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FITT: Individual Particle Silver Release Behavior at 1100–1600°C

DOE ART Gas-Cooled Reactor (GCR) Review Meeting
Virtual Meeting
July 25 – 27, 2023

ART
Advanced Reactor Technologies

It takes a village… critical staff supporting this effort

- John Hunn – program and technical guidance
- Darren Skitt – IMGA and furnace operation
- Zach Burns – furnace operation
- Grant Helmreich – statistical analysis
- Danny Schappel – data interpretation
- Irradiated Fuels Examination Laboratory – furnace operation/particle handling

Why do we care about silver release…

- "…the limiting criterion in a given reactor design is derived from the accessibility and maintainability of boilers/circulators/reformer tubes/inspection chambers." – Nabielek[1]

- ^{110m}Ag is of particular interest due to relatively rapid release and uncertainty surrounding our understanding of the release mechanism.

[1] H. Nabielek, P.E., Brown, P. Offermann, "Silver release from coated particle fuel," Nucl. Technol., 35, 483-493 (1977).

Goals for individual particle thermal exposure experiments

- Provide direct confirmation of silver release out-of-pile and insight on silver release kinetics and active mechanisms
  - Confirm elevated silver release below 1600°C observed during full compact tests
  - Expand range of temperature and times to observe release of fission products (FP) from intact particles
    - Alternative system is needed as safety testing (CCCTF and FACS) is limited by schedule and cost

Furnace for Irradiated TRISO Testing (FITT) for long-term* thermal exposure of individual TRISO particles

*relative to traditional safety testing

- Flexible, intentionally-simple, cost-effective capability to heat small batches of irradiated TRISO particles up to 1700°C over times >1500 h outside a hot cell
  - Closed-bottom ceramic tube in box furnace containing 10–30 particles under flowing inert gas *or oxidizing environments (up to 21% O_2)*
  - Installed in the Irradiated Fuels Examination Laboratory (IFEL) radiological facility at ORNL where AGR hot cell work is performed
  - **Intended to supplement integral release tests in the Core Conduction Cooldown Test Facility (CCCTF) and Fuel Accident Condition Simulator (FACS) systems**
  - R&D artifacts from system:
    - Skitt et al., Comparison of unirradiated and irradiated AGR-2 TRISO fuel particle oxidation response, JNM 580 (2023) 154409
    - Skitt et al., Oxidation Testing and Examination of AGR-2 Particles, ORNL/TM-2021/2092
    - Gerczak et al., AGR-2 Loose Particle Heating Tests in the Furnace for Irradiated TRISO Testing, ORNL/TM-2020/1715

