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**Blister Threshold Based Thermal Limits for the U-Mo Monolithic
Fuel System**

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ABSTRACT

1. Introduction

Fuel failure is most commonly induced in research and test reactor fuel elements by exposure to an under-cooled or over-power condition that results in the fuel temperature exceeding a critical threshold above which blisters form on the plate. These conditions can be triggered by normal operational transients (i.e. temperature overshoots that may occur during reactor startup or power shifts) or mild upset events (e.g., pump coastdown, small blockages, mis-loading of fuel elements into higher-than-planned power positions, etc.). The rise in temperature has a number of general impacts on the state of a fuel plate that include, for example, stress relaxation in the cladding (due to differential thermal expansion), softening of the cladding, increased mobility of fission gases, and increased fission-gas pressure in pores, all of which can encourage the formation of blisters on the fuel-plate surface. These blisters consist of raised regions on the surface of fuel plates that occur when the cladding plastically deforms in response to fission-gas pressure in large pores in the fuel meat and/or mechanical buckling of the cladding over damaged regions in the fuel meat. The blister temperature threshold decreases with irradiation because the mechanical properties of the fuel plate degrade while under irradiation (due to irradiation damage and fission-product accumulation) and because the fission-gas inventory progressively increases (and, thus, so does the gas pressure in pores).

Analytical simulations of thermal events have been performed to demonstrate that, while at low temperatures, the fuel meat is held in compression by the cladding (due to residual stress from the fabrication process) [1]. While in the compressive state, the fission-gas inventory and pore/defect size must be relatively large to overcome the constraint of cladding and induce blistering or pillowing (behavior typically characterized as breakaway swelling unless attributed to significant fabrication defects). As the temperature of the fuel plates increased, the aluminum cladding expands more than the

fuel meat due to differences in the coefficient of thermal expansion (CTE). If a sufficiently high temperature is reached, this expansion mismatch ultimately relieves the compressive stress applied by the cladding and puts the fuel meat into tension.

This stress state transformation significantly affects the fuel meat. Prior to the increase in temperature, the fission gas bubbles¹ within the fuel meat have achieved a quasi-equilibrium state during irradiation, where the external constraint of the material surrounding the bubble matches the stress applied by the gas pressure within the bubble. The bubbles grow over time as the gas inventory increases during irradiation and the external constraint remains relatively constant. An increase in temperature upsets this balance due to relaxation of the external constraint, and the bubbles expand slightly. This relatively sudden growth may lead to formation of micro-cracks in the fuel meat. This affect is further exacerbated by the increase in gas pressure that accompanies the increasing temperature². In regions where the fission-gas-bubble density is very high and the mechanical constraint is neutral or tensile, the potential for interconnected crack formation increases, and the resistance to blister formation drops as gas bubbles may coalesce into macroscopic defects.

This behavior was observed in the AFIP-6 irradiation experiment [ii]. This experiment was designed to demonstrate monolithic fuel performance at very high fission rates and, due to the thermal hydraulic design, operated at excessive cladding surface temperatures. This over-temperature condition led to accelerated oxide growth that insulated the fuel plate from the coolant. During the irradiation, the oxide layer grew exponentially thicker and progressively drove the peak local fuel-meat temperatures to ~400°C (although projections are difficult due to uncertainty in thermal properties of the aluminum oxide layer). Eventually the fission-gas inventory and mobility were sufficient in high-burnup regions of the fuel meat to allow localized coalescence into larger defects (on the order of several millimeters) with fission-gas inventories large enough to deform the cladding and form blisters (as shown in Figure 1).

¹ Typically, only bubbles larger than 1 micron are considered here because the gas pressure in smaller size bubbles is dominated by surface tension effects rather than external constraint.

² It is assumed that small increases in temperature do not accelerate gas diffusion rates meaningfully

