RERTR 2019 – 40TH INTERNATIONAL MEETING ON
REDUCED ENRICHMENT FOR RESEARCH AND TEST REACTORS

OCTOBER 6-9, 2019
THE WESTIN ZAGREB
ZAGREB, CROATIA

Fission Product Release Testing to Support Qualification of an LEU Fuel for the Advanced Test Reactor

K. Metzroth, PhD
MPR Associates
320 King Street, Alexandria, VA 22314 – USA

S. Kwon, PhD, J. Henley, J. Cole, PhD
Idaho National Laboratory
1955 N. Freemont Ave., Idaho Falls, ID 83402 - USA

ABSTRACT

To support the qualification of a low-enriched uranium (LEU) fuel that can meet the mission of Advanced Test Reactor (ATR), fission product release rate data on the LEU fuel (U-10Mo) must be obtained to update the ATR maximum hypothetical accident (MHA) evaluation. The MHA in the ATR safety analysis report (SAR) is currently supported by a detailed severe accident analysis (SAA) that was used to estimate the offsite and onsite consequences of the MHA. In order to support the ATR LEU SAR, this accident analysis must be updated to incorporate models that can predict the degradation and fission product release behavior of the new LEU fuel (U-10Mo) so that offsite and onsite source terms can be estimated. At present, limited data are available on the behavior of U-10Mo under degraded conditions, particularly on fission product release rates, which is a necessary piece of information to perform a comparable severe accident analysis to what was performed for the existing fuel system. This paper discusses the framework that will be used to evaluate the MHA for ATR LEU fuel and summarizes an evaluation of options that was performed to determine the best approach for obtaining fission product release data for U-10Mo fuel.

1 Introduction

The mission of the Department of Energy (DOE) National Nuclear Security Administration Office of Material Management and Minimization's (M^3) is to convert, remove, and dispose of vulnerable nuclear material located at civilian sites worldwide. As part of its mission, M^3's Office of Convert works around the world to convert research reactors and isotope production facilities to non-weapon-usable nuclear material, both domestically and abroad. The Office of Convert is working with the Idaho National Laboratory (INL) to develop and qualify new fuels and technologies to support conversion efforts domestically and abroad. The INL also is working on converting the Advanced Test Reactor (ATR) and the Advanced Test Reactor Critical

(ATRC) Facility from using High Enriched Uranium (HEU) fuel to Low Enriched Uranium (LEU) fuel.

The conversion of the ATR to LEU fuel will be require a Severe Accident Analysis (SAA) to evaluate the ATR Maximum Hypothetical Accident (MHA). The SAA will determine offsite dose for a range of hypothetical accident scenarios for comparison to siting criteria. Accurately quantifying offsite dose relies upon detailed analysis of severe accident progression events, including the degradation of the core and fission product transport. A key input to this analysis is the release rates of fission products from degraded fuel, which have historically been determined experimentally.

This paper describes the following: 1) the motivation and needs for obtaining fission product release data for ATR LEU conversion, 2) a summary of an evaluation of options to obtain U-10Mo fission product release data, and 3) the high level plan to obtain test data that will support the insertion of high power lead test elements.

2 Needs for Fission Product Release Data to Support ATR LEU Conversion

The ATR MHA is used to demonstrate that DOE siting criteria can be met. Specifically, both onsite worker dose and potential offsite dose are evaluated based on the defined MHA. The current ATR MHA evaluation consists of a detailed severe accident analysis that evaluates the melt progression, fission product release from the fuel, fission product transport through the primary system, and offsite release behavior. This severe accident analysis utilized the SCDAP/RELAP [1] computer code, which was capable of modeling the plant thermal-hydraulics during an MHA scenario including the relevant severe accident phenomena noted above.

Key input for the SCDAP/RELAP model are correlations that provide estimates of the fission product release rate from the fuel as a function of temperature. There were several experimental programs in the 1950s – 1970s where fission product release data from uranium-aluminide fuel were taken to support development of these models. The U-10Mo fuel system is a metallic alloy uranium fuel system as compared to an aluminum dispersion fuel. Limited data are currently available in the literature on the fission product release behavior of the U-10Mo fuel system under degraded conditions. Without more comprehensive information, it is challenging to estimate the quantity, timing, and chemistry of fission product release to perform dose evaluations for a severe accident.

A model that has been historically used in severe accident codes to predict fission product release is known as the CORSOR model [2]. The CORSOR model relates the total amount of fission product release over a given time interval as a function of time-at-temperature.