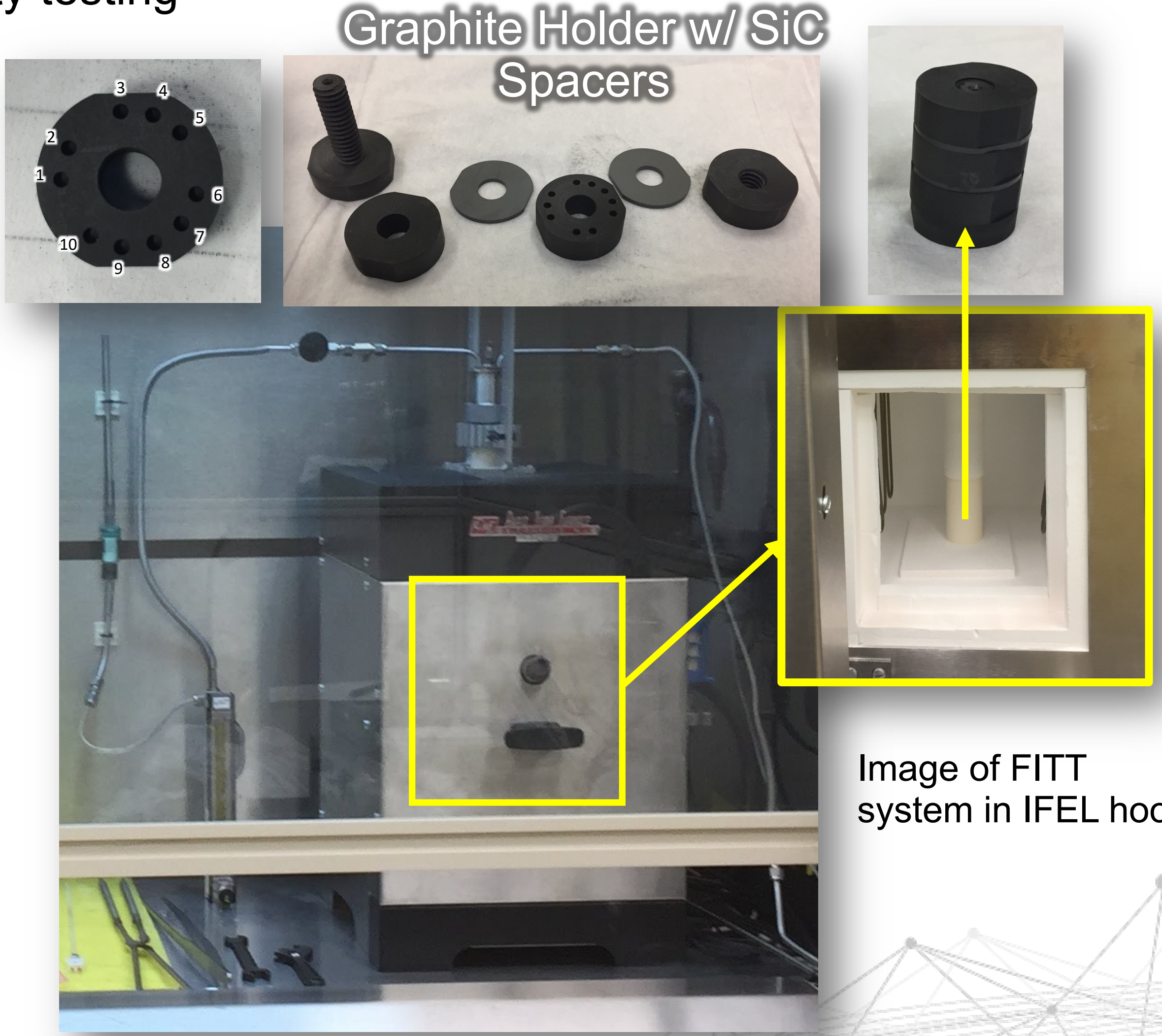
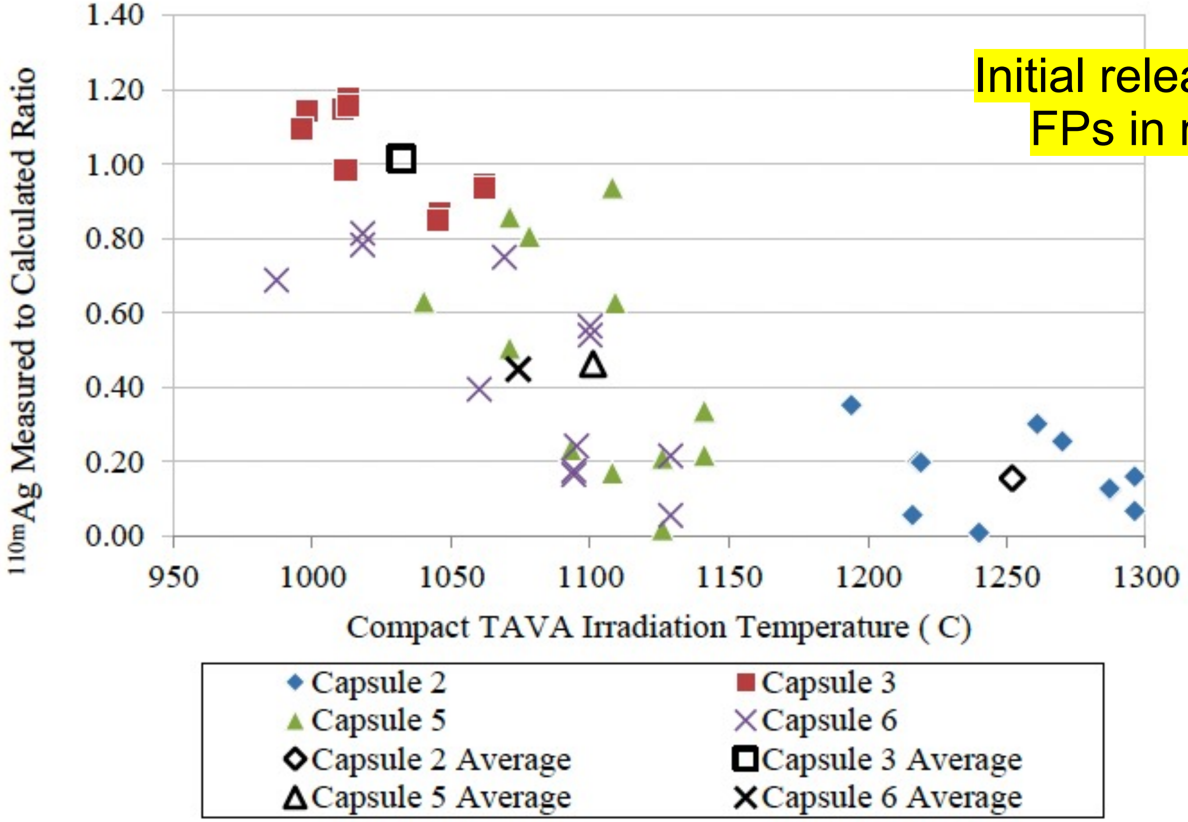
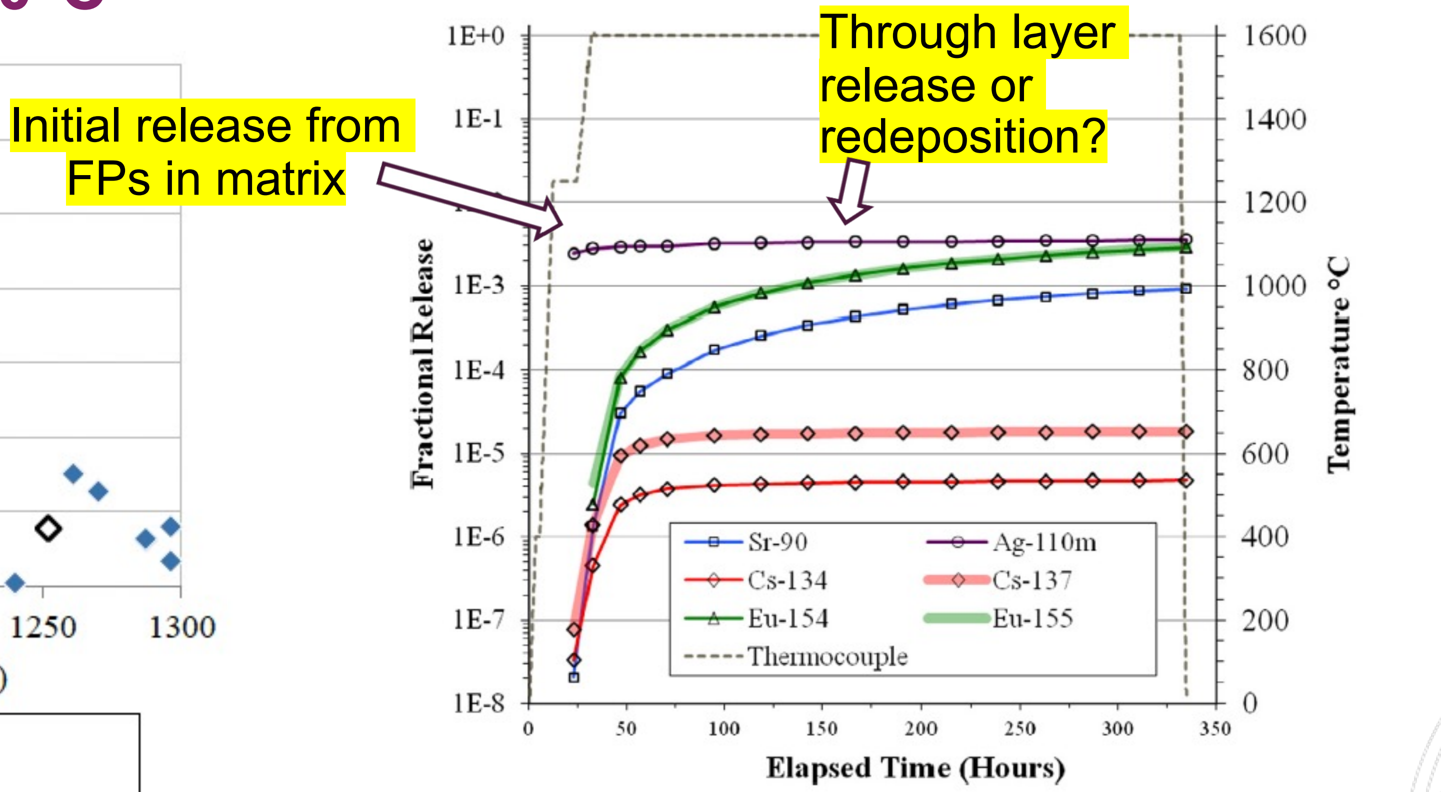


Image of FITT system in IFEL hood

In-pile release above 1100°C but challenges in interpreting release during out-of-pile safety testing at 1600°C



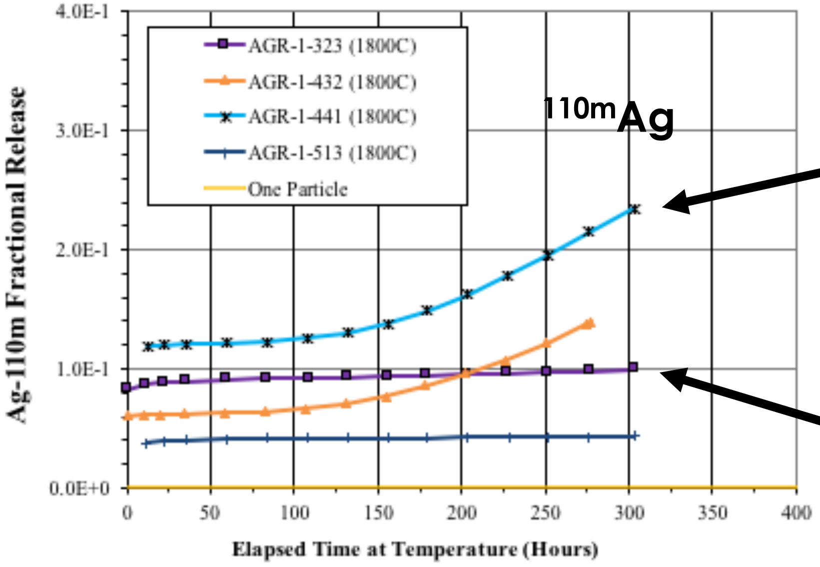
AGR-2 Measured-to-calculated ^{110m}Ag fraction in compact ($M_{compact}/C_{compact}$) versus compact time-average volume-average (TAVA) temperature (AGR-2 Final PIE Report, Stempien et al. 2016)[1]