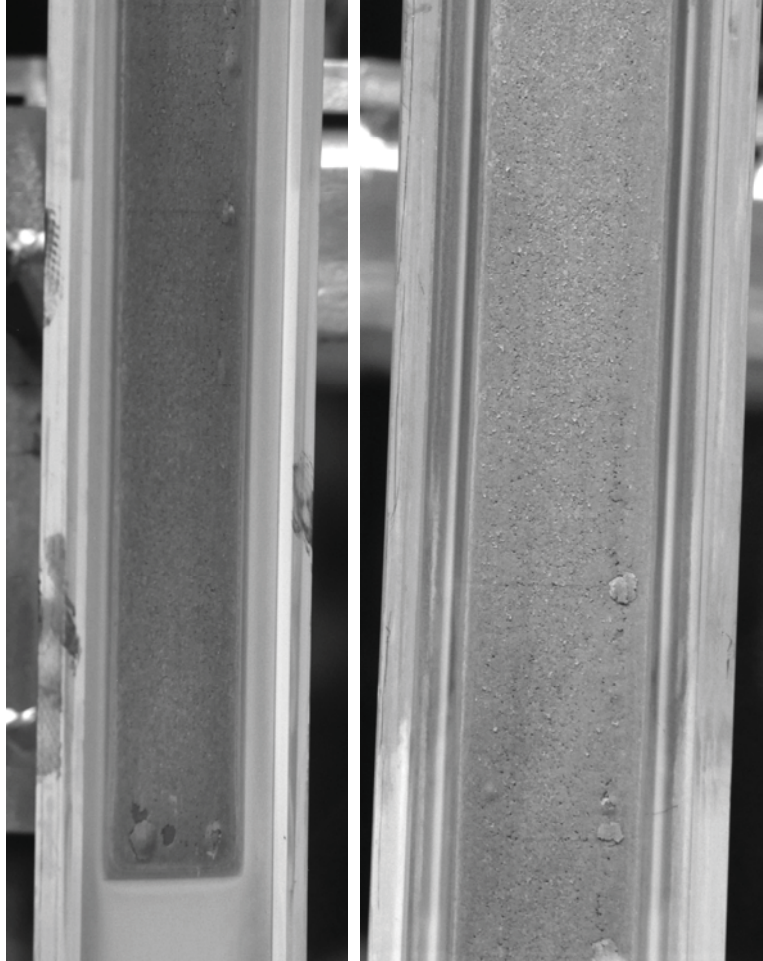


Figure 1. Irradiation induced blisters on the surface of the AFIP-6 experiment.

When blisters form, the mechanical deformation can be large enough to rupture the cladding and immediately release accumulated fission gases followed by a slow release of additional gaseous and solid fission products into the coolant. In AFIP-6, this process resulted in observation of a series of discrete fission-product-release indications over the course of several days. Release amounts were well within the administrative limits of the ATR but were clearly visible using standard monitoring practices.

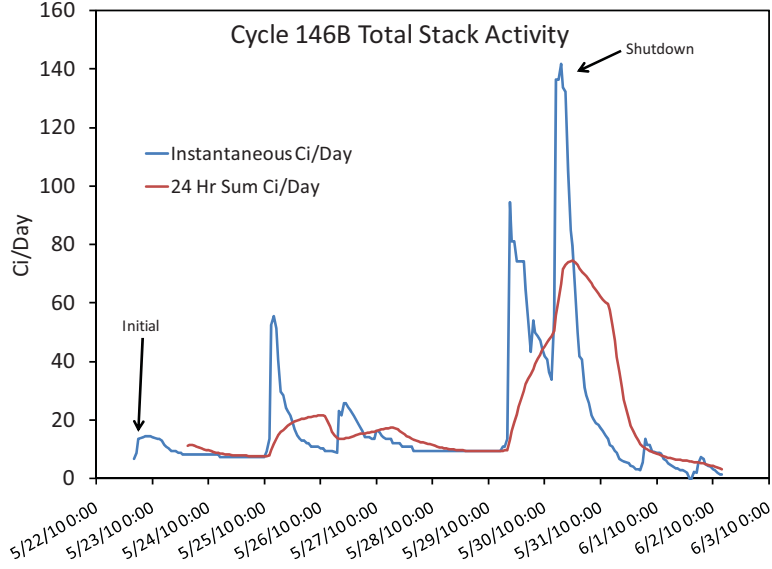


Figure 2. ATR stack gas activity measured by the Real Time Monitor during ATR cycle 146B.

The historic explanation for blister formation is consistent with these observations. A simple model describing fission-gas-driven blister formation was proposed in [iii], based on the stress in a circular disk with fixed edges and uniform pressure applied to the bottom surface of the disk. The maximum stress in the disk can be represented by the following equation.

$$\sigma = \frac{3Pa^2}{4h^2} \quad (1)$$

where P is the applied force (the gas pressure), a is the disk diameter (the defect size), and h is the disk thickness (the cladding thickness). If the stress σ in the disk (cladding) exceeds the yield strength σ_y , the disk will deform and produce a blister.

The gas pressure is a function of the quantity of fission gas generated, its ability to migrate into the open defect, and the temperature of the gas and can be represented by the ideal gas law,

$$P = \frac{nRT}{V} \quad (2)$$

where the volume V refers to the defect volume which is proportional to the defect size ($V \propto a$) and the number of gas atoms in the volume n is roughly proportional to the fission density ($n \propto \phi''$).

The yield strength of the cladding is also a strong function of temperature and decreases rapidly as the melting point is approached. The blister equation can then be reduced to (after solving for temperature)

$$T_{blister} \propto \frac{h^2 \sigma_y}{a f''} \quad (3)$$

Based on this model, the blister threshold would be expected to increase as the cladding thickness increased and to decrease as the defect size and fission density increased. The maximum gas-driven blister threshold is likely set by the limits of the aluminum as the plate temperature approaches its melting point (e.g. ~550-650°C). Variations in the blister threshold historically observed for different dispersion fuel-plate designs could readily be explained by this model. The threshold should be tightly coupled to variations in fission-gas-retention attributes and the fuel material's inherent resistance to defect formation and growth.

