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Progress Report on the Measurement of Fission Gas Release from Irradiated Fuel during Temperature Transients

Nuclear Technology Research and Development

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SUMMARY

A device for real-time fuel heating observations and collection of fission gas release data is being developed to study nuclear fuel behavior during a temperature transient. The proposed system incorporates high heating and cooling rates, a digital violet laser microscope, mass spectrometry, and gamma ray spectrometry. By placing the system in a hot cell or in a hot cell–like enclosure, the capability could support study of a range of samples, including low and high burnup nuclear fuels. This report documents a conceptional design that incorporates a commercially available system that can be modified to achieve the desired specifications and proposed modifications.

This report has been submitted in fulfillment of milestone M3FT-19OR020204082 titled, "Progress Report on Measurement of Fission Gas Release from Irradiated Fuel during Temperature Transient," for the US Department of Energy Office of Nuclear Energy, Advanced Fuel Campaign of the Fuel Cycle R&D program.

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ACRONYMS

AFC	Advanced Fuels Campaign
HBS	high burn-up structure
LOCA	loss-of-coolant accident
MOX	mixed oxide
NEAMS	Nuclear Energy Advanced Modeling and Simulation
ORNL	Oak Ridge National Laboratory
UC	uranium carbide
UN	uranium nitride

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PROGRESS REPORT ON THE MEASUREMENT OF FISSION GAS RELEASE FROM IRRADIATED FUEL DURING TEMPERATURE TRANSIENTS

1. Introduction

The behavior of pores and bubbles in UO_2 remains a research area with several gaps in knowledge, yet the migration of porosity and bubbles has important impacts on nuclear fuel performance of UO_2 in light-water reactors. The mobility of these features impacts steady-state fission gas release and transient accident conditions [1–3]. Pore migration also has an important impact on the fuel performance of UO_2 and mixed oxide (MOX) fuel for fast reactors [4–6]. This behavior becomes particularly important when considering UO_2 that has developed high burnup structure (HBS) during irradiation [7]. If this structure is subjected to a loss-of-coolant accident (LOCA) in operation or LOCA testing, then the highly pressurized Xe bubbles in the HBS will pulverize the fuel [8], and the fuel will be dispersed readily through any cladding breach that also typically occurs in a LOCA scenario. Heating tests have been performed on fuel to observe fission product release and fuel fragments left over after an out-of-pile heating transient. However, direct observation of pore and bubble migration in UO_2 during a heating transient with real-time measurement of the fission gas release has not been demonstrated previously. Direct observation of bubble and pore migration in UO_2 will compliment other transient techniques and will aid in the development of a more mechanistic model of HBS and its behavior during normal and accident conditions in UO_2 fuel. The technique developed here can also be applied to (1) pore migration that is important for UO_2 and MOX fuel performance and (2) to novel fuel forms beyond oxides such as uranium nitride (UN), uranium carbide (UC), and metallic U alloys. The developed mechanistic models can then be implemented in Nuclear Energy Advanced Modeling and Simulation (NEAMS) tools such as MARMOT to enhance fuel performance simulations.

Exploration of nontraditional fuel geometries such as those proposed for investigation by the Advanced Fuels Campaign (e.g. MiniFuel and Fission-Accelerated Steady-state Testing, FAST) will also introduce numerous uncertainties regarding their ability to accurately capture mechanisms that affect fuel performance. It will be necessary to understand how phenomenon such as fission gas bubble behavior translate to observed performance in nontraditional fuel geometries. Improved experimental data will facilitate improved mechanistic models, which can in turn allow for extrapolation to performance of fuel pellets sized according to actual reactor application.

To better understand all these important phenomena, a preliminary design has been performed to establish a nuclear fuel transient testing system at Oak Ridge National Laboratory (ORNL). This system will mirror the system discussed in Turnbull et al. [8] and Fontana et al. [9], but it is proposed that a visualization technique also be included to increase its utility. The pathway to establishing this system has been launched. This report documents a conceptional design that incorporates a commercially available system that can be modified to achieve the desired specifications and proposed modifications. Corresponding challenges have also been identified.