In the CORSOR model, the fraction of initial radionuclide inventory in fuel that is released over the course of an accident can be calculated using (Section 5.3.3 of [3]):

$$F = 1 - e^{-\dot{f}t}$$

Where:

F = release fraction [unitless],
$\dot{f}$ = release rate [fraction / min], and
t = time [min].

Within the CORSOR model, there are multiple sub-models available for computing the release rate, $\dot{f}$ for a given radioactive species (e.g., noble gases, cesium, iodine). The use of a specific model depends on the application; more information can be found in [2]. An often used release rate model is the CORSOR-M model, which defines the fission product release rates as ([2]):

$$\dot{f} = Ae^{-Q/RT}$$

Where:

$\dot{f}$	=	release rate [fraction / min],
A	=	empirical coefficient [fraction / min],
Q	=	activation energy [kJ/mole],
T	=	temperature [K],
R	=	ideal gas constant [J/mole-K)].

Once fission product release rate data are acquired, they are fit to the above release rate equations for each fission product of interest to determine fission-product specific values of Q and A.

The release rate models for each radionuclide of interest are coupled with a thermal hydraulic and core degradation model (typically using an integrated package like MELCOR [4] or SCDAP/RELAP) so that specific accident scenarios can be evaluated and the magnitude of release from the fuel can be determined for each case. Experimental data based on release under isothermal conditions have historically been used in accident codes to estimate the release over a given computer code time step (assuming isothermal conditions over the time step). In this way, steady-state, isothermal test data can be applied to a variety of transients.

This approach is most appropriate for cases of purely diffusive release from molten fuel but may not capture all phenomena in cases of mechanical fuel failures or behavior at the onset of melting. Hence, some characterization of these phenomena also must be considered. For example, commercial light water reactor fuel rods will initially rupture before fuel melt, releasing all of the fission products contained within the fuel/cladding gap. Metal fuels can undergo different phenomena at the onset of melt, such as “burst” releases upon melting of the cladding and fuel foaming depending on the amount of fission product gas pressure within the fuel, and the amount of fuel constraint.

In addition to fission product release from the fuel, these codes are also typically capable of modeling fission product transport in the primary system and containment (confinement for the ATR) and are used to estimate the source term to the environment. Given the environmental source term, codes such as RSAC [5] or MAACS [6] are used to estimate the onsite and offsite dose which can then be compared to regulatory limits.

At present, the chemical forms of fission products released U-10Mo fuel are not expected to be significantly different from the chemical forms for other uranium fuel systems. While some effort is required to characterize the specific chemical forms released from U-10Mo fuel, it is currently expected that the existing database (e.g., from commercial reactor experimental programs) on fission product transport behavior in the primary system can be utilized to support ATR LEU conversion without additional experimental characterization. Hence, the key data required are U-10Mo fission product release rates.

3 Fission Product Release Testing for Aluminide Fuel

The fission product release rate models currently utilized in the ATR SAR for aluminide fuels are based on a series of release rate tests performed in the 1950s to 1970s. The data were taken at multiple laboratories across a variety of temperature and atmosphere (e.g., inert vs. air vs. steam) conditions. Since the ATR is a high power density reactor and since the aluminide fuel design used in ATR is characterized by high thermal conductivity, low heat capacity, and relatively low melting point materials, the accident progression that occurs is typically much faster than a severe accident progression in a commercial reactor. The period encompassing melting and relocation of the fuel occurs on the order of minutes. Hence, several of the tests that were conducted on aluminide fuels consisted of setups that could attain rapid heating rates (20-30 °C/s) and heat samples for short hold times.

A summary of the tests that form the basis of the current aluminide fuel fission product release rates is provided in Table 1. The key experimental parameters to note are the range of temperatures considered (several hundred degrees above the UAl_x melting temperature), environment (air, steam, inert), and heat-up rates.

This information was used to inform the basis for the type of testing that must be performed for U-10Mo fuel to provide a level of characterization similar to that of aluminide fuels.