This model suggests that the blister threshold is a function of the fuel-plate design (cladding thickness), irradiation history (fission gas content), and the relative resistance to defect formation. The defect size at failure should be a strong function of fission-gas availability, the stress state of the fuel plate, and mechanical properties of the fuel meat.

Fission-gas availability can clearly be affected by fuel-plate microstructure. In irradiated U-Mo alloys, the fission gases are distributed between intragranular and intergranular bubbles. The behavior of the fuel meat is dominated by the larger intergranular gas bubbles that form directly on the grain boundaries. The gas in these bubbles can more readily coalesce into large bubbles than can the nanometer-sized bubbles distributed within the grains. The integrated amount of gas in these bubbles is roughly proportional to the grain boundary density; thus, as the grain size drops, the relative amount of gas in these bubbles would be expected to increase. It is important to note that the presence of intergranular bubbles is also expected to reduce the fuel meat's resistance to crack propagation. This contributes to the marked increase in brittleness observed after irradiation. This would presumably allow the defects to propagate much more easily in a fine-grained than in a coarse-grained fuel meat.

However, it is critical to recognize that small, bubble-based defects will not grow to a critical size without a macroscopic stress driver. Fission-gas accumulation occurs so slowly over the course of irradiation that irradiation-enhanced creep effects easily manage the stresses. The specifics of the thermally induced stresses during a given event must be evaluated to determine where and under what conditions defects are expected to form. However, it can be assumed that blistering generally occurs when the applied stress (σ) exceeds material strength (σ^*), as describe in Figure 3.

Blister Formation

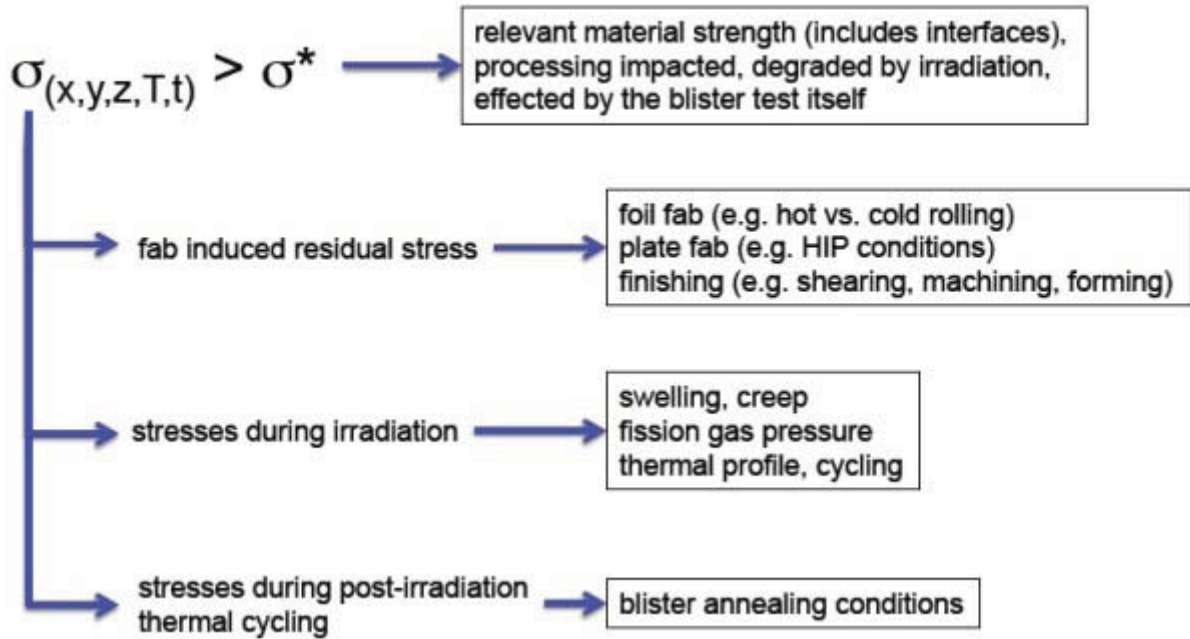


Figure 3. General condition for blister formation.

1.1.1 Out-of-Pile Blister Simulation

The direct experimental assessment of a fuel design's resistance to irradiation induced blistering under all potential thermal conditions would be extremely unrealistic. A simple test that bounds the stresses applied to the fuel plate during these events has therefore been used for decades to establish a universal blister threshold.