2. Conceptional Design of Nuclear Fuel Transient Testing Station

To simulate the transient conditions occurring in nuclear reactors, a high-temperature heating stage coupled with necessary characterization tools is required to assess nuclear fuel behavior in terms of fission gas release and fuel fragmentation. Figure 1 shows a schematic plot of a conceptual nuclear fuel transient testing station capable of in-situ observation of the fuel's structural evolution and fission gas release measurement during a rapid temperature ramping process. Irradiated nuclear fuels are expected to be heated in a clean sealed chamber located inside either hot cell or an enclosure. The use of infrared heating can give rise to

the desired rapid temperature ramping rate (>5°C/s) simulating the transient conditions; the small size of the specimen reduces the time required for the heat pulse to reach the central regions of the specimen. This is a reasonable approximation to the volumetric heating that would be seen under an actual LOCA. A visualization instrument—a specialized short wavelength microscope—will be placed above the heated fuel to record the fuel's behavior during the transient testing. The released fission gases will be identified and quantified using gamma ray spectrometry for the radioactive isotopes (high sensitivity), and mass spectrometer for both, providing lower sensitivity for the radioactive isotopes, along with the ability to detect the stable isotopes. Given the extremely high radiation dose of irradiated nuclear fuel, small samples are required for testing. Because the use of small samples results in the generation of weak signals, a low background is desired and could be achieved in an ultra-high vacuum environment by employing turbo pumps and high vacuum techniques. However, this may substantially increase the internal volume and footprint of the system. Since high-pressure testing may be desired to better simulate the actual conditions of a LOCA, the tradeoffs between a vacuum vs. a positive pressure system must be better understood before proceeding with final design.

To locate the system in a hot cell or a shielded glovebox, an easy sample loading procedure is required. Following transient testing, the nuclear fuel may require additional detailed microstructural characterization using a scanning electron microscope or transmission electron microscopy to characterize the redistribution of fission products, grain growth, fission gas bubble distribution, etc. This would require additional specimen preparation, along with the use of other analysis tools. The desired specifications of the nuclear fuel transient testing station are summarized in Table 1.

