Mechanistic Fuel Failure Evaluation System (MFFES)

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Abstract. Failure thresholds and fission product releases will be used to define the allowable safety margins for U-Mo monolithic and dispersion fuel plates. These margins can be improved through high fidelity testing. Failure points will be defined at clad blistering, clad failure, then diffusion barrier (as applicable) failure, and finally at fuel failure. In order to characterize these failure points, mechanistic behaviors, and the fission product release, a robust state of the art, multipurpose furnace and fission product analysis system must be developed.

1. INTRODUCTION

Previous studies performed on metal and oxide fuels demonstrate sound methodologies that can be applied to characterize these emerging fuel types. Literature from 1966 [1] and 1970 [2] discuss gas retention properties studied under meltdown conditions for UAlx and U_3O_8 dispersion plates. More recent literature, from 1993 [3] and 1994 [4], reports both fission gas behaviors in metallic fuels in addition to fuel/cladding chemical and mechanical interactions. An additional report from 1994 [5], discusses the deposition of fission product volatiles on the reactor coolant system. Work to characterize these behaviors in fuels developed in the last fifteen years has not been conducted. Therefore it is important that a system be developed to define the mechanistic behaviors of the monolithic fuels, new dispersion fuels, and transmutation fuels.

1.1. Hypothesis

Mechanistic fission product behavior of monolithic, dispersion, and transmutation fuels can be evaluated as released fission products are measured in solid and gaseous states, if a stepped approached to fuel failure is followed. Once these techniques are developed for the specific fuel type, a more comprehensive understanding of in-pile fission product mechanistic behavior can be formulated as they affect the material failure thresholds. As these failure parameters are determined and empirical data is gathered, the safety basis for new fuel types can then be established. Further development of the system and experimental methods can be initiated so that the furnace can mimic reactor over power thermal conditions to form eutectic and chemical interactions between clad and fuel. Fuel can be sectioned and examined to characterize these layers and interactions to define how they affect fuel performance.

2. SYSTEM DEVELOPMENT

A furnace will be placed in HFEF main hot cell at MFC. Irradiated fuel and cladding specimens will be put in the furnace for thermal degradation of materials so that fission products will be liberated at

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established failure points. As the temperature of the furnace is increased step wise or ramped, to follow known failure points and the fission products, both gases and volatized solids, will be measured using gamma spectroscopy (online), gas mass spectroscopy (grab sample) and chemical dissolution examinations (sectioned samples). Attached to the furnace port, in series, will be a thermal gradient tube, a HEPA filter, a gas expansion segment and a port for the gas grab sample. The gases collected in bottle will then be analyzed by gas mass spectroscopy.

The HEPA filter gradient tube sections can be further analyzed using a gamma detection system located in the AL hot cells. Additionally, the HEPA filter can then be dissolved in the hot cells and dilutions made for transfer out of the hot cells for further analysis of other solid fission products as well as solid activation products. A combination of chemical separations and analytical instrumentation for quantitative and qualitative measurements will be used. Some of these techniques will include anion exchange separations (both gravimetric and automized separation methods), inductively coupled plasma mass spectrometry and atomic emission spectroscopy (ICP-MS, and ICP-AES).

Analysis approach will be broken up into four stages and applied to each irradiated plate. The first will be to initiate blisters on the plate and measure any fission gas release. The second will be to ramp up the temperature at a rate yet to be determined, to breach and then completely melt the plate clad. Thirdly, the temperature of the furnace will continue to increase until the entire sample is melted and fission gases and volatilized solids measured by online gamma detectors. Stage four is comprised of three grab samples to be further analyzed for non-gamma emitters in the thermal gradient tube and the melted sample, and gases using gas mass spectroscopy.

3. BLISTERING

Blistering thresholds are the standard by which the safety margin for reactor operation is established. The assumption is that once blistered, the plates are presumed to be at the point of failure and total release of fission product is imminent. What has not been determined is the following:

Are blisters really indicative of imminent plate failure?

1. Are the blisters created by fission gases?
2. Is there is fission gas release at the point of blistering?
3. If not, how long at blister temperature before a release might occur?
   a) Can a blistered plate safely remain in the reactor for any period of time before a breach can be expected?
   b) How is the clad corrosion rate at the blister point increased?
4. If clad still remains intact following a hold period at blister temperature, at what temperature do the fission gases begin to escape?
5. If fission gases do not initiate blisters what does?