In-pile testing to determine the blister threshold in plate-type fuel is very difficult and expensive and has historically been deemed impractical for research and test reactor fuel qualification testing. The post-irradiation blister anneal test was consequently developed as a conservative simulation of the behavior and has been routinely used for many research-reactor fuel designs currently in use $[^{iv}, ^v]$. In this test, the irradiated fuel plate is inserted into a furnace at a given temperature, soaked for a period of time, and removed for inspection. If no visible blisters appear, the furnace temperature is increased, and the plate is soaked and inspected again. This process continues until blisters are observed.

The test is assumed to be conservative relative to an in-pile blister test for several reasons. First, the thermal- expansion-induced tensile stress in the fuel meat is maximized when the cladding temperature is equal to or greater than the fuel temperature (which occurs naturally during furnace heating), because cladding strength is relatively low and differential thermal expansion stresses are maximized (since aluminum has a significantly higher thermal expansion coefficient than does the fuel). In addition, the fission-gas availability in-pile is reduced by resolution of fission gases out of gas bubbles and back into solid solution due to gas atom collisions with neutrons (thereby reducing

the gas pressure within bubbles). An attempt to benchmark this methodology using an in-pile device was unsuccessfully attempted in the 1970s due to the complexity of the test methodology [vii].

The blister test as a simulation of in-pile over-temperature events does have some important limitations. For example, the total amount of fission gas that may diffuse into a defect is dependent on both temperature and time. While a fuel plate in a compromised position in pile may remain at power for very long periods of time (e.g. days, weeks or even months), practical limitations on the time spent in the hot cell furnace typically limit test durations to less than an hour. The absence of irradiation may also impact performance. In the case of U-Mo alloys, fission events are known to encourage the formation of a meta-stable gamma phase in the material that is crucial to the stability observed during irradiation. Exposure to temperatures in the 300-500°C range without irradiation could lead to premature decomposition of the alloy into the alpha phase and the release of fission gas. As mentioned previously, the overall temperature distribution in the fuel plate has a significant affect of its stress state, which drives the formation and growth of blisters. In a dispersion fuel meat, the temperature and stress mismatch occurs at the particle level and is only weakly impacted by the global stress conditions. In the monolithic fuel plate, the stress is integrated over the plate and may lead to large stress concentrations that are amplified by the heating mode (i.e. internal vs. external). In sum, all of these considerations suggest that the blister testing methodology is a conservative test that is even more conservative for the monolithic fuel design than for the dispersion fuel designs.

1.1.2 Monolithic Fuel Plate Blister Threshold Testing

The standard blister threshold method developed for dispersion fuel designs was applied to the newly developed U-Mo monolithic fuel system. Blister annealing studies on the U-Mo monolithic fuel plates began in 2007, with the Reduced Enrichment for Research and Test Reactors (RERTR)-6 experiment, and they have continued as the U-Mo fuel system has evolved through the research and development process.

Preliminary blister anneal threshold temperature measurements were performed on plates from early irradiation experiments (RERTR-6 through RERTR-10) and results ranged from 400 to 500°C (see Table 1) and showed the expected relationship with fission density. These temperatures were consistent with those observed in dispersion fuel designs and are projected to be acceptable for NRC-licensed research reactors and the high-power Advanced Test Reactor (ATR) and the High Flux Isotope Reactor (HFIR) based on current safety-analysis reports (SARs).

Table 1. Blister threshold temperature data for monolithic fuel plates from the RERTR-6, 7, 9A, 9B, 10A, and 10B experiments.

| Experiment | Plate ID | Test Train Position | Fuel Foil Thickness (inches) | Diffusion Barrier | Plate Bonding Method | Average Fission Density, $\times 10^{21}$ (fissions/cm ³) | Peak Fission Density ² , $\times 10^{21}$ (fissions/cm ³) | Measured Blister Temperature (°C) | Blister Types ⁹ |
|------------|----------|---------------------|------------------------------|-------------------|----------------------|---|--|-----------------------------------|----------------------------|
| RERTR-6 | L2F020 | A-4 | 0.020 | None | FB ⁴ | 2.45 | 4.53 | 450 | 2 |

| | | | | | | | | | |
|-----------|---------------------|-----|-------|--------------------|------------------|------|------|------------------|---|
| RERTR-6 | N1F060 ¹ | C-6 | 0.010 | None | FB | 3.63 | 6.90 | 450 | 2 |
| RERTR-7 | L1F110 ⁸ | C-3 | 0.010 | None | FB | 5.06 | 12.1 | 400 | 2 |
| RERTR-9A | L1F27C ³ | A-5 | 0.010 | None | FB | 3.64 | 9.64 | 425 | 2 |
| RERTR-9B | L1F330 | B-2 | 0.010 | None | FB | 5.99 | 14.6 | 400 | 2 |
| RERTR-9B | L1F35T | D-1 | 0.010 | Al-Si ⁵ | FB | 6.67 | 14.3 | 425 | 2 |
| RERTR-9B | L1P10T | D-5 | 0.010 | Zr | HIP ⁶ | 5.71 | 12.1 | 400 | 2 |
| RERTR-10A | L1P30Z ⁸ | A-1 | 0.010 | Zr | HIP | 2.88 | 6.41 | 400 | 2 |
| RERTR-10A | L2P15Z | A-3 | 0.020 | Zr | HIP | 1.34 | 3.82 | 475 | 1 |
| RERTR-10B | L2F47Z | D-7 | 0.010 | Zr | FB | 1.75 | 3.72 | 500 | 1 |
| RERTR-10B | L2F46Z ⁷ | B-6 | 0.020 | Zr | FB | 2.25 | 4.52 | 350 ⁷ | 1 |