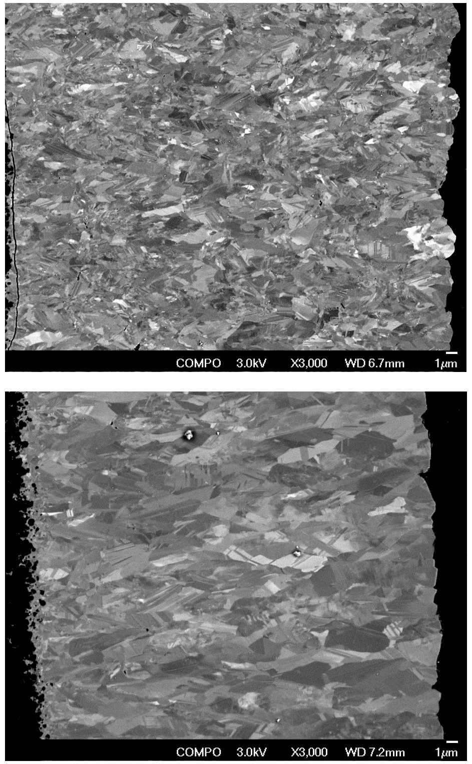
AGR-1 Compact 6-2-1 Fractional release (1600°C, 300 h) during safety testing showing initial release of silver and slow continuous release of other fission products (FP)s (e.g. Sr and Eu) presumed to be in matrix (Morris et al. 2016)[2]

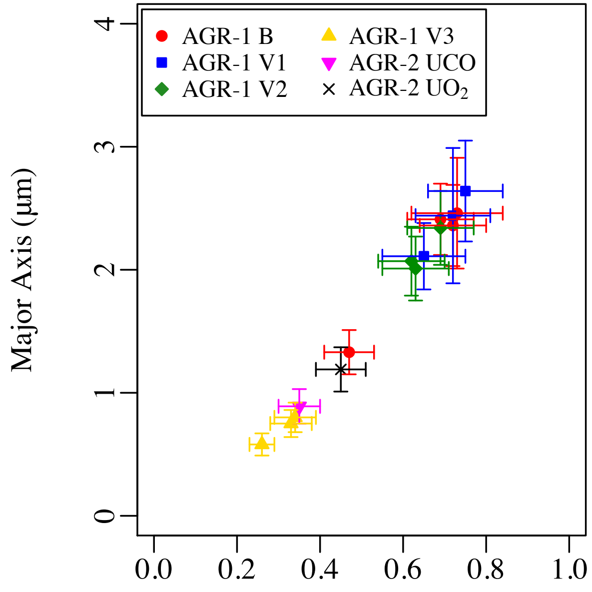
[1] Stempien et al., "AGR-2 TRISO Fuel Post-Irradiation Examination Final Report," INL/EXT-21-64279, (2021).
[2] R.N. Morris et al., “Performance of AGR-1 high-temperature reactor fuel during post-irradiation heating tests,” Nucl. Eng. Des., 306, 24-35 (2016).

But observed ^{110m}Ag release at 1800°C in out of pile safety tests



Fractional release measurements of ^{110m}Ag from AGR-1 Compacts exposed to 1800ºC for 300 h, Reproduced from Morris et al. (2016)[1]



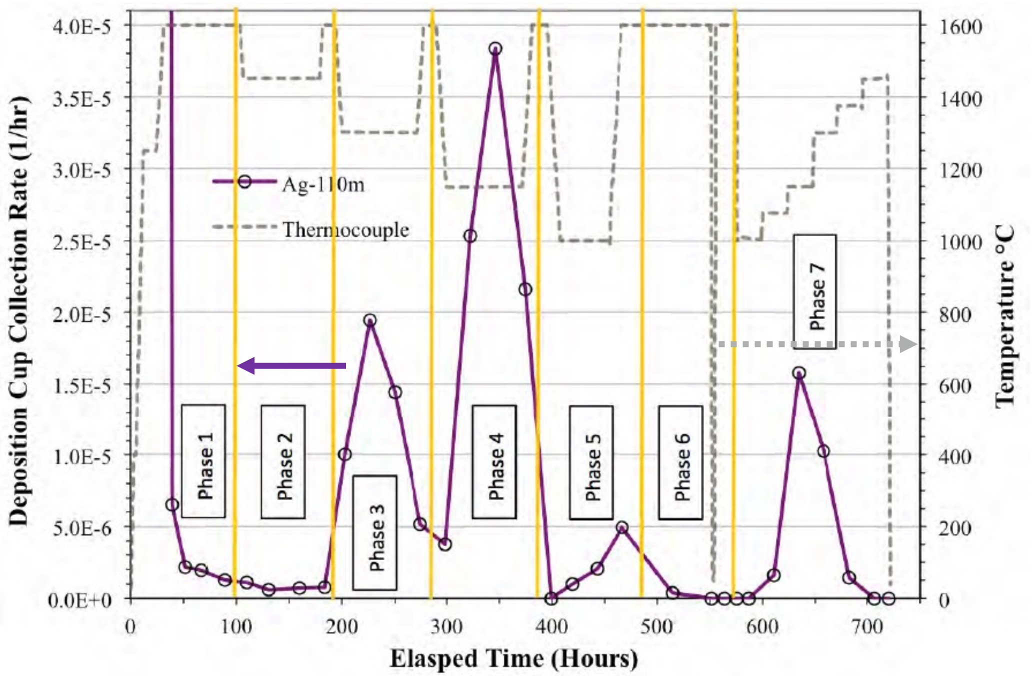


Grain size for each AGR batch. Reproduced from Gerczak et al. (2016) [2]

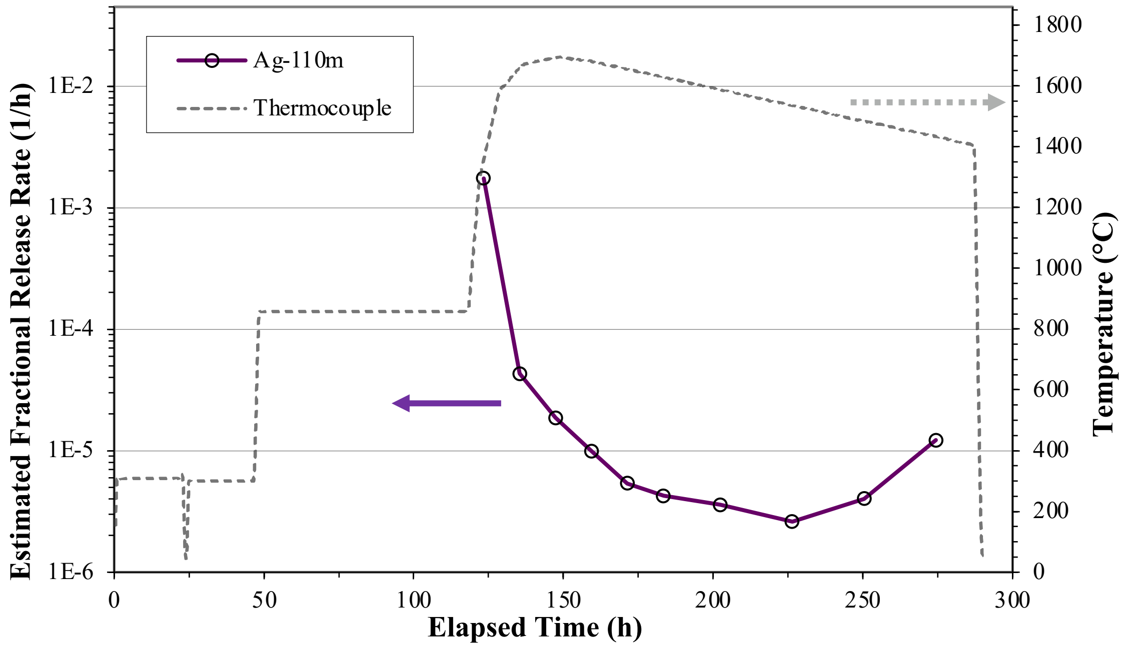
- SiC microstructure leads to varying FP release behaviors during safety testing
- Calculated increase in effective diffusion coefficient (D_{eff}) for fine grain is 3.1x versus large grain variants
- Grain boundary diffusion dominates out-of-pile transport mechanism for silver and likely strontium and europium (not shown)[2]

