INL/CON-08-13781
PREPRINT

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ICONE 16

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May 2008

Idaho National Laboratory

Proceedings of the 16th International Conference on Nuclear Engineering
ICONE16
May 11-15, 2008, Orlando, Florida, USA

ICONE16-48453

SECOND GENERATION EXPERIMENTAL EQUIPMENT DESIGN TO SUPPORT VOLOXIDATION TESTING AT INL

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ABSTRACT
Voloxidation is a potential process used to prepare fuel for spent-oxide-fuel treatment. The spent oxide fuel is heated to an elevated temperature in oxygen or air to promote separation of the fuel from the cladding. Low pressures are also required to enhance volatizing the fission products. The Idaho National Laboratory and the Korea Atomic Energy Research Institute have been collaborating on voloxidation research through a joint International Nuclear Energy Research Initiative project. The new high temperature multiple zone furnace and off-gas trapping system (OTS) are designed for remote operation in the Hot Fuel Examination Facility. The OTS is designed to handle vacuum, oxidizing environments, and high temperatures. Distinctive temperature zones of the furnace are used to selectively capture the fission products. Vacuum conditions at elevated temperatures are achieved by incorporating various metal seals. The OTS has proved durable under these aggressive operating conditions. A detailed description of the second generation furnace and OTS with enhanced capabilities will be presented.

INTRODUCTION
Idaho National Laboratory (INL) and the Korea Atomic Energy Research Institute (KAERI) have been investigating an advanced head-end processing concept to declad and oxidize (DEOX) spent oxide nuclear fuels. The objective of the DEOX process is to achieve separation of the spent fuel from the cladding. The feed material is spent fuel from light water reactor uranium oxide (UO_2) or fast reactor mixed oxide (MOX). Remote tools are used to cut the spent fuel into shorter segments that fit within the operating envelope. The main pieces of equipment are used to heat the segments in an oxygen-rich environment and to collect the off-gas fission products. As the UO_2 oxidizes to U_3O_8 per the reaction $3UO_2 + O_2(g) \rightarrow U_3O_8$, it undergoes a corresponding volume increase, which pulverizes the fuel. This volume increase causes the pin cladding to swell or split, allowing the fuel to escape as shown in Figure 1. The fuel may then be treated by either aqueous or pyrochemical methods. In addition to decladding and pulverization, application of the DEOX process to fuel results in the release of a portion of the volatile fission products.

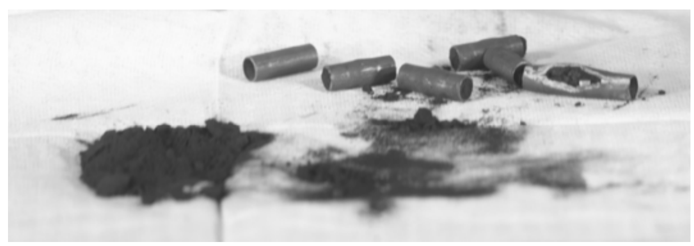

Figure 1 Spent fuel after voloxidation experiment

The oxidation process utilized in DEOX is based on the Atomics International Reduction Oxidation (AIROX) process developed by the Atomics International Corporation in the 1960s and 1970s [1]. AIROX studies found the release of fission products correlates to the extent of oxidation, the degree of pulverization, the processing temperature, and the processing pressure. After oxidation was completed and the fuel was returned to an inert atmosphere, additional volatilization was observed when the fuel was raised to temperatures as high as 1100°C. Operating at slightly under atmospheric pressure also increased total release of the fission gases [2].

The first-generation experimental equipment for DEOX [3] (see Fig 2) was designed primarily for separating the spent oxide fuel from the cladding and for removal of the fission products. One objective of the research was to study the effects of cladding, air, vacuum, and elevated temperatures on fission-product removal from the spent nuclear fuel [4-8]. Several tests were conducted to determine the optimum conditions for de-cladding and fission-product removal [7].

The focus of further research is to optimize collection of fission gases via an off-gas trapping system (OTS). A new furnace and OTS, with enhanced capabilities, were required to perform additional tests. It was initially thought that the first-generation equipment could be retro-fitted with an OTS; however, it was concluded that this would be difficult so a new furnace was designed. The first generation furnace was also only capable of 1100°C and temperatures in excess of 1100°C would be required for complete removal of selected fission products. Another obstacle to using the first generation equipment was maintaining an elevated temperature in the interface tubing between the furnace and a new OTS. If cooling were to occur, the fission-product gases would plate out and never reach the filter media.

