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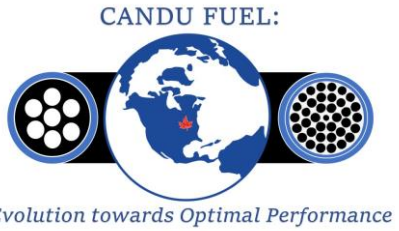

Post-Irradiation Examination of Bruce Nuclear Generating Station Elements at Chalk River Laboratories (CNL)

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ABSTRACT – Bruce Power shipped five fuel elements from five irradiated fuel bundles to Chalk River Laboratories, Canadian Nuclear Laboratories (CNL) in 2013 for routine non-destructive examination (NDE) and destructive examination (DE). Four of the elements were examined to characterize anomalous deposits observed on the fuel cladding.

The deposits were examined in detail using optical microscopy and secondary electron (SE) imaging. The composition of the deposits was examined using a hot-cell analytical Scanning Electron Microscope (SEM) equipped with Energy Dispersive X-Ray Spectroscopy (EDX). The EDX analyses determined the deposit to be composed of primarily iron. The UO_2 pellets and Zr-4 cladding, and hence the fuel performance, appeared unaffected by the deposits. The cladding hermetic integrity for all four fuel elements were confirmed intact.

Introduction

Post-Irradiation Examination (PIE) is used to characterize anomalous cladding deposits and assess their fuel performance implications. Cladding deposits are occasionally observed on irradiated fuel and in most cases, do not affect fuel performance. However, more severe cases of cladding deposits have resulted in increased cladding corrosion and fuel cladding defects, such as the crud induced power shift (CIPS) experienced in pressurized light water reactors [1], [2] and on CANDU fuel rods irradiated in experimental loops under abnormal water chemistry conditions [3]. To date, cladding deposits on CANDU fuel in Canadian reactors have not resulted in fuel cladding failure [4].

In 2013, Bruce Power shipped four fuel elements with unknown deposits on the cladding to Canadian Nuclear Laboratories (CNL) for PIE. The fuel elements were taken from four fuel bundles discharged from two units (Unit 4 in 2010 and Unit 1 in 2012). The fuel elements were partially coated with what appeared to be a thin deposit layer.

It has not been unusual for Unit 4 fuel bundles residing adjacent to the coolant inlet to experience some deposits. However, deposits observed on the Unit 4 elements sent for PIE were from bundles residing adjacent to the coolant outlet. Two Unit 4 elements, which resided adjacent to the coolant outlet and were observed with deposits on the cladding, were sent for PIE to evaluate the composition of the deposits and verify that the deposits had no impact on fuel performance.

As part of the refurbishment and return to service of Units 1 and 2, hot-conditioning was used to prepare the primary heat transport system (PHTS) for operation. This process introduces a protective magnetite (iron oxide) coating on PHTS carbon steel surfaces [4] before the reactor

begins to produce power. Iron is also expected to precipitate on other surfaces, such as the fuel [2] if present, which was the case for the Unit 1 and 2 PHTS during hot conditioning. Consequently, deposits were observed on fuel bundles discharged shortly after Unit 1 and 2 were returned to service. Two Unit 1 elements from bundles present in the core during hot conditioning and discharged after the startup were sent for PIE to evaluate the composition of the deposits and verify that the deposits had no impact on fuel performance.

1. Hot Cell Non-Destructive Examination

Each of the four elements was examined using a specialized stereomicroscope system commissioned in 2011 to operate in CNL's Universal Cells. The new stereomicroscope system replaced a previous stereomicroscope which ported an optical image outside the cell. The replacement stereomicroscope has modernized visual capabilities, including:

- Newvicon Tube Cameras[1] were fixed to the microscope, providing real-time video for fuel element inspections.
- In-cell radiation tolerant optical lenses were used to provide varying magnifications between 8.4x and 37.7x.
- A calibrated measuring system capable of dimensioning the deposit features was implemented.
- Magnification, focus, image/video capture and X-Y stage movement became computer controlled.

Dark deposits were observed on the two elements discharged from Unit 4 (Figure 1). The cladding was covered in a dark coating, with the exception of the endcap regions (Figure 2). The deposits on the two elements discharged from Unit 1 appeared lighter and more granular (Figure 3).