Several studies were reviewed to evaluate some of what is known about clad blistering behaviors. Development work for ATR fuel [6] observed that while gases do not contribute to creep or yielding behaviors but do fill in those voids that remained after fabrication and those created during the U_3O_8-Al reaction. Fission gases are credited (by calculation) with volumetric void expansion increase of 2.8 percent which is almost that of the 3 volume percent voids increase measured during irradiation. If there is an interconnection mechanism of these voids the volume is claimed to be sufficient to cause blistering.

An Olander authored [8] article suggests otherwise. He argues that the pressure of the fission gases contained within the measured volume is not sufficient to deform the cladding and that mechanical failure of the clad can be attributed to the tensile stress normal to the plate. A journal paper [9] on non-fueled aluminum was also explored for any non fission gas contributions to blistering behaviors that

might be better understood. While this article did not point to anything specific within our current systems, the blistering documented, was a function of the presence of hydrogen introduced during the fabrication processes. As most of RERTR fuels are clad with aluminum in addition to being in the matrix material for dispersions, it is important to be cognizant of the fact that gases, to some small degree are already present with the clad and matrix materials.

4. PROJECTED DATA

Expected fission product inventories following a forty day cooling in the ATR canal are:

- Plate blistering temperatures for given fuel type, clad type and clad bonding method.
  - Initial blistering temperature.
  - Video capture of blister propagation.
- Detection of fission gas release via HpGe detector during clad blistering phase of plate thermal degradation.
  - Xe, Kr and Br.
- Fission gas release as a function of temperature, clad degradation and fuel degradation.
  - Xe, Kr and Br.
- Total fission gas release collected as a grab sample (gas mass spectroscopy analysis) following completed fuel failure.
  - Xe, Kr and Br.
- Temperature dependent volatilized solids deposition behavior.
  - Real time measurement using CZT gamma detectors of volatilized solids.
    - I, Cs, Sb, Ag, Te, Mo, Ba, Rh, Eu, Ru, Ce, Pr, La Nb and Zr.
- Type and quantity of deposited volatiles.
  - Mass balance for determination of weight of solids deposition.
  - Chemical dissolutions of thermal gradient tube sections for determination of presence of non-gamma emitters.
- Waste plate material will be sampled and chemically analyzed for any retained fission gases and isotopic elements (some may not be present due to decay).
  - H, Be, C, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Kr, Rb, Sr, Y, Zr, Nb, Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Te, I, Xe, Cs, Ba, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er and Tm.
- Re-irradiated future samples for reactivation of decayed fission products for a more comprehensive analysis of inventories while in reactor.

5. MECHANISTIC FUEL FAILURE EXAMINATION SYSTEM COMPONENT SPECIFICATIONS

The furnace components are front access (see Fig. 5.1). This furnace is engineered and fabricated for high temperature operation in a controlled atmosphere, the hot zone, in the shape of a cylinder, is resistance heated and is insulated from the cold wall by fibrous graphite shields. Access to the furnace for loading and unloading is manual via remote operated manipulators with sample melt contained within a retort as shown in Fig. 5.2. Door clamps are provided for operation to slightly positive pressure. The entire inside of the vacuum chamber is designed to conform to the best high vacuum practices with particular attention given to surface finishes and choice of materials. Visual observation of the specimen is provided for by a sight port with a rotatable viewing glass. Ports are also provided for inert gas inlet, thermocouples and pertinent instrumentation.

FIG. 5.1. Main furnace and thermal gradient furnace at the bottom and top respectively.

FIG. 5.2 Retort (sample holder) mounted inside main furnace.

All controls and instrumentation are positioned to optimize monitoring and control. Furnace temperature is controlled by closed-loop circuit between the temperature sensor, the control instrument, and the power supply. System protection is provided by water-flow interlock. Components are readily accessible for inspection and maintenance.

5.1. Thermal gradient tube and furnace

Movement of volatilized fission product solids will be facilitated along the length of the Inconel or stainless steel gradient tube using the thermal gradient furnace shown on top of main furnace in Fig. 5.1 and in Fig. 5.3 with 6 discreet temperature zones.

FIG. 5.3 Thermal gradient furnace with deposition tube in place.

5.2. Camera system

In order to capture blister formation with removing plate from the furnace, the system will be equipped with 2 cameras and 4 lights located at view ports with quartz windows. The configuration can be seen in Fig. 5.4.