- 1- U-7Mo alloy was used
- 2- Estimated using beginning of life (BOL) plate power peaking factors
- 3- Information for plate L1F27C is preliminary and requires additional verification
- 4- Friction Bonding
- 5- Al-Si thermal spray
- 6- Hot Isostatic Press
- 7- Blister tested in 2012
- 8- Blistered at starting temperature of the blister anneal test
- 9- Confirmation is required via optical metallography of plate cross-sections through blisters

However, subsequent results from the RERTR-12 blister anneal plates showed a decrease in the blister-threshold temperatures. These temperatures range from 300 to 400°C (see Table 2). Later, blister testing results from selected plates from the AFIP-4 experiment—which used a fabrication process similar to that of RERTR-12—fell within the same blister-threshold temperature range measured in the RERTR-12 plates.

Table 2. Blister threshold data from the RERTR-12 and AFIP-4 experiments.

| Experiment | Plate ID | Test Train Position | Fuel Foil Thickness (inches) | Diffusion Barrier | Plate Bonding Method | Average Fission Density, $\times 10^{21}$ (fissions/cm ³) | Peak Fission Density ¹ , $\times 10^{21}$ (fissions/cm ³) | Measured Blister Temperature (°C) | Blister Types ³ |
|---------------|---------------------|---------------------|------------------------------|-------------------|----------------------|---|--|---|----------------------------|
| RERTR-12 (X1) | L1P460 | C-4 | 0.010 | Zr | HIP | 2.35 | 3.98 | 400 | 1,2 |
| RERTR-12 (X1) | L1P592 | C-7 | 0.010 | Zr | HIP | 2.69 | 5.09 | 350 | 2 |
| RERTR-12 (X2) | L1P463 | D-8 | 0.010 | Zr | HIP | 2.86 | 4.86 | 350 | 1 |
| RERTR-12 (X2) | L1P595 | D-7 | 0.010 | Zr | HIP | 3.41 | 6.38 | 325 | 2 |
| RERTR-12 (X2) | L1P758 ² | D-6 | 0.010 | Zr | HIP | 5.00 | 9.20 | 300 | 2 |
| RERTR-12 (X1) | L1P774 | C-5 | 0.010 | Zr | HIP | 5.59 | 7.49 | 325 | 2 |
| RERTR-12 (X1) | L1P772 | C-1 | 0.010 | Zr | HIP | 5.77 | 7.83 | 325 | 2 |
| RERTR-12 (X2) | L1P756 | D-5 | 0.010 | Zr | HIP | 7.05 | 9.10 | 300 | |
| | | D-1 | 0.010 | Zr | HIP | | | blistered in the core at 205.3 °C (est. peak T) | 2 |
| RERTR-12 (X2) | L1P754 | | | | | 8.13 | 11.9 | | |
| | | B-5 | 0.010 | Zr | HIP | | | blistered in the core at 210.1 °C (est. peak T) | TBD |
| | | | | | | 9.15 | 13.08 | | |
| RERTR-12 (X3) | L1P785 ⁴ | | | | | | | | |
| AFIP-4 | L1H34Z | B-1 | 0.013 | Zr | HIP | 2.51 | 2.99 | 350 | 2 |

| | | | | | | | | | |
|--------|--------|-----|-------|----|-----|------|------|-----|---|
| AFIP-4 | L1H36Z | B-6 | 0.013 | Zr | HIP | 4.45 | 5.03 | 300 | 2 |
| AFIP-4 | L1B33Z | A-2 | 0.013 | Zr | FB | 4.06 | 4.59 | 300 | 2 |
| AFIP-4 | L1B51Z | A-4 | 0.013 | Zr | FB | 4.56 | 5.20 | 300 | 2 |

- 1- Estimated using beginning of life (BOL) plate power peaking factors
- 2- Blistered at starting temperature of the blister anneal test
- 3- Information is preliminary, additional metallography work is required to confirm types of the blisters
- 4- Post Irradiation Examination of plate L1P785 has not been performed