[1] All images in 2013 were taken in grey-scale (a limitation of Newvicon tube camera technology). High resolution 10 mega-pixel colour CMOS cameras were tested in 2014 and replaced the Newvicon tube cameras as the standard in 2016.

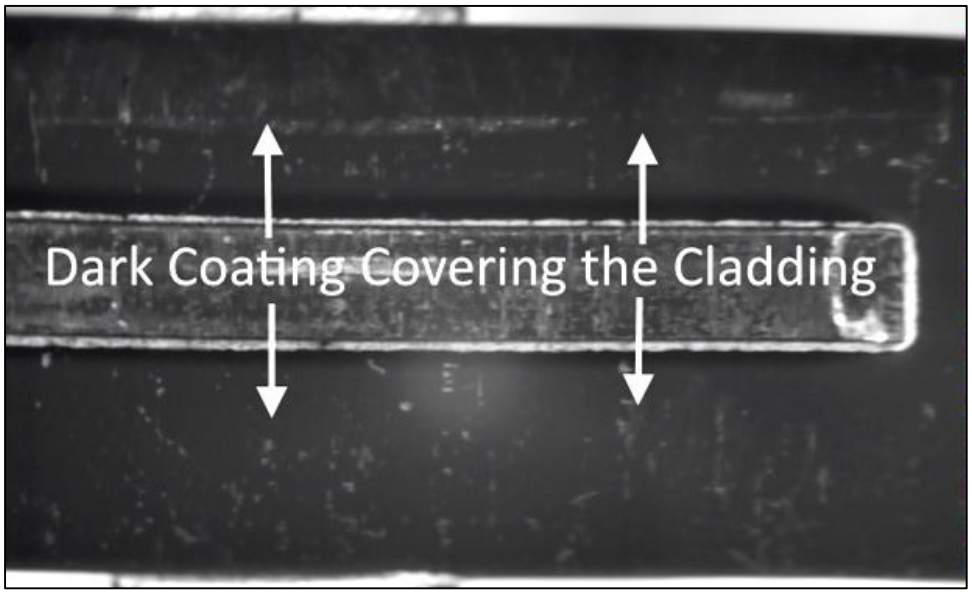

Figure 1 - Example of Deposits Observed on Unit 4 Elements

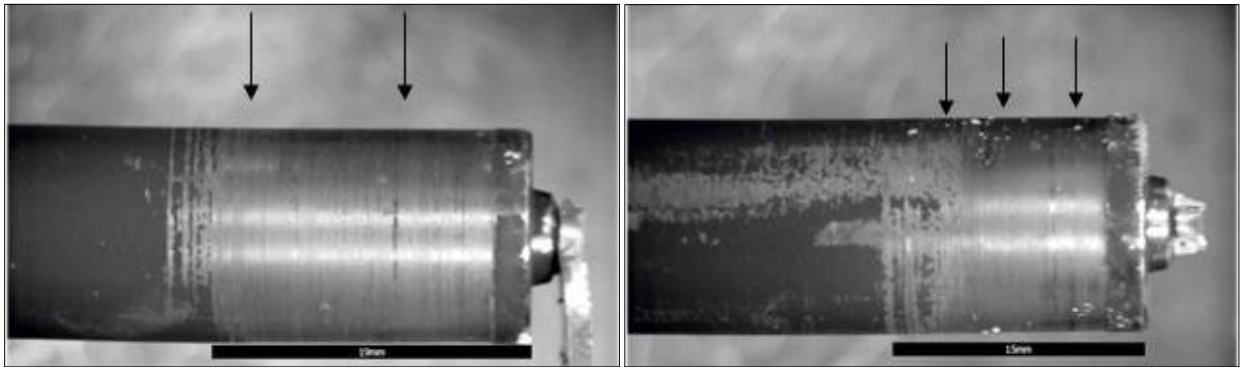

Figure 2 - The Ends of the Unit 4 Elements Appeared to be Relatively Free of Deposit

