AN APPROACH TOWARD IN-CORE DETERMINATION OF POSSIBLE FUEL ELEMENT CLADDING RUPTURE

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ABSTRACT

Routine pool water sampling at the Annular Core Research Reactor (ACRR) detected a low-level presence of radionuclides that might be expected if a fuel element cladding failure had occurred. Air sampling showed no increase in airborne radioactivity levels. Reactor operations and a wide team of supporting staff began a detailed investigation to determine the source of the radionuclides. Ultimately, the determination was made that no detectable fuel element cladding failure had occurred. However, demonstrating the continued integrity of the fuel required significant time and effort. The intent of this paper is to present the method of measurements employed to identify the source, to facilitate a return to normal operations in similar events.

BACKGROUND

Some radionuclides are expected to develop in the ACRR pool water due to activation of structural materials and the creation of radionuclides from irradiation of water and air (H-3, O-19, N-16, Ar-41). Particulate activation based nuclides have been detected in pool water samples, filtration media and water polishing resins throughout the sampling history of the facility. Their concentration in bulk pool water is typically very low. No residual contamination has ever been detected during activities requiring open access to the Bulk or Cleanup Loop Systems. On 27 August 2015, ACRR conducted a high power (5600 MJ), long-term (multiple hours at elevated power) steady-state operation. Shortly after the reactor was shut down, a standard 500 mL Marinelli water sample was collected from the top of the reactor tank. Following the collection protocol, it was delivered to the Radiation Protection Sample and Diagnostic (RPSD) Laboratory for gamma spectroscopy analysis.

The results indicated a high activity of Np-239, with a concentration of 1070±105 pCi/mL and minimum detectable concentration of 1.79E2 pCi/mL. U-238 transmutation is the only source of Np-239. Its presence in these concentrations could be indicative of a fuel element failure. All available water sample data from previous years was reviewed to determine if this was a unique event or a common occurrence. An incomplete set of water samples results dating back to 1999 existed, and changes to RPSD sample archival systems during the intervening years thwarted further search efforts. This set was found to have only occasionally detected Np-239 at levels from 0.08 - 132 pCi/mL. Previous RPSD measurements occurred at irregular intervals, both in terms of timing and power history. A determination had to be made if the current measured levels were acceptable or too high given the recent power history. Coincidentally, a pre-filter from the Cleanup Loop System was replaced before the long operation. Fission products such as I-131, Cs-134, Cs-137, Ce-144, Ru-106, Zr-95, Nb-95 were detected by RPSD. The possibility of a fuel element failure could not be ruled out.

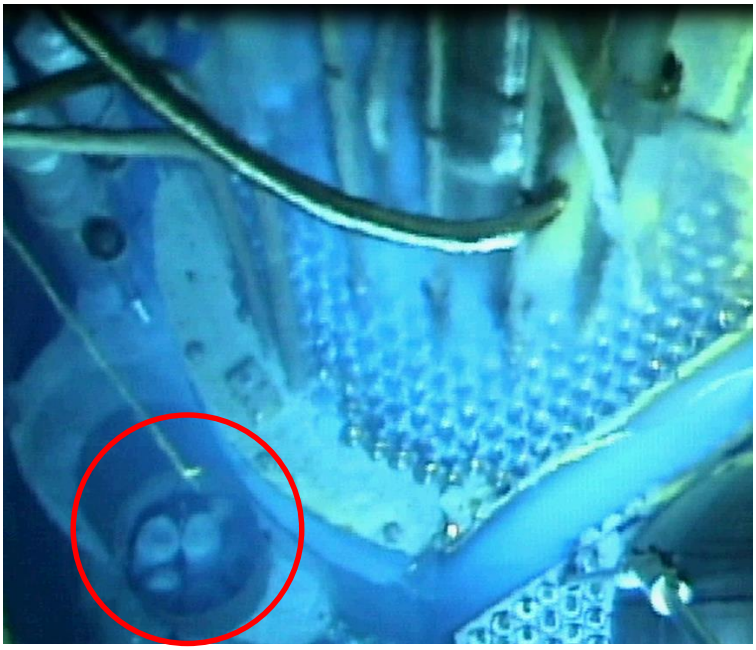
The Radiation Metrology Laboratory (RML) offered its gamma spectroscopy equipment and expertise in the effort to help determine the source of the radionuclides. The RML provides support for experiments conducted at ACRR. Typically, this includes independent, NIST-traceable, standards-based measurements of flux monitors and thermo-luminescent dosimetry. Unlike most radiometric laboratories associated with reactors, the RML does not provide personnel dosimetry services nor surveys of record. Radiation Protection provides gamma spectrometry of routine water samples and maintains and operates the continuous air monitors (CAMs) integrated into the reactor's safety equipment.