Figure 2 First-generation furnace and DEOX equipment

NOMENCLATURE

AIROX	Atomics International Reduction Oxidation
AL	Analytical Laboratory
DEOX	Declad and Oxidize
EMM	Electromechanical Manipulator
HFEF	Hot Fuel Examination Facility
LWR	Light Water Reactor
MSM	Master Slave Manipulator
OTS	Off-Gas Trapping System

DEOX EQUIPMENT DESIGN CRITERIA

The DEOX experiments are conducted in a large argon-gas-filled hot cell located at the INL, called the Hot Fuels Examination Facility (HFEF). Technicians perform their work inside the hot cell facility using electromechanical manipulators (EMM's) while viewing components through four-foot-thick glass windows. The new furnace and OTS needed to be designed for remote operation and maintenance, as well as designed to resist radiation degradation.

INL and KAERI representatives met together during the conceptual design process to develop the design criteria for the furnace and the OTS. The overall general requirements were that the system had to accommodate the use of spent nuclear fuel and be capable of operating in the HFEF. Further discussions were used to refine and establish the following specific requirements.

- The furnace must be remotely operable and maintainable using EMMs.
- All heating elements must be remotely replaceable using EMMs.
- The furnace and OTS shall be designed to minimize the footprint area (HFEF in-cell storage requirement).
- The system must be able to accommodate 100 g of spent oxide fuel.
- The furnace must be capable of heating the spent nuclear fuel to 1200ºC and holding for up to 3 hours.
- The OTS must be capable of operating with air or oxygen, with a flow rate up to 0.5 L/min at elevated temperatures.
- The furnace and OTS must be capable of operating in an inert argon-gas environment.
- The OTS shall be designed so that all the off gas flows through the filter media.
- The OTS that holds the spent nuclear-oxide fuel and the filter media shall be designed for remote disassembly using manipulators.
- The filter cartridges (which include the filter media) shall be designed so that they can be shipped from the HFEF to the Material and Fuel Complex Analytical Laboratory (AL) using the pneumatic rabbit system.
- The filter cartridges and crucibles shall be designed so that before and after weight measurements can be taken.
- The materials chosen for the system shall be compatible with the elevated temperatures and gas media (air or oxygen).

Three major components were identified as needed to meet the design criteria.

- New furnace
- Off-gas trapping system
- Loading/unloading equipment

DEOX Furnace

The new furnace (see Fig. 3) was designed with four separately-controlled heated zones. The bottom zone is capable of operating at 1400°C. This provides ample potential to volatize the spent-fuel fission products. The upper three zones are capable of 1100°C, 1000°C, and 300°C respectively. A split tube design was chosen so the equipment could be operated on a worktable, located at window 10M of HFEF. The split tube design makes loading/unloading at the worktable height easier. The split tube design also allows for independent heat control at each zone. The vertical configuration was chosen to minimize the footprint of the furnace because real-estate space within the HFEF hot cell is limited. The overall dimensions of the furnace are 24” x 24” x 50” height. During design, the vendor worked with the INL to make the heater elements replaceable. The bottom-zone heating elements are silicon-carbide rods, and the upper three zones use standard-resistance heaters in two separate halves. The upper three zones have a working volume of 3” diameter by 6” height. The bottom zone dimensions are 4” x 6” x 6” height. Some remote handling fixtures and connectors were added to the furnace during qualification testing to support remote installation and maintenance.

Figure 3 Second-generation DEOX furnace

Off-Gas Trapping System

The OTS is the vital part of the design effort. The OTS consist of four separate zones (see Fig 4), which fit inside the DEOX furnace. The upper three zones are used to hold various filter assemblies [9]. The bottom zone holds the spent-fuel segments. Oxygen or air is introduced through the inlet at the bottom, and a vacuum is attached to the outlet. The entire assembly is leak tested to make sure the majority of the fission-product gases stay within the system. Creating a design that would hold a vacuum at the requested temperature limits was a challenge because standard vacuum equipment is only rated for 650°C.

Figure 4 DEOX Off-Gas Trapping System

The structural hardware for the filters was designed to meet the established criteria. The mode of transportation for the filters to the AL through the pneumatic-tube system was the major factor in determining the diameter. The filter had to fit in the largest available sample container designed for the pneumatic system. The height of the individual filters was chosen so it would be possible to see if a gradient existed in fission-gas capture along the column. Having the option to choose the number of filters was also a factor in determining the height. The filter dimensions are one inch diameter by half inch height. Each DEOX test may required more or fewer filters depending on the character or quantity of the spent fuel used in each experiment. The filter media used in each zone is as follows; Zone 1—Silver impregnated Zeolite, Zone 2—Calcium Oxide, and Zone 3—Fly Ash [10].