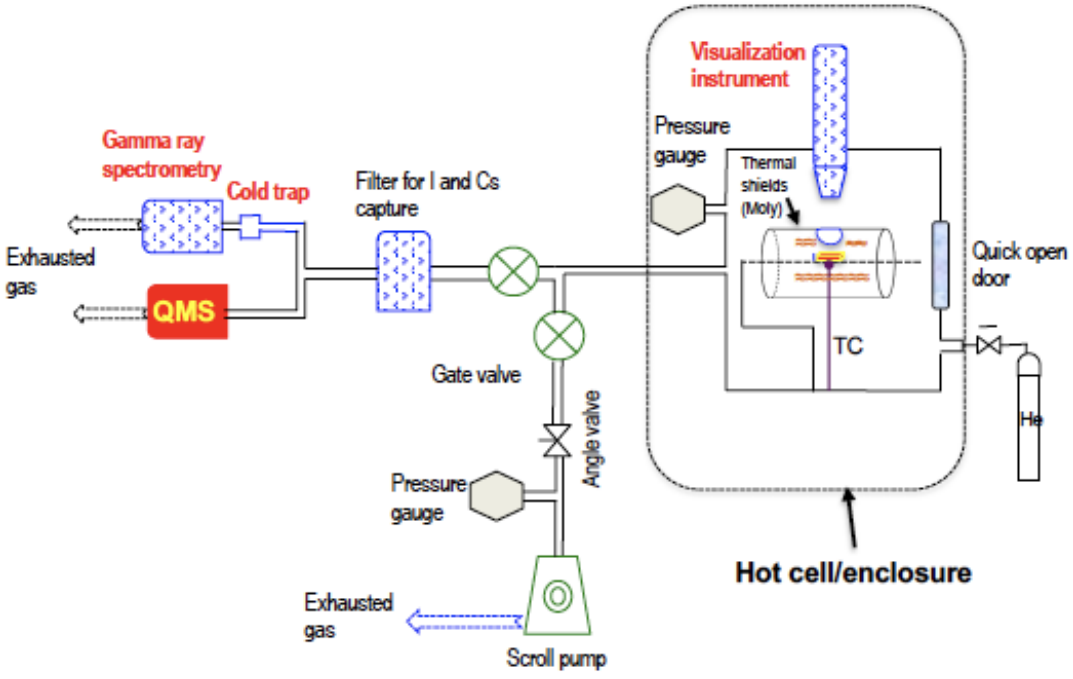

Figure 1. Schematic Plot of Nuclear Fuel Transient Testing Station.

Table 1. Specifications of the desired nuclear fuel transient testing station

Parameters	Desired Values
Maximum temperature	>1500°C
Temperature ramping rate	>5°C/s
System pressure	TBD, may be dual – vacuum or positive pressure
Visualization	< 1μm resolution
	High-temperature compatibility
Mass spectrometer	<1appm resolution
γ ray spectrometry	<100eV energy resolution, < 1 μCi
Sample loading	Easy access
Maintenance	Easy to disassemble and clean under semi-remote conditions

3. Pathways to Establish the Capability of Nuclear Fuel Transient Testing

The current design strategy is to build the transient testing system from commercially available components as much as possible, especially those with short lead times. It is possible to take advantage of commercially available ultra-high–temperature microscopes and make corresponding modifications to satisfy the system requirements for the nuclear fuel transient testing station in either a hot cell or a shielded glovebox. An ultra-high–temperature infrared heating observation violet laser microscope fabricated by Yonekura MFG Co LTD in Japan has been identified based on data gathered in an intensive market survey. This product claims the only real-time high-temperature in-situ observable laser microscope in the world.

Model VL2000DX-SVF18SP (shown in Fig. 2) is an ultra-high–temperature infrared heating real-time digital violet laser microscope. The elliptical sphere reflection focusing sample chamber could support the use of inert gas, ordinary atmosphere, vacuum, or reduction atmospheres. The attainable ultimate pressure is 10^{-4} torr. The equipped gas scroll flow system prevents adhesion of sublimed substance to the observation window, providing a clear observation path at all times. The maximum temperature of the embedded infrared image heating system is 1800°C using a 1.5kW halogen lamp with a cylindrical heating area of 10 mm in diameter and 10 mm in height. Ultra-high–speed temperature rise and fall can be achieved. For example, the maximum heating rate is 60°C/s, and the highest cooling rate is 100°C/s by rapid cooling with high pressure helium gas, but the gas analysis system may not be able to process this high flow rate.

This model has a high resolution, high contrast VL2000DX digital laser camera head using a 408 nm semiconductor laser as the light source. It is expected that a 0.45 μm resolution could be obtained by using the ultra-long working distance lens (TPS35X) for high temperature observation. The VL2000DX can respond with a maximum of 60 frames/s fast scan for a violet laser wavelength of 405 nm, offering a good chance of capturing rapid structural changes in the fuel test specimen. In addition, a standard objective lens is also available for room temperature 3D imaging prior to beginning the test to establish a baseline for the time analysis.