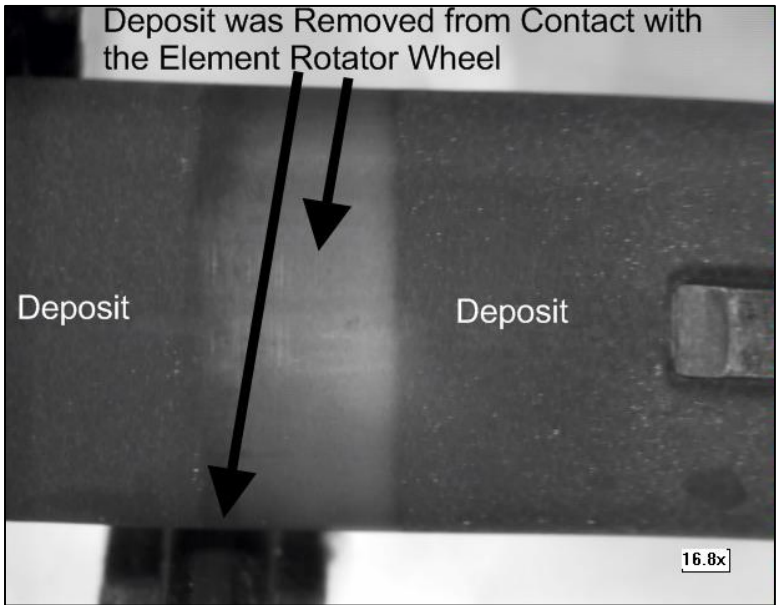

Figure 3 - Example of Deposit Observed on Unit 1 Elements

The deposits on the Unit 4 and Unit 1 elements had several similar features. Deposits were observed on the sides of some of the appendages (Figure 4). Post-discharge handling marks were clearly visible and indicated that simply holding the elements with tooling/hot cell manipulators was sufficient to remove the deposit material. All four elements appeared to be intact. To observe the cladding condition underneath the deposits, it was necessary to destructively examine the elements as described in Section 2.

Spacer pad and bearing pad wear are routinely observed and classified during visual examination. The wear observed on the spacer pads and bearing pads was typical of normal operating conditions.

Endplate material is routinely removed from elements to check for incipient cracking in the endcap-to-endplate welds. This test is called "peel testing" and is performed if sufficient endplate material remains on the elements sent for PIE. The peel tests performed on the deposit elements revealed no indications of incipient cracking in the endcaps-to-endplate welds.

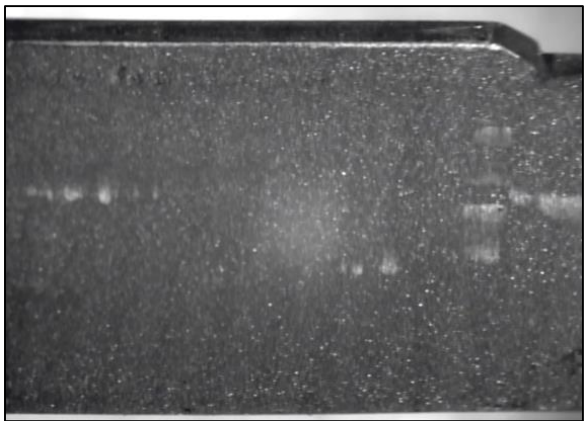

Figure 4 - Deposit Observed on the Cladding and Side Wall of the Bearing Pad

2. Hot Cell Destructive Examinations

CNL's Fuel and Material Cells (FMC) features a series of hot cells dedicated to destructive examination of irradiated fuel and in-core materials, including microscopy, radiography, mechanical testing and other PIE activities requiring high precision. The FMC are also used to prepare samples for chemical and material property analyses, which are performed in active laboratories at CNL.

Each of the four elements was prepared for destructive examination the FMC. Several tests were performed to determine the nature of the deposits and their effect on fuel performance. Sample preparation and examination included the following tasks:

- Sample Cutting
- Macroscopic Visual Examination of Selected Samples
- Microscopic Optical Examination of Transverse Cladding Samples (Metallography)
- SEM Examination
- Deposit Removal and Activation Analysis

Microscopic optical examination (metallography) was performed on transverse samples in a dedicated cell. A shielded Scanning Electron Microscope (SEM) attached to the FMC was used to characterize the deposit morphology and elemental composition. Finally, deposits were removed from the fuel element and transferred to CNL's analytical chemistry laboratories to measure gamma emitting radionuclides for activation analysis.

2.1 Sample Selection and Cutting

At least one metallographic, SEM and activation analysis sample was performed on each element. The activation analysis samples required the largest surface area to collect sufficient deposit material for gamma spectrometry. Metallographic and SEM samples required only 5 to 15 mm of element length per sample and were taken from areas where the deposits were continuous and relatively undisturbed. SEM and metallographic samples were taken adjacent to each other to facilitate direct comparison of their results.

