior to release
Release dependent on target burnup for both pore sizes
- Small pore 70 GVR temperature decreased 60°F on 10/3
- Tritium release from porous pellet microstructures enhanced relative to those without pores (next slide)

Larger grains may not offer significant advantage over standard size
- Cermet release is lower initially, but increased over Cycle 160A
- Also at a lower temperature relative to other pellets (564°F)
Liquid scintillation counting (LSC) of bubbler vials provides a quantitative measure of tritium in the gas stream.
- General trends are similar ion chamber data
- Data permits scaling of IC data and integration to develop cumulative release curves

Average and cumulative release rate data tabulated on next slide.
TMIST-3: Preliminary Results

<table>
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<tr>
<th>Pellet Capsule</th>
<th>GVR</th>
<th>Average TC Reading (°F)</th>
<th>Cycle-Average Release Rate (Ci/day)</th>
<th>EOC Cumulative Release (Ci)</th>
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- Cycle-average rate provided for a sense of magnitude only (i.e. mCi)
- Rate changes with time over cycle
- Overall tritium release rates and totals are relatively low
  - Gas stream concentration calculated to be < 1ppm
  - Mass spectrometer minimum detection limit is 1ppm
    - No signals other than helium detected

Cycle 160A Summary

- Overall test train performance is successful
  - Reactor physics were satisfactorily captured and used to inform a robust mechanical design with desired thermal performance
  - Capsule temperature control compensated for low lobe power
  - All thermocouples, temperature control gas lines, sweep gas lines, and ion chambers functioned throughout Cycle 160A
    - Ensures target test environments are achieved and recorded for each pellet
- Flow-through capsule data provides insight on tritium release from pellets
  - Tritium entered gas phase upon reactor startup
    - No significant delay or incubation period observed
  - Tritium release increased with burnup and burnup rate (140 GVR > 70 GVR)
  - Addition of engineered porosity, large or small, enhances tritium release
  - Unclear if larger grains (> 10micron) offer a significant advantage relative to the nominal grain size (~2 micron)
  - Cermet pellets exhibit a slower initial release before approaching concentrations comparable to standard pellet designs at a lower test temperature (~50°C)
Cycle 160B

TMIST-3: Preliminary Results

Cycle 160B Reactor Operation
- December 19, 2016 to February 23, 2017
  - Scrammed January 18th thru 23rd
  - ~60 calendar days or EFPD at ~23 MW_th

![Graph showing power output over time](image-url)
TMIST-3: Preliminary Results

Cycle 160B Reactor Operation

- Nominal capsule thermal performance (standard 140 GVR closed capsule)

Temperature control gas composition adjusted for higher lobe power

- Gas mixtures typically less neon-rich than Cycle 160A
- Average temperature for Capsule 4A below lower tolerance limit
- Temperature control gas composition is dictated by Capsule 5A, which has the highest temperature among the capsule sharing this gas line

### TMIST-3A-6B Cycle 160B

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Flow Rate Data:

- Temperature control gas composition adjusted for higher lobe power
- Gas mixtures typically less neon-rich than Cycle 160A
- Average temperature for Capsule 4A below lower tolerance limit
- Temperature control gas composition is dictated by Capsule 5A, which has the highest temperature among the capsule sharing this gas line
TMIST-3: Preliminary Results

Cycle 160B

- Acceptability of the test train thermal performance
  - Majority of capsules achieved thermocouple readings within specified range
    - Temperature control gas compositions were adjusted to be less neon-rich to accommodate the higher lobe power (i.e., 23 vs. 20 MWth)
    - Average temperature of Capsule 4A is slightly below the lower specified limit
      - Temperature is controlled to Capsule 5A, the high temperature capsule on the common gas line with Capsule 4A

Overall, satisfactory thermal performance was achieved by the TMIST-3A test train during Cycle 160B

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TMIST-3: Preliminary Results

TMIST-3A Cycle 160B Ion Chamber Response

Solid Lines = 140 GVR
Dashed Lines = 70 GVR

- Reactor operations began at 8:45pm on December 19, 2016
- Tritium detected in gas phase as reactor brought to Nf = 1
Reactor operations began at 8:45pm on December 19, 2016
Tritium detected in gas phase as reactor brought to \( N_f = 1 \)

Higher concentrations detected for pellets with higher target burnups
Influence of pore size is remains unclear
Enhanced release from pellets with large pores observed at 70 GVR only
Standard pellet exhibits enhanced release relative to Large Grain pellet at target burnup of 140 GVR
- 140 GVR standard pellet similar to 70 GVR Large pore pellet

Liquid scintillation counting (LSC) of bubbler vials provides a quantitative measure of tritium in the gas stream
- General trends are similar ion chamber data
- Suggests that standard pellet designs are exhibiting enhanced tritium release relative to large grain and cermet pellet designs
Cycle 160A and 160B Conclusions

- Overall test train performance is successful
  - Reactor physics were satisfactorily captured and used to inform a robust mechanical design with desired thermal performance
  - Capsule temperature control compensated for varying lobe powers
  - All thermocouples, temperature control gas lines, sweep gas lines, and ion chambers functioned throughout Cycle 160A and 160B
    - Ensures target test environments are achieved and recorded for each pellet

- Flow-through capsule data provides insight on tritium release from pellets
  - Tritium entered gas phase upon reactor startup during Cycle 160A
    - No significant delay or incubation period observed
  - Tritium release increased with burnup and burnup rate (140 GVR > 70 GVR)
  - Addition of engineered porosity, large or small, enhances tritium release
  - Data suggest that larger grain (> 10micron) pellets may offer an advantage relative to the nominal grain size (~2 micron)
  - Cermet pellets exhibit a slower initial release before approaching concentrations comparable to standard pellet designs
    - Note the cermet pellets are at a slightly lower temperature
Motivation for improving current TPBAR performance models

- Current predictive code does not accurately predict tritium release to the reactor coolant system (RCS)
  - Initial prediction of ~1Ci/TPBAR is less than the observed ~4Ci/TPBAR
- Shape of measured tritium release is non-linear and reveals a delay/incubation period (~80-100 days) prior to measurable tritium release to the RCS
  - Disparity between measured and predictive curves indicates that underlying mechanisms are either not included or adequately captured by the model
TMIST-3: Pellet Performance Irradiation Experiment

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TMIST-3: Pellet Performance Irradiation Experiment

- TPBAR Materials Irradiation Separate Effects Test (TMIST)
  - Evaluate in-reactor performance of individual TPBAR components
- TMIST-3
  - Two test trains (3A and 3B) with a total of 41 pellet capsules
  - Tritium speciation and release kinetics as a function of temperature, time, burnup, burnup rate from pellets with standard and alternative designs
TMIST-3: Experiment Description

- Test Train Designs – 3A and 3B
  - Short (3A) and long (3B) term test trains to be irradiated sequentially to 300 and 600 EFPD at 23 MWth, respectively

Ex-Reactor Measurement Systems