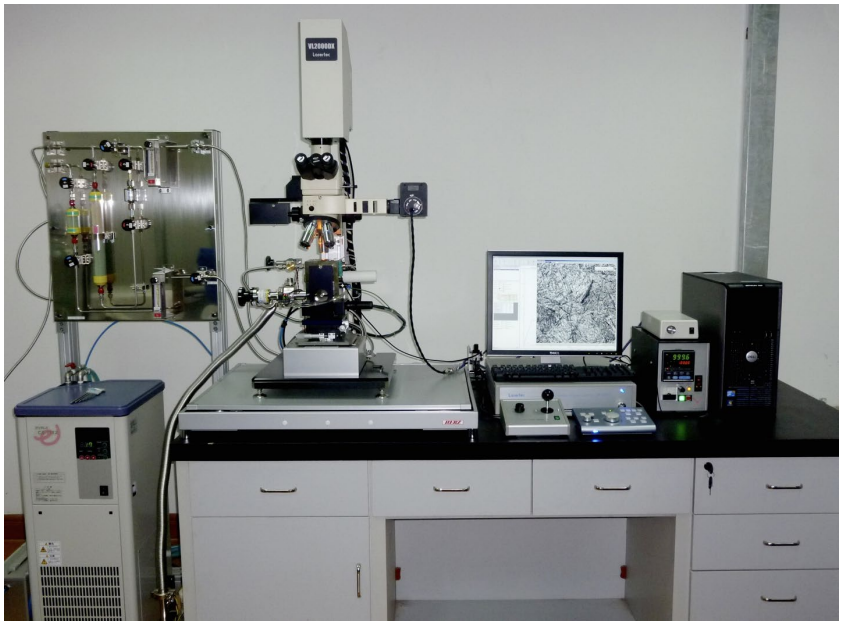

Figure 2. Yonekura VL2000DX-SVF18SP Ultra-High Temperature Infrared Heating Real-Time Digital Violet Laser Microscope (Reproduced with permission from YONEKURA MFG Co LTD)

The VL2000DX-SVF18SP provides a high-temperature capability with excellent ramping and cooling rates, both of which are critical for the fuel transient testing. This equipment will serve as the nucleus of the desired nuclear fuel transient testing system. Challenges associated with applying the system for nuclear fuel testing are identified below.

3.1 Location

The main section of the VL2000DX-SVF18SP system will be placed in a hot cell or a verified hot cell–like enclosure to provide the necessary shielding. Figure 3 shows the sample loading process of this system: removing the 4 screws on the furnace lid, opening the lid, and loading the sample. The sample loading process is not straightforward, making the sample exchange challenging if the system is located in the hot cell. However, the process is simple enough that it is possible to load the sample into the hot cell using fixtures and vacuum tweezers. Considering the cost, operation, and maintenance of this system, including the vacuum unit, a custom-designed or modified glovebox-like enclosure is desired to host the system at ORNL Building 3525. This will enable easy and economical access to the system for sample exchange and maintenance.