The top zone (Zone 1) operates at a much lower temperature (150-200°C) so that initially Viton O-rings were used between the filters (see Fig 5). Presently, Viton O-rings have been replaced with Kalrez 4079 O-rings, which have an operating temperature limit of 316°C. This new material supports a sustained 200°C bake-out that is performed on the top filter assembly prior to testing to remove moisture. Each filter assembly zone has a spacer at the bottom with a 320-mesh screen to capture any filter media which might break off during testing. This spacer also allows the fission gases to contact the entire surface area of the filter instead of being concentrated at the center of the filter as they emit from the 3/8-in. tubing.

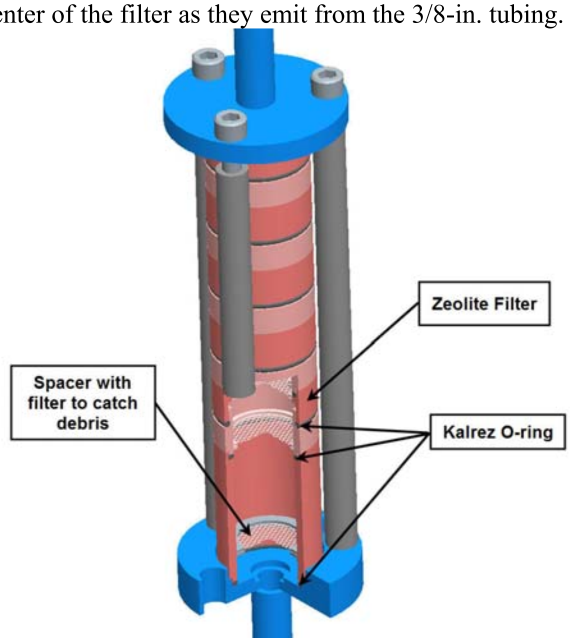

Figure 5 Zone 1 Filter Assembly

Providing a seal between each filter in the middle zones (2 and 3) at elevated temperatures was accomplished by using a metal C-ring (see Fig 6). The C-ring selected is shaped in such a way that external pressure pushes on the inside of the ring, helping to maintain the system seal. The C-ring material is Waspaloy with a nickel coating. The original design criteria required this seal to function at 800°C. Further tests showed that an operating temperature of 1000°C would improve the performance of the trapping media. Subsequent tests have shown that a vacuum of less than 1000 mtorr can be maintained at this operating temperature, indicating the seals functioned adequately above their design temperature during the relatively short test.

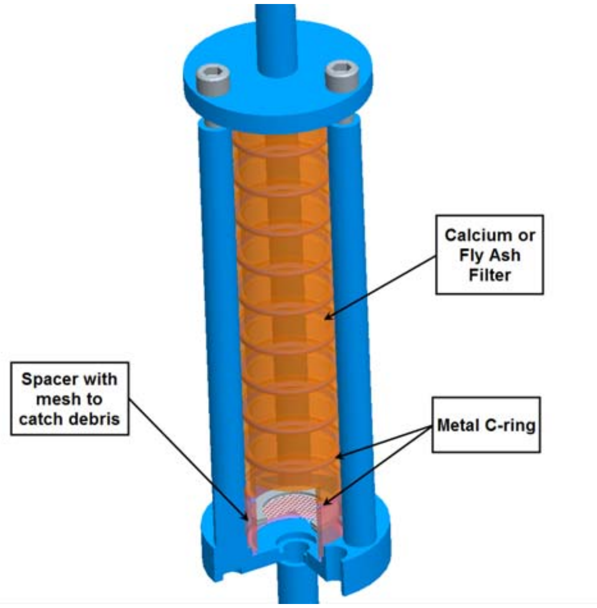

Figure 6 Zone 2 and 3 Filter Assembly

The bottom (Zone 4) of the DEOX furnace is where the highest temperatures and the varied oxidizing environments are encountered. In one of the earlier tests, a ceramic crucible and Inconel fuel holder were used to hold the fuel segments (see Fig 7). Complications were encountered. First, the alumina crucible contained large amount of silica that reacted with the fission gases prior to reaching the filters. Second, the oxidization was not ideal, because oxygen had a torturous path to get to the fuel. The design was modified so the crucible and fuel holder were both fabricated from Inconel. The Inconel crucible has a hole through the center where the oxygen is introduced directly to the fuel (see Fig 8). The fuel holder, which sits atop the crucible, is designed to support the fuel during handling and loading operations (see Fig 9). The diameter of the holder into which each segment is placed is sized sufficient to allow the cladding and fuel to swell. Holes in the holder's bottom allow the fuel to fall into the crucible.