Table 1. Summary of Testing Basis for Aluminide Fuel Fission Product Release Rates

Fission Product Species	Source	Comments
Noble Gases	[7], [10]	Combination of long (24-30 minute) and short (2 minute) hold-time data at temperatures of 700°C – 1145°C in helium, air, and air+steam environments
Iodine	[8], [9], [10]	Short (2 minute) hold time data in a steam environment consisting of rapid heating rates to test temperature (20-30 °C/s)
Cesium	[8], [9]	Short (2 minute) hold time data in steam environments with rapid heating rates to test temperature (20-30 °C/s).
Tellurium	--	Due to significant differences between estimates of tellurium release from multiple sources, iodine release rates are used to model tellurium release

4 Requirements for U-10Mo Fission Product Release Testing

Preliminary requirements were developed to provide a framework for evaluating a set of high level options to obtain U-10Mo fission product release data (see Section 5). The requirements developed (See Table 2) are general to allow for flexibility in the options for completing the testing. Once a general experimental approach is selected, more specific requirements will be developed for detailed experiment design.

The requirements are split between technical (setup capabilities) and programmatic requirements (cost, schedule, risk). The most challenging technical requirements relate to the heating rate

(Requirement #1), test environment (Requirement #6), and the ability of the test apparatus to capture release phenomena at the onset of fuel degradation (Requirement #10).

Table 2: Preliminary U-10Mo Fission Product Release Testing Requirements

Category	#	Requirement
Technical Requirements		
Heating Capability	1	The test apparatus shall be able to heat to the test temperature rapidly and allow for testing of samples at isothermal conditions to the maximum extent practical.
	2	Tests shall be performed between the melting temperature up to several hundred degrees above the fuel melting temperature (T_{melt} ~1130°C).
	3	Tests shall be performed long enough for a significant release of pertinent fission products to occur (e.g., noble gases, cesium, iodine).
Fuel Samples	4	Fuel sample initial composition shall be quantified and representative of LEU fuel at peak ATR burnup.
	5	Fuel sample design and fabrication shall ensure that the fission product release behavior is representative of a LEU fuel plate.
Data Acquisition	6	Tests shall characterize the impact of an oxidizing environment on fission product release rates.
	7	Test assembly materials shall minimize chemical interactions with the fuel sample and fission products.
	8	At a minimum, fission product release measurements shall consider noble gases, iodine, and cesium.
	9	The cumulative fraction of fission products released over the duration of a given test shall be determined.
	10	Rapid “burst” releases shall be characterized to the maximum extent practical.
	11	The temperature history of the fuel samples shall be measured.
	12	The chemical behavior of the cesium and iodine release products shall be identified.
Programmatic Requirements		
Programmatic	13	Technical risk of testing should be minimized by focusing on proven technologies.
	14	Cost should be minimized to the extent practical and appropriately balanced with technical requirements.
	15	The data shall be available to support the ATR ET-3 test.

5 Alternatives Assessment for Acquiring Fission Product Release Data for U-10Mo Fuel

In order to meet the requirements discussed above, an evaluation of alternatives was performed to determine the best testing approach. As part of this evaluation, three major options were considered to perform U-10Mo fission product release testing:

1. Thermal heating using a furnace within the Hot Fuels Examination Facility (HFEF) or other suitable hot cell
2. Nuclear heating in the Transient Test Reactor (TREAT)
3. Utilize an existing capability to perform fission product release testing (i.e., a pre-existing facility for measuring fission product release rates for nuclear fuel).

The three options were evaluated against the set of technical (test capability) and programmatic (cost, schedule, risk) requirements discussed above.

For Option 1, the key considerations are as follows:

1. Heating (furnace) capability
2. Data acquisition capability
3. Environment

For furnace capability, use of both an existing furnace and developing a new furnace capability were considered.

With respect to data acquisition, two different approaches were considered: 1) real-time measurement using on-line data collection with gamma spectroscopy and 2) static measurement, which also utilizes gamma spectroscopy but relies on pre- and post-test assays of the sample to determine the relative change in fission product concentration over a particular time period. While real-time measurement can better characterize the release rates, it is also more complex and costly. The static measurement approach is easier to implement, but has higher uncertainties and requires a larger number of samples in order to provide sufficiently high fidelity estimates of time-resolved fission product release rates.

Finally, the evaluation considered inert, air, and steam testing environments. Inert and air environments are easier to implement but are less prototypic. However, it was determined that testing in a steam environment would not be cost-prohibitive and is preferred due to it being much more prototypic for ATR accident conditions.

For Option 2, the evaluation considered two different approaches for utilizing the TREAT reactor to perform fission product release testing for U-10Mo fuel:

1. Utilize existing test vehicles planned for near term use in TREAT that were originally developed to support other fuels programs.
2. Develop new TREAT capability specifically for performing U-10Mo fission product release testing.