METHODOLOGY

To eliminate the possibility of a false positive, after suspect isotopes were first identified from RSPD analysis of routine water samples from the top of the core tank and filtration media, the RML performed an independent analysis of similar samples. The same isotopes were identified.

The ACRR pool requires approximately 280 gallons per month of make-up water to maintain pool height at the operable level. Supply water is filtered through a standard resin purification tri-bed (anion-cation-mixed bed) system. The feed water is potable water supplied to the facility and contains naturally occurring uranium, which, in Albuquerque, typically ranges from 1 to 2 pCi/L. Because the water purification system is not selective for uranium some uranium is able to pass into the reactor pool. Assuming no filtration ~ 2.1 nCi of U-238 and ~0.01 nCi of U-235 would be added to the tank each month. To determine if the Makeup Water System allowed sufficient natural uranium to enter the

reactor pool, an experiment was conducted which placed sealed bottles of make-up water near the core grid, as shown below. The water was analyzed after irradiation and determined to contain no radioactivity levels above the detection limit.



Make Up Water Activation Experiment

All initial routine water samples were drawn directly from the pool's surface using a dip technique. Detection would require any released fission products to transit through the 20-feet of pool water, diluting significantly as it moved. In order to determine if a more sensitive core effluent sampling technique could detect fission products thereby locating leaking fuel element if it was occurring, water samples were drawn from the surface of the pool and from 10-feet down and 20-feet down while the reactor was operating at 95% steady-state power. While these samples did detect the presence of trace amounts of U-235, no increased fission product concentration was detected.