[1] R.N. Morris et al., "Performance of AGR-1 high-temperature reactor fuel during post-irradiation heating tests," Nucl. Eng. Des., 306, 24-35 (2016).
[2] T.J. Gerczak, et al. *SiC layer microstructure in AGR-1 and AGR-2 TRISO fuel particles and the influence of its variation on the effective diffusion of key fission products*, J. Nucl. Mater., 480 (2016), 257–270.

Expanded out-of-pile tests suggest a "Goldilocks" zone for silver release



^{110m}Ag deposition cup collection rate for AGR-1 Compact 4-2-2 (Hunn et al. 2015)[1]



^{110m}Ag fractional release rate for AGR-2 CCCTF transient safety test (Hunn et al. 2019)[2]

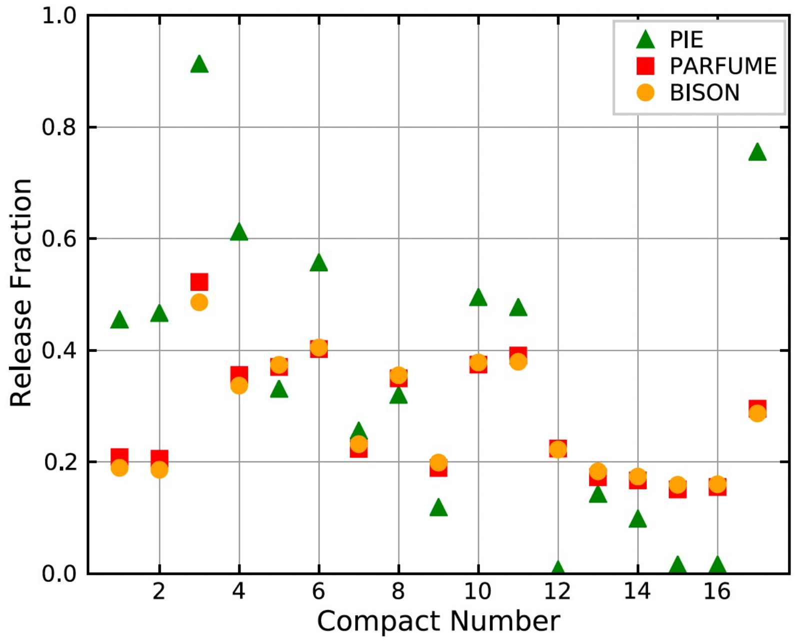
*Independently observed in AGR-1 transient test at INL by Stempien et al (2016)[3]

- Apparent maximum out of pile ^{110m}Ag release rate observed below 1600°C and between 1000–1400°C?

[1] Hunn, John D., Robert N. Morris, Charles A. Baldwin, Fred C. Montgomery, Tyler J. Gerczak, 2015. PIE on Safety-Tested AGR-1 Compact 4-2-2. ORNL/TM-2015/033, Revision 0. Oak Ridge: Oak Ridge National Laboratory.
[2] Hunn, John D., Robert N. Morris, and Zachary M. Burns. 2019. Transient Temperature Safety Test of AGR-2 UCO Compacts 5-1-1, 5-1-2, and 5-1-3. ORNL/TM-2019/1292, Revision 0. Oak Ridge: Oak Ridge National Laboratory.
[3] Stempien John D. Paul A. Demkowicz, Edward L. Reber, and Cad L. Christensen. 2016. "High-Temperature Safety Testing of Irradiated AGR-1 TRISO Fuel." Proc. 8th International Topical Meeting on High Temperature Reactor Technology (HTR-2016). Las Vegas, November 6–10, 2016.

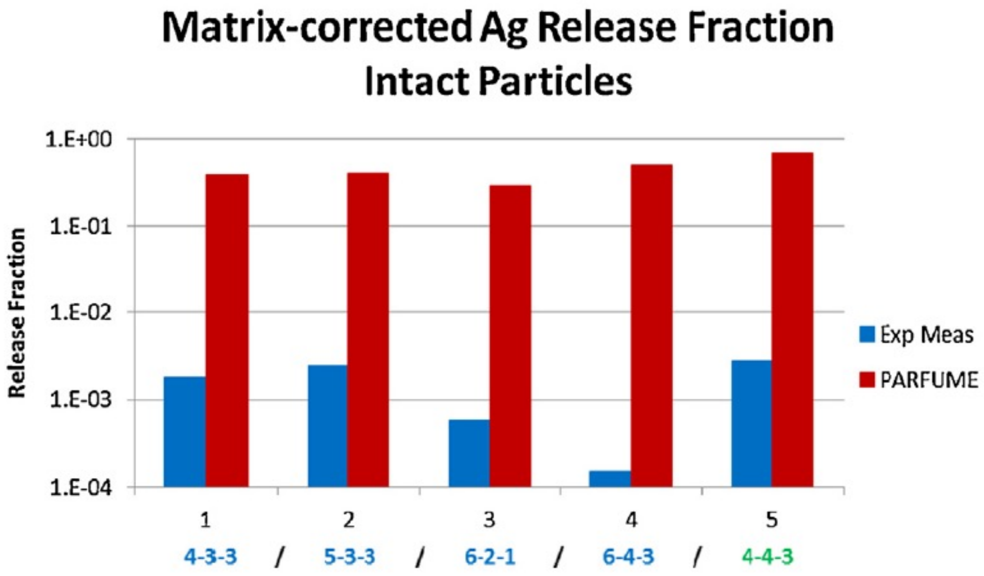
Silver release behavior likely influences modeling interpretations

AGR-1 In-pile release behavior



Compact silver release compared to PARFUME calculated release. Reproduced from Hales et al. (2021)[1]

Safety tested release behavior



Comparison of compact fractional release from experimental analysis and PARFUME for compacts with no failed particles. Reproduced from Collin et al. (2016)[2]

- BISON/PARFUME do a good job of estimating in-pile release for several cases[1], however, estimation of safety testing data is overestimated by orders of magnitude[2]
- Suggests disconnect with D_{eff} used to simulate in-pile performance and D_{eff} responsible for release in out-of-pile safety testing

[1] Jason D. Hales, Wen Jiang, Aysenur Toptan , Kyle A. Gamble, Modeling fission product diffusion in TRISO fuel particles with BISON, Journal of Nuclear Materials 548 (2021) 152840
[2] B.P. Collin et al. J. Nucl. Mater. 301, 378-390 (2016).