TREAT offers significant capability to perform prototypic sample heating in prototypic environments (including water/steam). However, test capability planned for near term deployment has not been developed with fission product release rate measurements as a focus and, therefore, lacks suitable instrumentation. While it may be possible to utilize a “static” approach similar to what was discussed for furnace testing, the currently planned TREAT test vehicles are all closed systems that could allow re-contaminating the sample with released fission products, leading to errors in the estimates of release. Addressing this issue would likely require modification of planned test vehicles which introduces risk and likely cost.

Development of new TREAT capability to support U-10Mo fission product release testing was determined to be to have higher costs and risks relative to furnace options and was not expected to meet the schedule to support the ATR LEU conversion program.

For Option 3, facilities that have existing fission product release testing capability were considered. However, performing U-10Mo testing in the time frame required did not fit within the current testing schedule of these facilities. Additionally, use of these facilities would require international shipments of irradiated fuel, which introduces additional cost and complications.

Based on the evaluation of all three options, the approach chosen for U-10Mo fission product release testing is to utilize a new furnace to be installed within a hot cell. A new furnace could attain the heat-up rates necessary to meet U-10Mo testing requirements that current furnace capability lacks. Additionally, a static measurement approach and testing in a steam environment are recommended.   The additional uncertainties due to using a static measurement approach were considered acceptable and results in much less risk and cost to implementation. Additionally, testing in steam environments is most prototypic for the ATR. In general, the recommended option was determined to be the best balance of limiting technical, risk, schedule, and cost requirements.

The notional test setup for the preferred approach is shown in Figure 1.

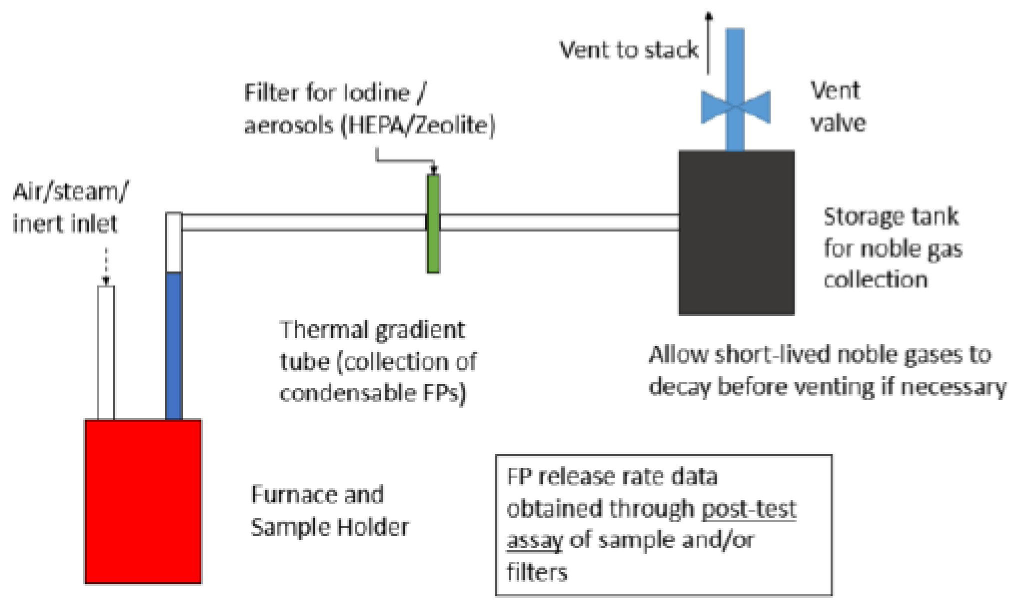

Figure 1: Notional Fission Product Release Test Setup

The approach consists of melting samples in a furnace that is fed by a gas flow that will expose the melting sample to various environments and also sweep released fission products away from the sample to be collected downstream. Fission products are collected using various means depending upon their physical state:

- Condensable fission products: Condensable fission products are typically captured in a "thermal gradient tube." The tube is made of a chemically inert material that has a controlled temperature gradient applied to it that allows condensable fission products to plate out on the tube inner surface at locations corresponding to the condensation temperature of the substance.
- Aerosol fission products: Aerosol fission products (solid particulates or liquid droplets) are trapped using various filters, such as activated charcoal or zeolite filters.
- Gaseous fission products: Gaseous fission products are those that remain in the gas phase over a wide range of temperature such as noble gases (e.g., krypton, xenon). Gaseous fission products are typically captured by means of a storage tank to collect the gases and

potentially hold them for a period of time to allow them to decay. Depending upon the level of radioactivity in the sample, it may be possible to directly vent to the atmosphere.