To create a vacuum seal at 1200°C in an oxygen-rich environment created unexpected challenges. Material selection and seal designs were discussed with vendors of high-temperature seals with a few iterations. No viable solution could be obtained through the contacted vendors. It was decided that a knife-edge design with a flat pure nickel gasket could potentially provide the seal at the required temperature. Tests were performed, and results show that a vacuum seal can be maintained at 1200°C using this design.

Figure 7 Alumina crucible and fuel holder with handling tool

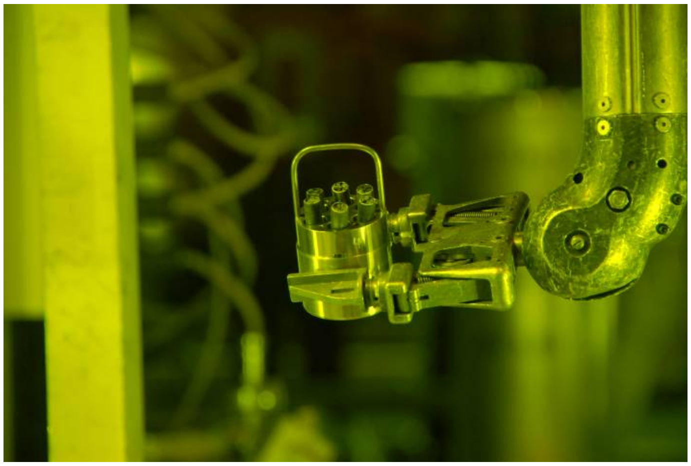

Figure 9 Spent fuel loaded into an Inconel crucible assembly

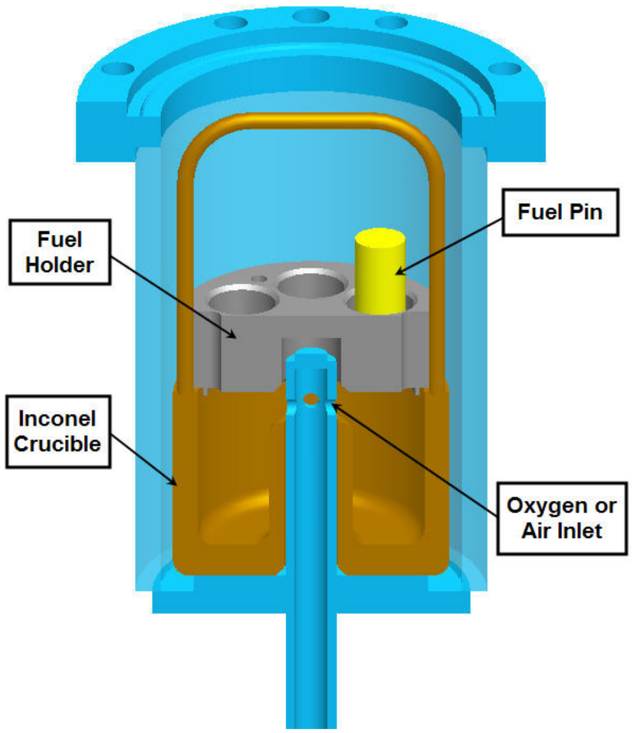

Figure 8 Inconel crucible and fuel holder (cross section)

Loading/Unloading Equipment

This equipment was designed to support the OTS during loading and unloading operations (see Fig 10). The entire OTS is leak checked prior to being transferred to the hot cell. In order to load the spent fuel, the filter zones and crucible lid must be removed. Because the structure of the filter zones is long and spindly, it requires additional support. A stand, located to the left, is used to support the upper portion of the OTS while fuel is loaded. The stand at the right supports the entire OTS assembly. A quick-release pin is used to hold the upper section. The crucible holder sits in the tube on two guide pins. These guide pins are used to resist the force created during bolt torque.

Figure 10 DEOX OTS loading/unloading equipment

RESULTS

The second generation equipment to support DEOX testing was designed, fabricated, and tested. The system meets all of the design requirements and has performed well during testing (see Fig 11). The furnace zones have operated at the specified temperatures with very consistent heating results for each experiment. The seals used in the OTS have functioned better than expected. The vacuum that has been achieved has been in the 100s of millitorr range. The Inconel 600 crucible and the fuel holder have withstood the elevated temperatures and oxygen environment (see Fig 12) and are reused. Sufficient oxygen is getting to the spent fuel for the reaction to U_3O_8 (see Fig 13). The loading/unloading equipment has made disassembly feasible using the MSM in the hot-cell environment. As an added benefit, the vertical design has allowed the system to be gamma scanned inside the HFEF hot cell [9]. This data provides a preliminary cesium distribution before the filters are sent for analytical chemistry.