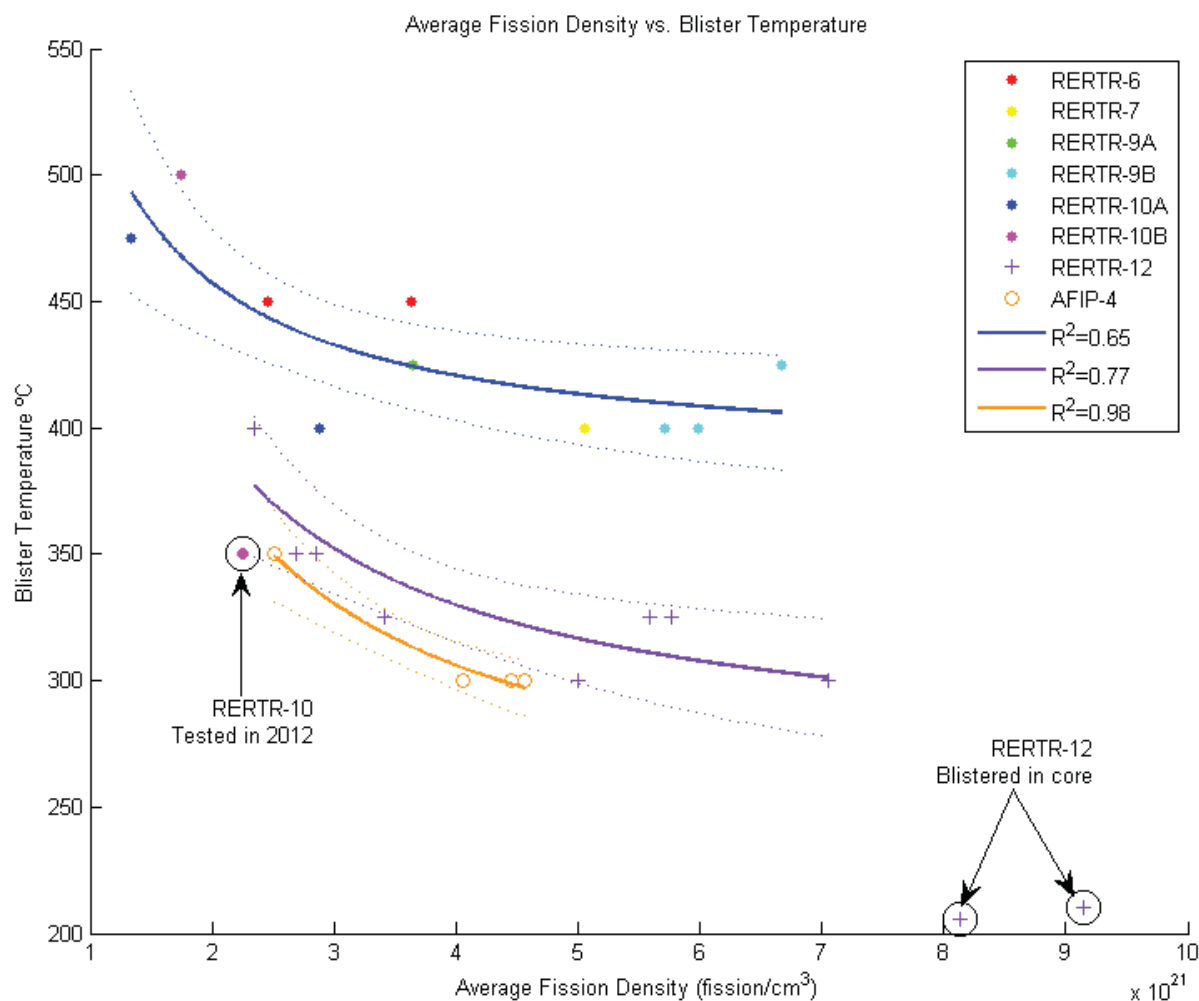


Figure 1. Curve fits of data from the two monolithic fuel blister testing campaigns. The data is fit using the average fuel plate fission density. Three circled data points (one from RERTR-10 and two from RERTR-12) were not included in the model fit.

Discussion

As seen in Figure 1, the decrease in blister temperature with fission density in each of the two groups of plates (prior to RERTR-12 vs. RERTR-12 plus AFIP-4) appears to follow a trend consistent with the model described earlier and is similar to other fuel systems,

decreasing with increasing fission density. Because of the iterative method historically established for collecting blister-threshold temperature data and the relatively small data sets, blister-threshold temperature data are inherently noisy. The data from the two blister test campaigns, nevertheless, appear to be statistically distinct (Figure 1) at the 95% confidence level.