Testing should include at least a steam environment (the most prototypic), but sensitivity to atmosphere, particularly to an air atmosphere, should also be better understood due to the potential for air ingress during a severe accident.

6 Plan for Obtaining Fission Product Release Data for U-10Mo Fuel

The high level plan for acquiring and utilizing U-10Mo fission product release data is shown in Figure 2. The plan consists of three major phases:

1. Pre-conceptual design (FY20): in this phase the furnace design concept will be finalized, detailed requirements developed along with a detailed cost and schedule estimate.
2. Design, procurement, installation, and test execution (FY21-FY23): During this phase, the detailed design will be developed, procurement and installation will occur, and the test will be performed.
3. Model Validation and SAR Updates (FY24-FY25): During this phase, the ATR LEU severe accident model (developed in the MELCOR code) will be validated using the fission product release data, and updates to the ATR SAR will be developed to support insertion of the high power lead test element (LTE; the ET-3 test) as part of the ATR LEU fuel qualification.

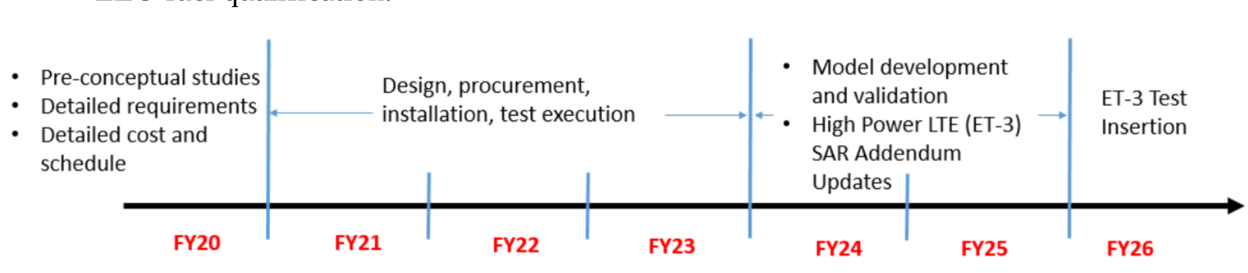

Figure 2: High Level Plan for Development and Utilization of U-10Mo Fission Product Release Data

7 References

[1] C.M. Allison et al., "SCDAP/RELAP5/MOD2 Code Manual, Volumes 1, 2, and 3", NUREG/CR-5273, EGG-2555, June 1989.

[2] Lorenz, R. A., Osborne, M. F., NUREG/CR-6261, "A Summary of ORNL Fission Product Release Tests with Recommended Release Rates and Diffusion Coefficients", Oak Ridge National Laboratory, July 1995.

[3] Sehgal, B. R., "Nuclear Safety in Light Water Reactors: Severe Accident Phenomenology," Academic Press, 2012.

[4] Lorenz, R. A., Osborne, M. F., NUREG/CR-6261, "A Summary of ORNL Fission Product Release Tests with Recommended Release Rates and Diffusion Coefficients", Oak Ridge National Laboratory, July 1995.

[5] Schrader, B. J., "Radiological Safety Analysis Computer (RSAC) Program Version 7.2 Users' Manual", INL/EXT-09-15275, Rev. 1, October 2010.

[6] Jow, H. N., Sprung, J.A., Rollstin, L. T., Ritchie, L.T., Chanin, D. I., "MELCOR Accident Consequence Code System (MACCS), Model Description", NUREG/CR-4691, SAND86-1562, Vol. 2, 1990.

[7] ORNL-3981, “Out-of-Pile Studies of Fission Product Release from Overheated Reactor Fuels at ORNL, 1955-1965”, dated July 1967.

[8] HEDL-7598, “The Release of Fission Products from Irradiated SRP Fuels at Elevated Temperatures, Data Report on the First State of the SRP Source Term Study”, dated March 1987.

[9] HEDL-7651, “The Release of Fission Products from Irradiated SRP Fuels at Elevated Temperatures, Data Report on the Second Stage of the SRP Source Term Study”, dated March 1987.

[10] Shibata, T., et. al., “Release of Fission Products from Irradiated Aluminide Fuel at High Temperatures”, Nucl. Sci. Eng. **87**, 405 (1984).