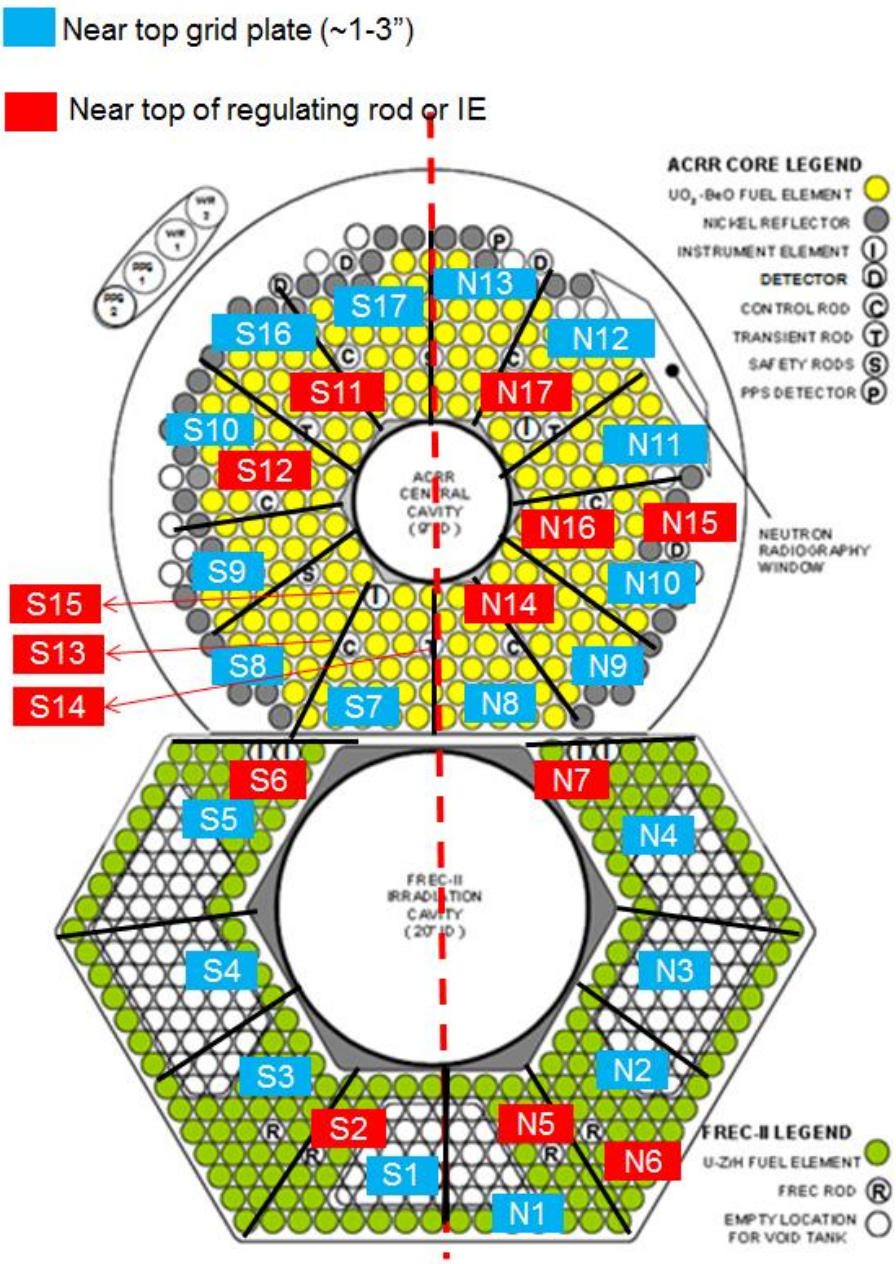
A literature search revealed that The University of Utah experienced a similar event at their 100 kW TRIGA Mark I. Traces of Cs-137 were discovered in their demineralizer resin purification system. A pool sipping technique was developed to draw water from directly above the fuel elements during operation and plumbed to a High Purity Germanium (HPGe) gamma detector. Short-lived noble gases (Kr-85m, Kr-87, Kr-88, and Xe-138) along with low-level decay daughters (Rb-88 and Cs-138) were detected and leaking fuel elements identified.

In an attempt to eliminate the possibility that fission product gasses were being vented or stripped out of solution while collecting the water samples, a similar method was engineered at ACRR to allow water to be pumped directly from the near-core sample location, to a tubing coil around a HPGe detector in an adjacent, lower radiation background room. Images below show elements of this system.

Elements of the Sipping Tool

The heart of the system is a mobile gamma spectroscopy cart (Canberra's ISOCS system) with a portable, electrically-cooled HPGe that can be pointed in various orientations and modular lead shielding. The annulus between shielding and detector was tightly coiled with tubing. As shown in the above image, the detector was turned parallel to the ground to minimize the impacts of any leaking water. The detector efficiency was modeled as a solid ring of water lined with tubing walls surrounding the detector. To cool the hot water coming directly from the core, water was first pumped into an ice bath. This had an additional benefit of permitting the N-16 normally present in the water during reactor operations to decay before coming near the detector. Specific locations at any height in the pool could be sampled with little or no dilution effect and the reactor could be operated at power while sampling was occurring. Two detectors were used to allow for simultaneous sampling of two locations.

To ensure a detectable fission product inventory existed in the fuel, the reactor was operated for at least one hour at a fuel temperature greater than 500 °C in the highest flux region. The lowest flux regions of the core still exceeded 200 °C. After establishing this baseline condition, water was pumped to the HPGe detectors for at least ten minutes in each location. When the sampling head was moved to a new location, a five-minute purging period was allowed to flush the previous sample from the tube before reinitiating data collection. During this period, the flow-rate was adjusted so that O-19, with its 26.9 second half life, would not overwhelm the detector. Sampling locations are shown below.