Goals for individual particle thermal exposure experiments: reiterated

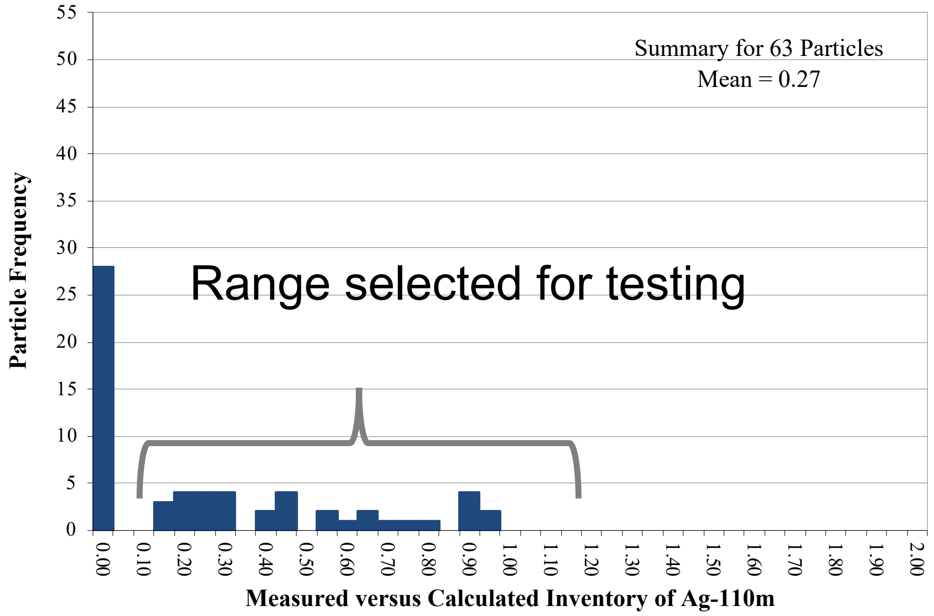
- Expand range of temperature and times to provide direct confirmation of silver release from intact particles
  - Confirm “Goldilocks” silver release between 1000ºC and 1400ºC
- Provide insight on kinetics and active mechanisms

AGR-5/6/7 testing to expand observations and confirm "Goldilocks" zone

Particles of interest

- AGR-5/6/7 Compact 2-2-1[1]
  - TAVA: 828/845°C, Burnup: 14.03% FIMA, Fast Fluence: 4.72×10^{25} n/m^2 (E>0.18 MeV)
- Target broad range in ^{110m}Ag to allow for bounds in particle retention behaviors
  - However, selection is biased as particles which had measurable ^{110m}Ag were selected for testing
- Release is confirmed via **direct observation** of change in FP inventory (ratio of pre- and post-test measured activity)

Example of ^{110m}Ag M/C distribution from AGR-5/6/7 Compact 2-2-1



$$^{110m}\text{Ag M/C} = \frac{A_i(^{110m}\text{Ag})}{A_{calc}(^{110m}\text{Ag}) \dfrac{A_i(^{137}\text{Cs})}{\sum_{i=1}^{n}\left(\frac{1}{n}\right) A_i(^{137}\text{Cs})}}$$

[1] B.T. Pham et al. "AGR 5/6/7 Irradiation Test Final As-Run Report" INL/EXT-21-64221 (2021).

AGR-5/6/7 testing to expand observations and confirm "Goldilocks" zone

Isochronal Testing (Completed)

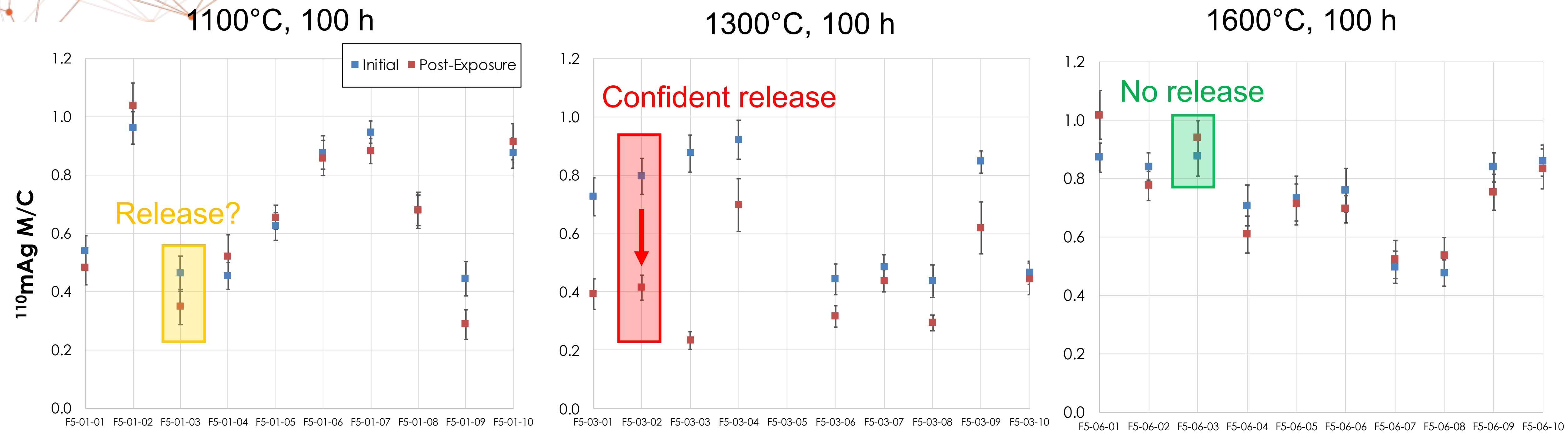
Temperature	Time	Particles
1100°C	100 h ✅ ✅	10× AGR-5/6/7 Compact 2-2-1
1200°C	100 h ✅ ✅	
1300°C	100 h ✅ ✅	
1400°C	100 h ✅ ✅	
1500°C	100 h ✅ ✅	
1600°C	100 h ✅ ✅	

Time Dependent Release Testing

Temperature	Time 1	Time 2	Time 3	Particles
1200°C	50 h ✅ ✅	25 h (75 h elapsed) 🔥	25 h (100 h elapsed)	10× AGR-5/6/7 Compact 2-2-1
1300°C	50 h ✅ ✅	25 h (75 h elapsed) ✅	25 h (100 h elapsed)	
1400°C	50 h ✅ ✅	25 h (75 h elapsed) ✅	25 h (100 h elapsed)	

🔥 Furnace testing in progress ✅ Furnace testing complete
✅ ✅ Furnace testing and IMGA analysis completed