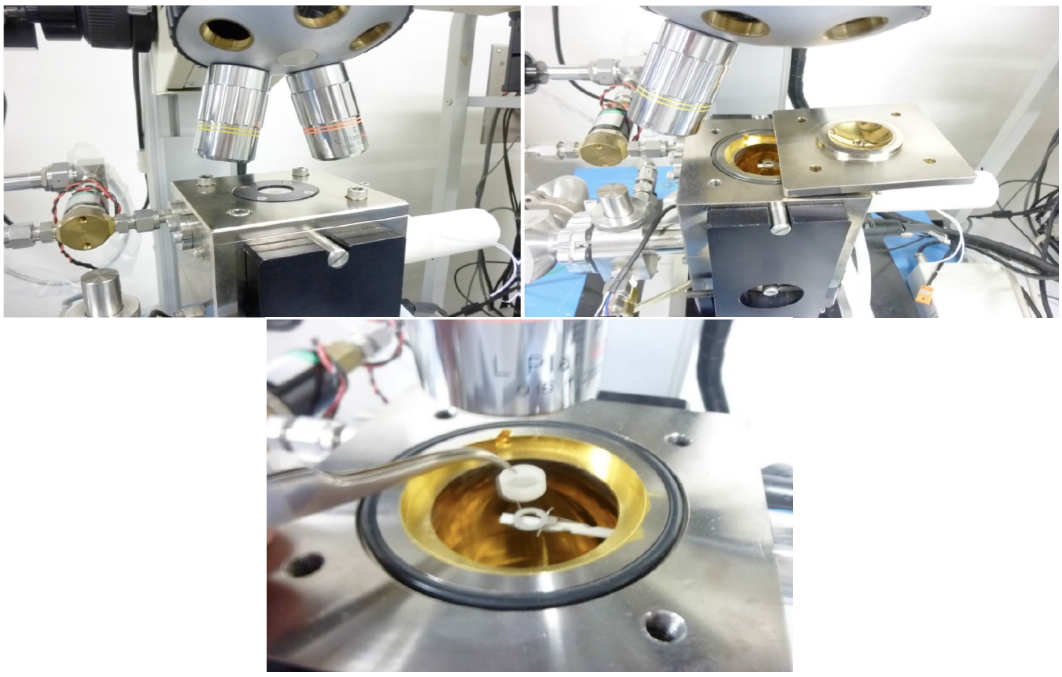

Figure 3. Sample Loading Procedure of VL2000DX-SVF18SP
(Reproduced with permission from YONEKURA MFG Co LTD)

3.2 Fission Gas Characterization

It is critically important to identify and quantify the fission gas release during the fuel transient testing. As mentioned in Section 2, gamma ray spectrometry and mass spectrometry will be part of the system. An inert gas scroll flow system is included with the VL2000DX-SVF18SP to prevent the adhesion of sublimed substances to the observation window and possible sample oxidation at high temperature. The flowing ultra-high–purity helium gas will sweep the fission gas out of the sample chamber for future characterization. After leaving the sample chamber, the gas will pass through a filter for capture of iodine and cesium and to eliminate dust, and then the gas will be split into two lines. Considering the possible particulates, additional filters may need to be included in the gas processing lines. One line includes cold traps and gamma ray spectrometry (^{85}Kr), and the other line has a mass spectrometer that will be used to accurately determine the amount of fission gases (Xe and Kr) with high resolution (~1ppm). Therefore, working with small amounts of nuclear fuels (<0.1 g) will be possible. Determining the relative ratios of the Xe and Kr isotopes that make up most of the fission gases observed in the irradiated fuel is particularly important, particularly when combined with the absolute determination of ^{85}Kr by gamma ray spectrometry of the fission gases in the cold trap. The combination of information enables all Xe and Kr isotopes to be measured quantitatively.

Significant efforts are needed to incorporate the two characterization tools into the gas flow system of VL2000DX-SVF18SP. Quantification of mass spectrometer readings will be challenging, and a separate gas calibration system will be required. The ability to create time resolved fission gas release data may be limited by the time necessary to collect a signal and is highly dependent on the method used to collect fission gas for analysis. Initially, a cold trap coupled with a HPGe detector is envisioned for this system, but a faster response time could be beneficial to enhance the capabilities of this system. The overall design philosophy is to remain flexible so that different detectors with different response times and sensitivities can be deployed to fully study all the associated phenomena.

3.3 High-Temperature Imaging

The crucibles provided by the vendor are 6.5 mm and 9.0 mm diameter alumina open crucibles. It has been observed that high-temperature annealing of high burnup fuel could result in fragmentation and sputtering. Therefore, it might be necessary to have a Knudsen crucible to keep the solid fuel pieces within the crucible. The opening of the Knudsen crucible must be determined based on the laser beam size. How the use of the Knudsen crucible will impact the imaging must be evaluated. The other option would be to make the chamber internals easily replaceable and to simply replace the internals after each use. This may require extensive redesign, however.

3.4 Testing Under Pressure

In out-of-pile LOCA testing, the fragmentation behavior of HBS UO_2 has been observed to change depending on the internal pressure of the cladding prior to the test. The limited data available suggest that LOCA tests are more representative if the cladding is pressurized to approximately 4 MPa (580 psi). Coupling of visualization and high-pressure may not be possible, but it may be possible to pressurize samples for heating transients without visualization while still observing fission gas release. This would allow for comparisons between lower pressure visualizations and high-pressure tests.

4. Summary

This report presents the pathway to establish a nuclear fuel transient testing station at ORNL. Based on the conceptional design, a commercially available ultra-high–temperature infrared heating observation violet laser microscope is identified as the basic component for the desired nuclear fuel transient testing system. Challenges are reviewed regarding its location, fission gas release characterization, and high temperature imaging.

In the next FY, the objective is to procure the identified high-temperature imaging system for further modification after the current work plan is fully approved.

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