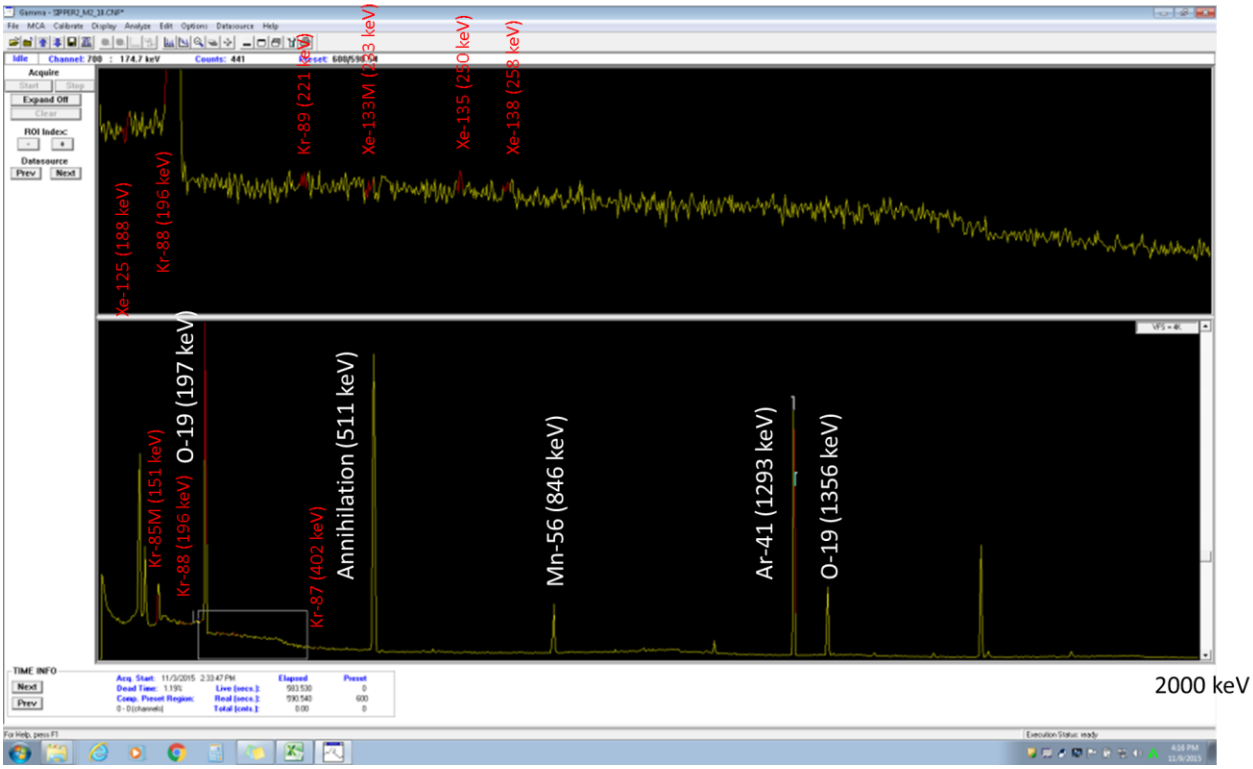


Sample Locations for Core 'Sipping'

Marinelli samples were also collected during the sipping activity for low background analysis in the RML. Overall, noble gases could not be confirmed during real-time sipping operations, and could only be detected when these grab samples were collected and analyzed in a lower background area, with longer count times.

Having acquired these spectra directly over the operating core, no “smoking gun” was found. A typical water sample spectrum is shown below. Results throughout the core were not consistent with a fuel

leak. The ability to perform this measurement in core without disturbing the fuel was invaluable during this investigation, and contributed to an expedited return to normal reactor operations.



Typical Water Spectrum

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