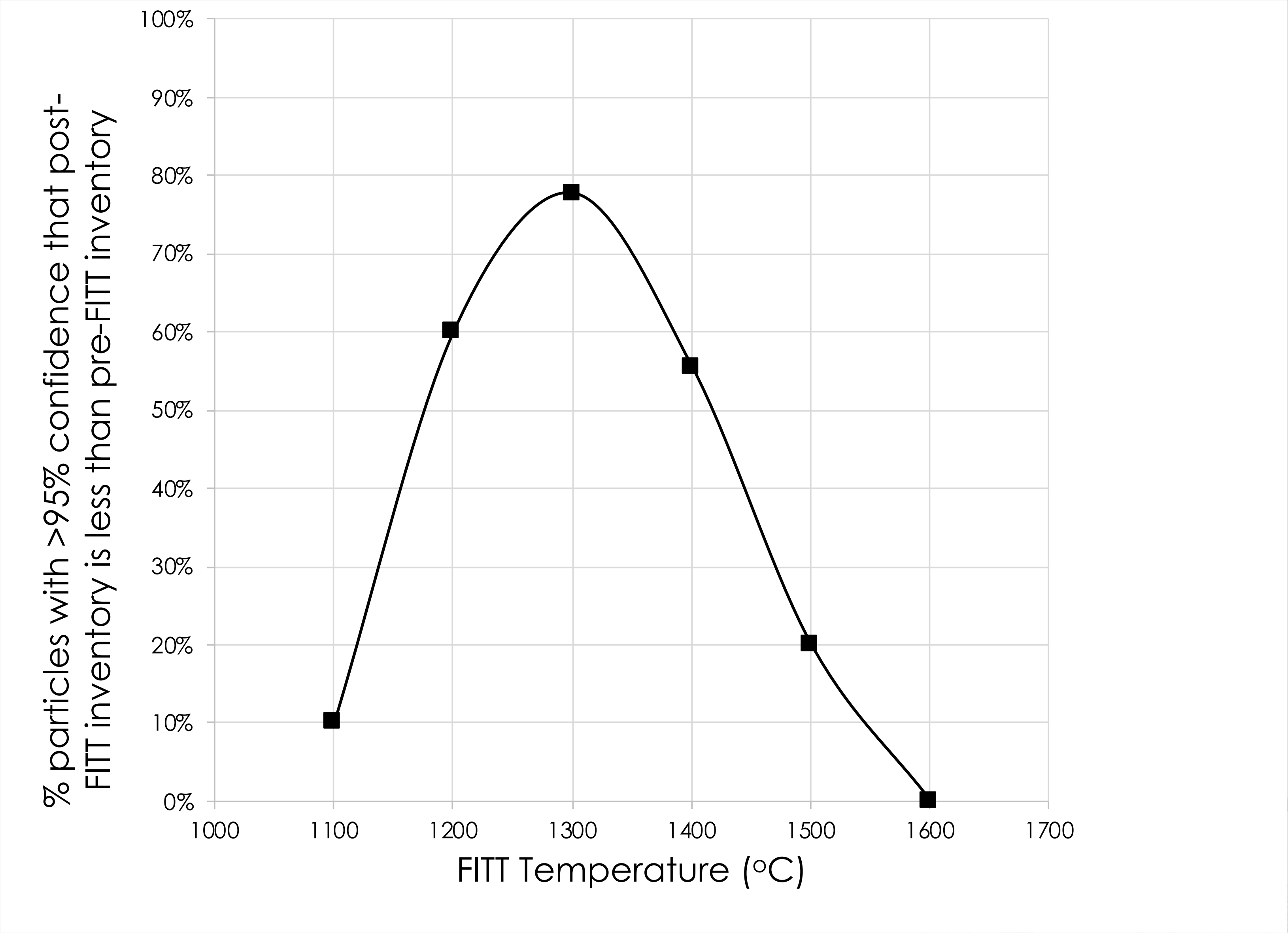
Direct confirmation of "Goldilocks" zone for release



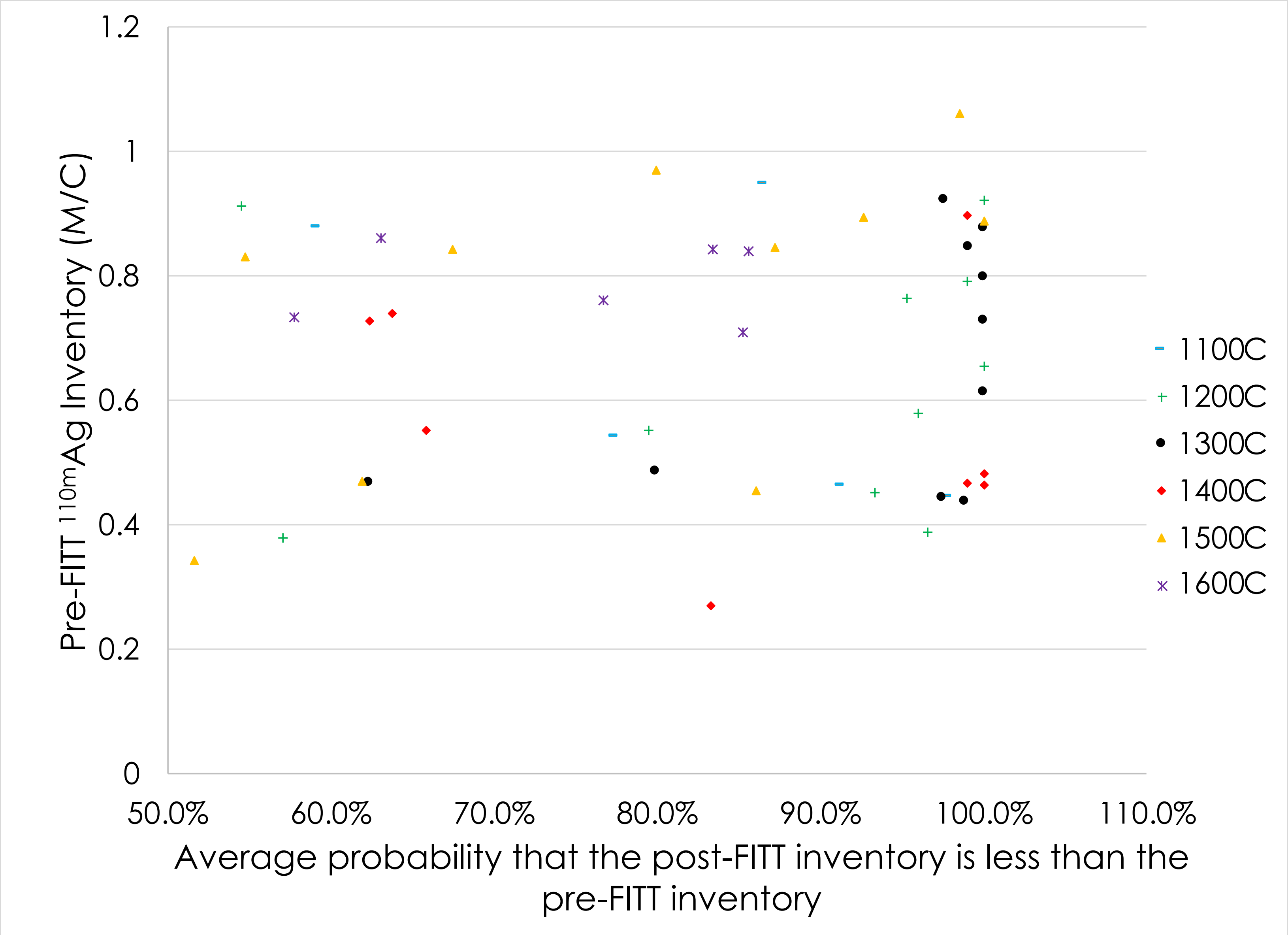
- Silver release observed with apparent temperature dependence
- Apparent particle to particle variation
  - What constitutes confident release?