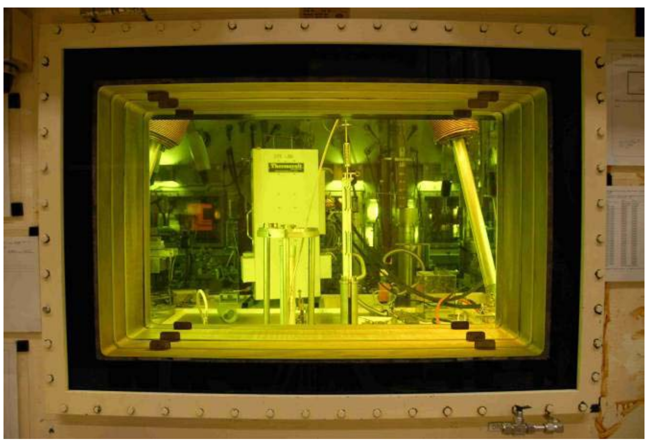

Figure 11 DEOX furnace and OTS installed in HFEF.

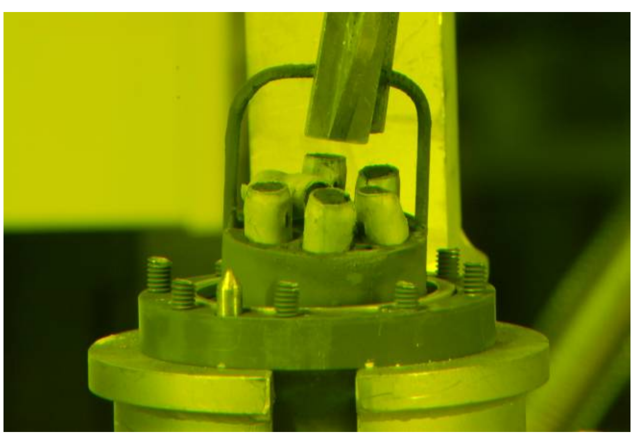

Figure 12 A close-up view of the Inconel crucible and spent fuel after DEOX processing.

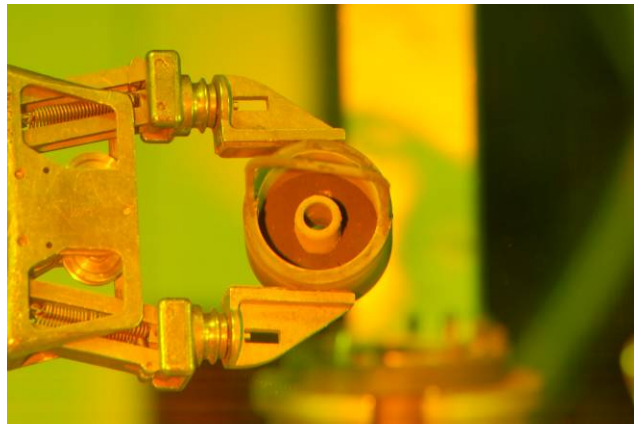

Figure 13 A close-up view of the Inconel crucible with powdered fuel created during DEOX processing.

CONCLUSION

The second-generation DEOX equipment was designed and fabricated. The system has performed well during experimental testing. The furnace has provided consistent temperature profiles during testing. The vertical design of the furnace gave it a small foot print of 24” by 24”. The once-through design of the OTS has resulted in a consistently leak tight system and has provided valuable information to the researchers on how the off-gases from the spent fuel are trapped in the filters. The Inconel crucible and fuel holder are reused. Both these items have proved durable, even under aggressive operating conditions. Overall, the system is performing well and the experimenters continue to push the operating parameters as new information about the process is obtained.

Future improvements to the equipment could include an improved high-strength high-temperature fastener for the 1200°C zone. The current fastener breaks during disassembly, which allows the equipment to be taken apart, but requires extra time and does not allow the crucible holder to be reused. Furthermore, the addition of flow controllers that can transmit data to the data logger would allow for more precise flow conditions of the oxidizing gas.

ACKNOWLEDGMENTS

This manuscript has been authored by Battelle Energy Alliance, LLC under Contract No. DE-AC07-05ID14517 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a nonexclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

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