All blisters examined were preferentially located either at, or connected to, the corners or the edges of the U-Mo fuel foil where both the stress gradients and fission density are maximized. There appear to be two types of blisters. On the RERTR-12 plates, Type 1 blisters are designated as small blisters, less than about 0.2 cm^2 in size, with some involvement of the clad-to-clad bond adjacent to the fuel-to-clad bond (see Figure 4). The upper limit of fission density for the formation of Type 1 blister is approximately $4 \times 10^{21} \text{ fiss/cm}^3$. It is apparent that at this low fission density, the fuel foil remains relatively strong because it has not yet become embrittled from the build-up fission products. In addition, the plates with Type 1 blisters manage to endure several thermal cycles in the course of the annealing tests. It has been hypothesized that during the cool-down phase of the test, due to significant differences between thermophysical properties of U-Mo fuel and aluminum cladding, the foil can initiate a debond at the clad-to-clad interface adjacent to the fuel foil. This characteristic was observed during examination of irradiated fuel via optical metallography. As is shown in Figure 5, the fuel in these cases remains mostly intact, but the clad-to-clad bond is opened, with the separation moving towards the outer edges of the plate.

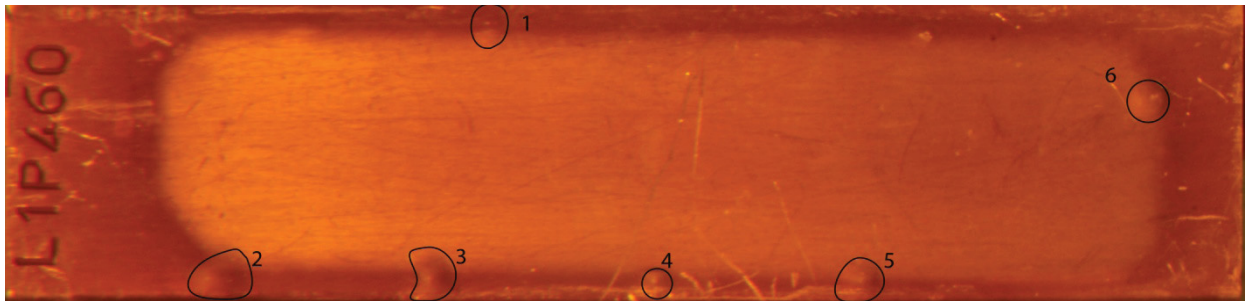


Figure 4. Type 1 blisters on the front side of plate L1P460 (RERTR-12). Average fission density $2.35 \times 10^{21} \text{ fiss/cm}^3$. Peak fission density $3.98 \times 10^{21} \text{ fiss/cm}^3$.

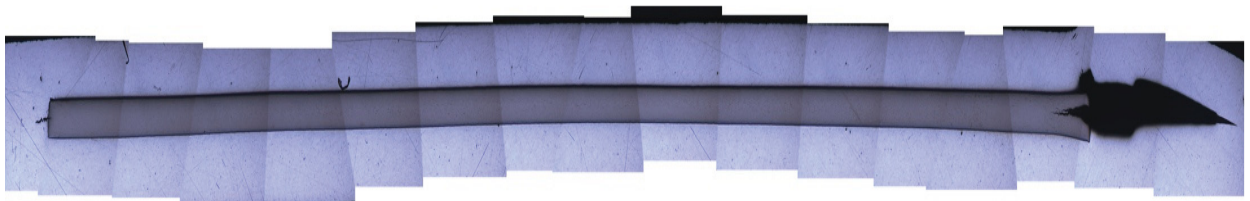


Figure 5. Montage of optical metallography images of the transverse cut through a Type 1 blister (#4) on plate L1P460 (RERTR-12). Average fission density $2.35 \times 10^{21} \text{ fiss/cm}^3$. Peak fission density $3.98 \times 10^{21} \text{ fiss/cm}^3$.

Type 1 blisters have not been observed on the plates with burnups exceeding $4.0 \times 10^{21} \text{ fiss/cm}^3$; instead, Type 2 blisters (see Figure 6) are present over the fuel zone and cover a

larger area. These blisters appear to grow larger with burnup, indicating that propagation may be fission gas driven. In fuel plates with fission densities exceeding $\sim 5.5 \times 10^{21}$ fiss/cm³, the Type 2 blisters become more “pillow-like” in appearance. At these high fission densities, the fuel foil is saturated with fission gases and other fission products and has become embrittled. A typical metallographic cross-section view of the Type 2 blister is shown in Figure 7, Figure 8 and Figure 9. As seen from these images, it appears that the tearing (cracking) in the Type 2 blisters usually occurs in the U-Mo foil, close to the U-Mo/Zr diffusion layer.