Direct confirmation of “Goldilocks” zone for release

- By addressing measured uncertainty…
- Confirm temperature dependence with 1300°C representing an apparent maximum in ^{110m}Ag release after 100 h
- Confirmed a particle-to-particle variation in release response

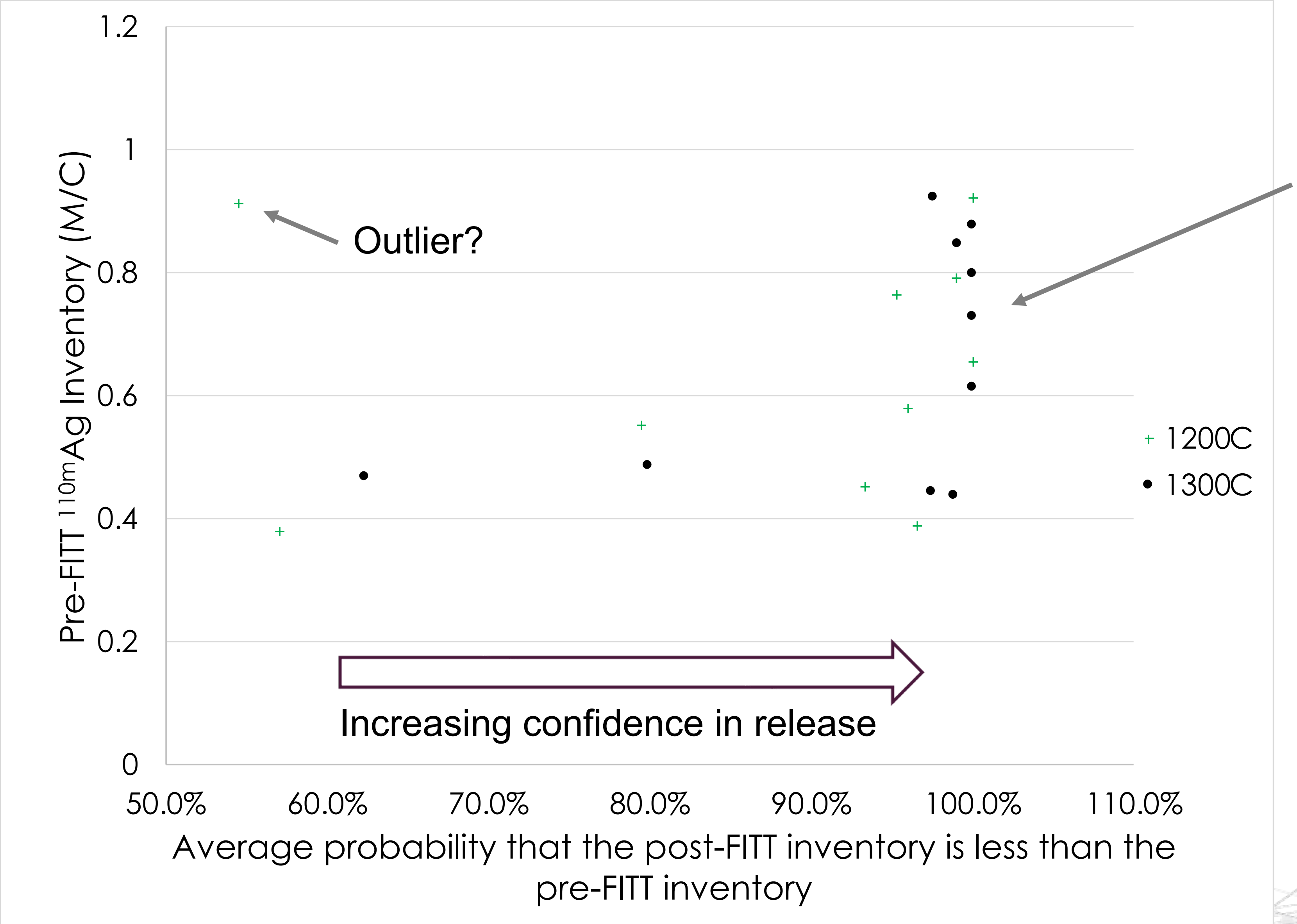


What particles are susceptible to release?



Note: 50% probability indicates pre- and post-FITT inventories are equal.

What particles are susceptible to release?

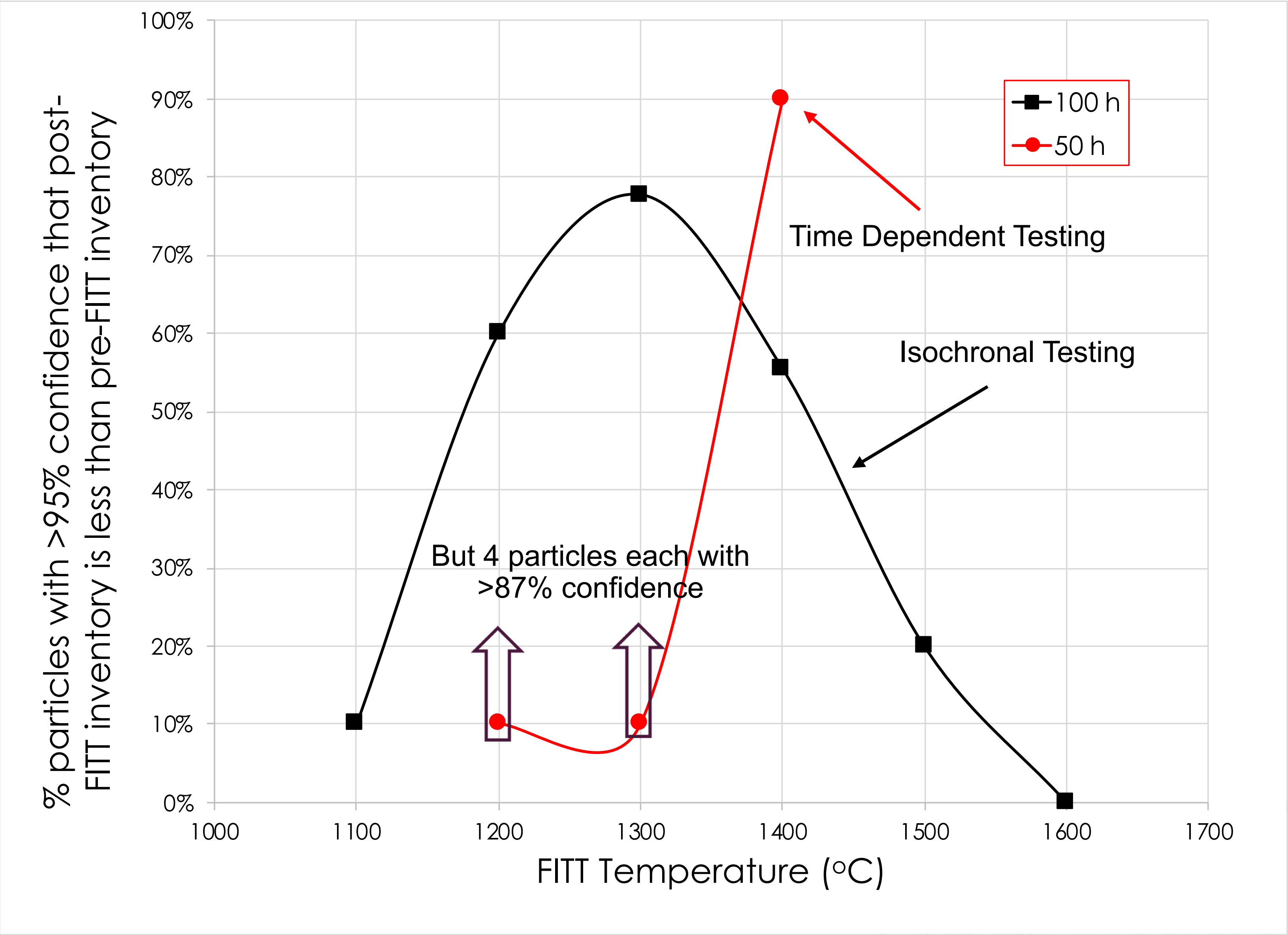


Higher starting inventory appears easier to measure release more confidently

Note: 50% probability indicates pre- and post-FITT inventories are equal.