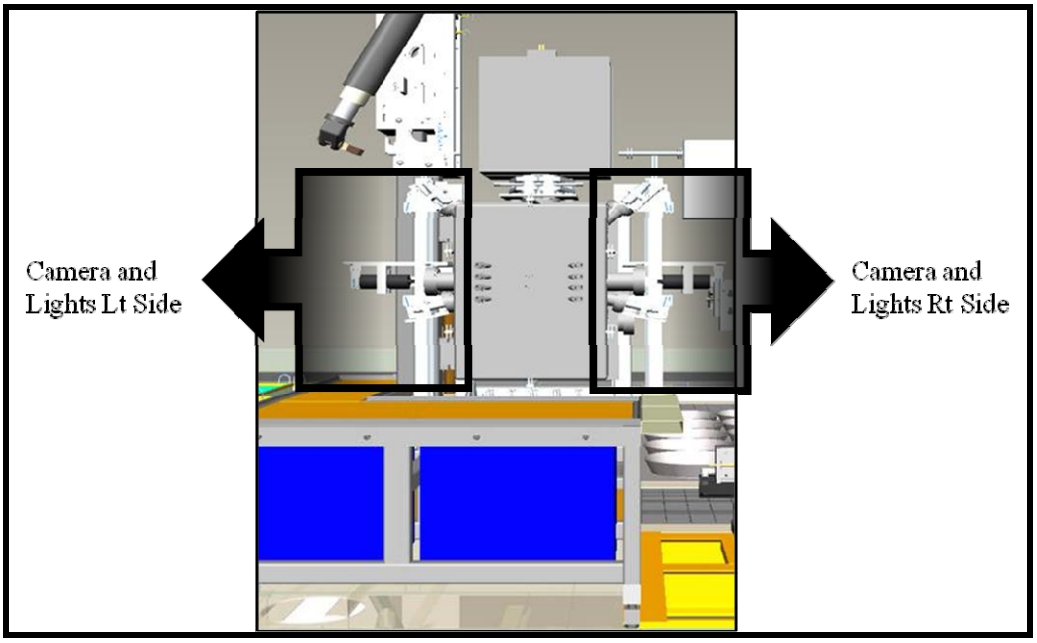

FIG. 5.4. Camera and light configuration.

5.3. Fission gas loop

The fission gas line will be placed in series with the thermal gradient tube. Gas line will pass through hot cell feed to the detection system located at the expansion segment of the system. Gases will loop back into the furnace for a measure of total fission gas accumulation attributed to the fuel plate failure during the thermal degradation regime. A grab sample of the accumulated fission gases will be collected at a port outside the hot cell following completion of test.

5.4. Fission product detection system

In cell detectors will be cadmium zinc telluride spear gamma detectors provided by eV products. The SPEARS will be shielded and non-stationary to facilitate optimal resolution. The out of cell for system will be the fission gas detection component will be an ORTEC HpGe detector.

5.5 In cell SPEAR gamma detector system specifications

CdZnTe detector dimensions (example shown in Fig. 5.5):

1. 5 × 5 × 5 mm with CAPture® contacts
2. Measurement capabilities:
3. Energy range: 1–10 MeV
4. Energy resolution: < 4% FWHM @ 122 keV @ 25ºC
5. Operating temperature:
6. Range: -10–50ºC standard

FIG. 5.5 Example SPEAR detectors.

SPEAR™ dimensions:

1. 13 mm diameter × 89 mm length (without cable) .
2. Total length of SHV HV output cable is 3’.
3. Total length of lemo connector cable is 6’.
4. Total length of BNC output cable is 3’.

SPEAR detector input requirements:

1. 9 PIN sub D connector: +12 VDC, ground.
2. SHV connector: HV bias (typically +500 to +1000 VDC).

Signal output:

1. BNC connector and preamplifier output.
2. As assembled, all input/output connectors will interface directly with any standard spectroscopy or counting system.

SPEAR performance:

1. + 7 to 12 VDC operation.
2. Input capacitance: 6.6pF.
3. Risetime @ C source: 1pF- 35nS.
4. Falltime @ C source: 1 pF - 725 μS.
5. Noise @ C source: 0 pF - ~160 e- (Si).
6. Sensitivity: 3.2 mV/fCoul.
7. Power dissipation: 130 mW (+ 12 VDC supply).

ORTEC GEM series coaxial HpGe detector specifications:

1. Energy resolution at 1.33 MeV photons from 60Co at optimum shaping time.
2. Relative photopeak efficiency for a 60Co 1.33 MeV peak.
3. Peak-to-Compton ratio for a 60Co 1.33 MeV peak.
4. Peak shape ratio for the full-width tenth-maximum to the full-width half-maximum for a 60Co 1.33 MeV peak.
5. Energy resolution at 122 keV photons from 57Co at optimum shaping time.

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