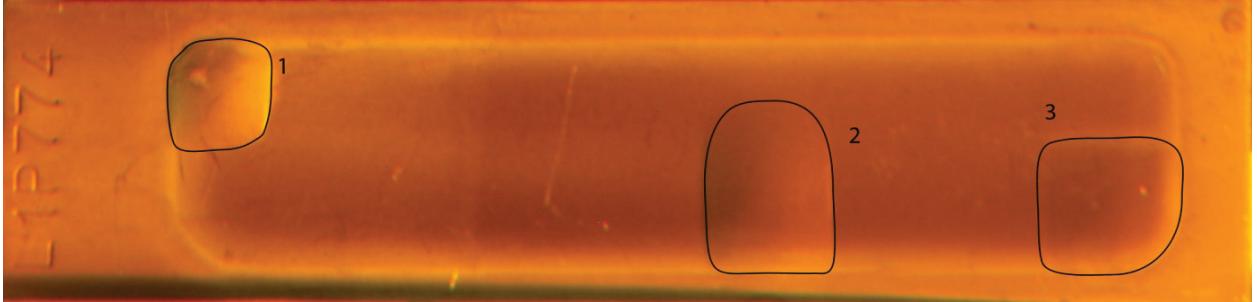


Figure 6. Type 2 blisters on the front side of plate L1P774 (RERTR-12). Average fission density 5.59×10^{21} fiss/cm³. Peak fission density 7.49×10^{21} fiss/cm³.



Figure 7. Montage of optical metallography images of the transverse cut through Type 2 blister on plate L1P758 (RERTR-12). Average fission density 5.00×10^{21} fiss/cm³. Peak fission density 9.20×10^{21} fiss/cc.



Figure 8. Optical metallography image (500 \times) of the right end of the transverse cut through Type 2 blister on plate L1P758 (RERTR-12) shown in Figure 6.



Figure 9. Optical metallography image (500 \times) of the axial cut through Type 2 blister on fuel plate L1P754 (RERTR-12).

It has yet to be determined whether these lower blister-threshold temperatures would be acceptable for the NRC-licensed research reactors and high-power reactors like the ATR and HFIR. Nevertheless, these low temperature values imply that some significant changes may have occurred between the RERTR tests performed prior to and those beginning with RERTR-12, changes which warrant further investigation. An investigation has been launched to identify differences between the tests and to determine the cause of the lower blister-threshold temperature observed in irradiated fuel specimens from the RERTR-12 and AFIP-4 experiments.

Several theories were identified to explain the systematic variation in observed blister threshold using a standard Failure Modes and Effects Analysis (FMEA) methodology. This analysis identified hypotheses related to the test methodology, irradiation environment, and fabrication processing history that may have lead to variation in performance. Each of these hypotheses and being further studied to determine the root cause phenomena that may have triggered the change. The hypotheses are summarized as;

- 1) The high temperature and low temperature data were each collected in distinct and separate time frames. If the blister furnace system was reconfigured in a manner that changed the testing environment between these measurements, inaccurate data may have resulted from one configuration or the other. A systematic study to re-characterize the test apparatus is underway.
- 2) The early, high temperature data was all collected from fuel plates that were irradiated in the 'edge-on' configuration such that a significant burnup gradient was present across the width of the plates. The more recent, low temperature data was collected from fuel plates irradiated in the 'face-on' configuration such that the burnup profiles were more uniform. A series of thermal-mechanical analyses are being run to project the presence of any significant residual stress patterns or fuel performance parameters that might drive the observed behavior. An irradiation test is being developed to support evaluation of these orientation phenomena.
- 3) Fuel plates irradiated in both data sets experienced differences in fabrication process history. The fuel plates irradiated in earlier experiments and that exhibited higher blister thresholds contained fuel foils that were typically hot rolled to thickness or annealed after rolling. Alternately, all the plates irradiated in RERTR-12 and AFIP-4, which exhibited low blister thresholds, contained fuel foils that were cold-rolled to final thickness. It has been hypothesized that the cold rolled foils contain microstructural features or residual stress that ultimately impacts the blister performance of the fuel plates. An effort to enhance understanding of as-fabricated-process dependent microstructure and post-irradiation microstructure is underway that is intended to identify any critical features. A series of out-of-pile and irradiation tests are being planned to collect the necessary data to describe and to confirm the anticipated role of microstructural features on blister behavior.

Summary and Further Work

Blistering of irradiated fuel plates at elevated temperatures has long been identified as the point of first release of fission products from research and test reactor fuel plates. In order to establish the safety limits for this fuel type, the blister threshold test was developed. This test was previously used to inexpensively establish a conservative threshold for current dispersion fuel designs (based on UAl_x , U_3O_8 , and U_3Si_2) that was commonly used in reactor safety analysis.

The standard test methodology was applied to the U-Mo alloy based monolithic fuel design currently under development by the RERTR program. Test results have shown a systematic difference in performance between fuel plates irradiated in different reactor experiments. The root cause of this difference is currently under investigation and has resulted in several hypotheses related to the irradiation environment, blister test apparatus, and the as-fabricated microstructure.

The extent to which the out-of-pile blister threshold test simulates in-pile behavior is not clear. Additional study will be performed to evaluate the inherent safety margins built into the test methodology and to determine if an alternate test would be more appropriate in the future.

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