What can we learn regarding time dependent release?

- 50 h exposures completed (1200–1400°C)
- 25 h time steps to be completed next (elapsed time 75 h then 100 h) tracking the same particles
- Our goal is to learn about the fraction of silver inventory available to be released from intact TRISO fuel particles
- Will the 1200°C and 1300°C continue to release silver and eventually reflect the isochronal testing observations?
- How do account for particle to particle variation when attempting to assess kinetics?



Discussion and takeaways

- FITT isochronal exposure experiments confirm the presence of a “Goldilocks” temperature dependence in the range of 1100–1600^{o}C
- This elevated release at FITT temperatures relative to safety testing suggests multiple diffusion mechanisms are active regarding silver release
  - Lower temperature release likely not dominated by thermal grain boundary diffusion
- Early in data interpretation and completion of the time dependence release behavior study is needed to more comprehensively understand silver release in this “Goldilocks” zone
  - Need to better establish D_{eff} calculation and comprehensively compare with Ag release data
  - Limited destructive analysis (cross-sectional analysis with imaging and compositional analysis) to understand particle to particle variation is release response
- Thoughts: Modeling & Simulations tools are extremely powerful for accelerating fuel development and qualification
  - This is an opportunity to communicate complex phenomena and relevant data to help refine models
  - Need to account for uncertainties in experimental results and isolate challenges with integral experiments
    - Complex temperature histories and temperature gradients result in particle-to-particle silver retention variation

Thank you

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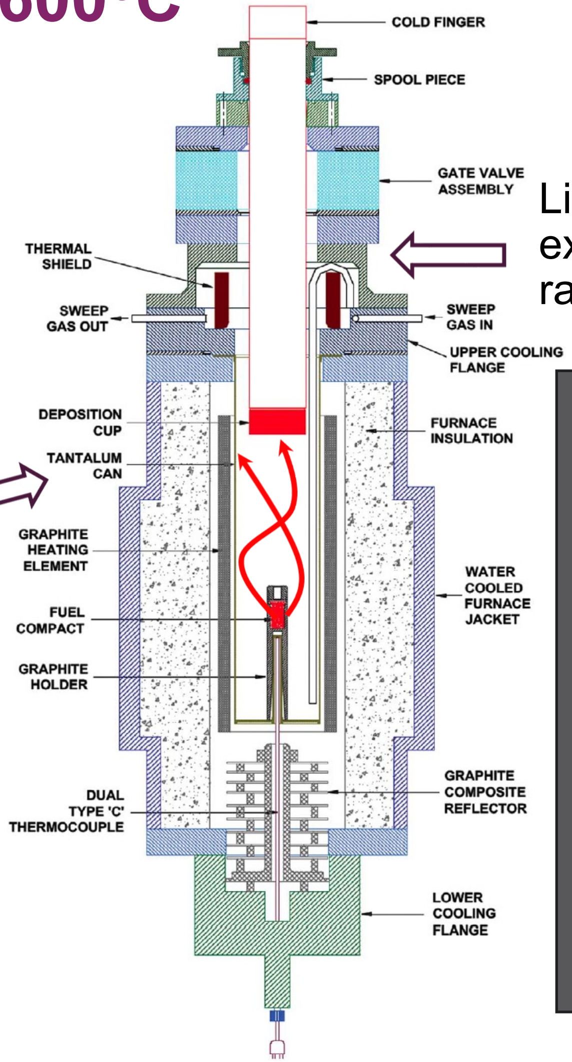
- This work was sponsored by the US Department of Energy Office of Nuclear Energy through the Advanced Gas Reactor Fuel Development and Qualification Program.

Matrix retention masks continued silver release during safety testing at 1600°C

Requires determination of collection efficiency at different temperatures



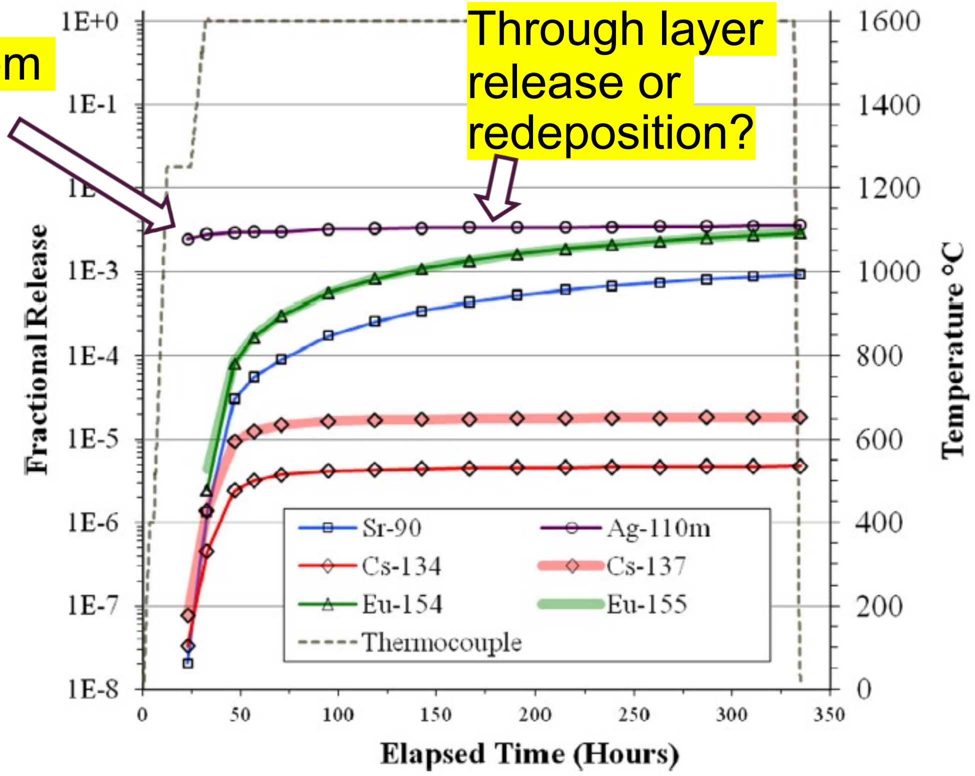
Limits on cup exchange rates

Schematic of the CCCTF furnace (Baldwin et al., 2014)[1]

Plate out on cold finger requires multiple transport pathways and reflects FPs sourced from different components- real challenge in interpreting through layer release from intact particles from compact tests

Initial release from FPs in matrix

Through layer release or redeposition?



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[1] Baldwin, C.A., Hunn, J.D., Morris, R.N., Montgomery, F.C., Silva, C.M., Demkowicz, P.A., 2014. First elevated-temperature performance testing of coated particle fuel compacts from the AGR-1 irradiation experiment. Nucl. Eng. Des. 271, 131–141.
[2] R.N. Morris et al., "Performance of AGR-1 high-temperature reactor fuel during post-irradiation heating tests," Nucl. Eng. Des., 306, 24-35 (2016).