2.2 Macroscopic Visual Examinations

The first stage in examining the metallographic and SEM samples was to select areas of interest for each examination. To observe surface details and select areas of interest on irradiated samples, the FMC uses a low-magnification microscope (up to 20x optical magnification). Features of interest were selected for metallographic and SEM examinations, as follows.

- For each metallographic sample, a specific grinding depth was selected where the deposit was most prominently observed.
- For each SEM sample, an optical image was taken to identify where the deposits were observed, for comparison with the SEM and EDX results. In general, optical imaging and SEM techniques may detect different features due to their intrinsic capabilities (i.e., optical light vs. electron interactions).

2.3 Microscopic Optical Examination of Transverse Cladding Samples

After selection of a grinding depth, four metallographic samples (one from each element) were vacuum impregnated and mounted in an epoxy to stabilize the fuel, cladding and deposit material for examination.

The length of each sample was measured, to determine mounted height and the required grinding depth. A custom built grinder/polisher was used to prepare the four metallographic samples.

Each sample was subsequently transferred to two dedicated hot cells housing optical microscopes used for macroscopic and microscopic imaging. Macroscopic images taken to identify the larger features were unable to resolve the deposits. A high-magnification microscope was used to perform detailed metallography and resolve the deposit features to a resolution of 1 µm.

The polished Zr-4 cladding and associated deposits were examined in detail to assess the clad performance beneath the deposits and provide an optical characterization of the deposits. Figure 5 and Figure 6 show examples of the deposits and underlying oxide from the Unit 4 and Unit 1 samples, respectively. All observed deposits appeared to be porous and were present in non-continuous patches. Deposit thickness on Unit 4 samples did not exceed 20 µm. The deposits on one of the Unit 1 samples were noticeably thinner than the Unit 4 samples; the deposit on the other Unit 1 sample was too thin to be resolved (<1 µm). The ZrO_2 layer thickness for fuel operating to <450 MWh/kgU is typically less than 10 µm [5], but can be greater under abnormal conditions and lead to defects in extreme cases [2]. The ZrO_2 thickness underneath and remote of the deposit patches was less than 10 µm.

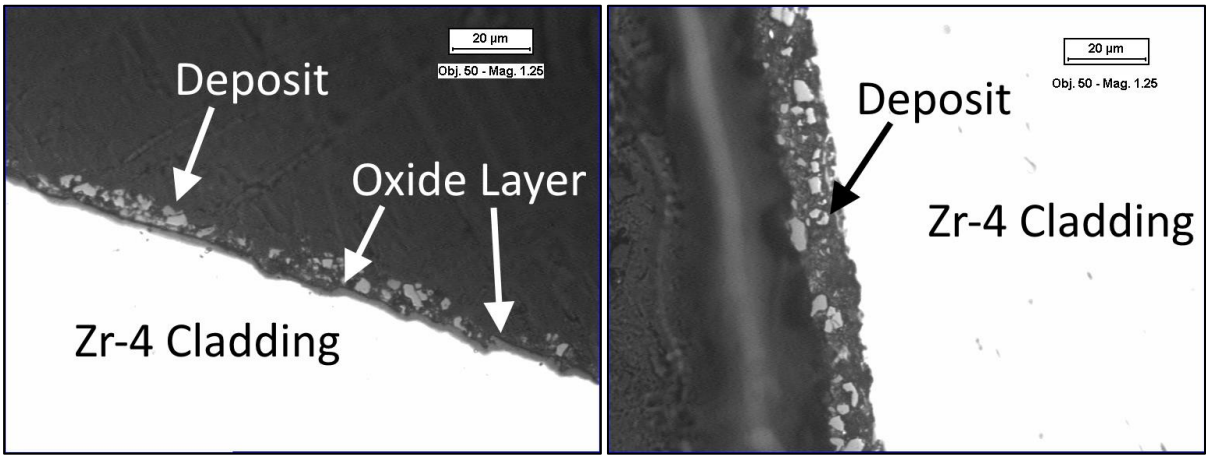

Figure 5 - Microscopic Images of Deposit and Oxide Layers from Unit 4 Elements

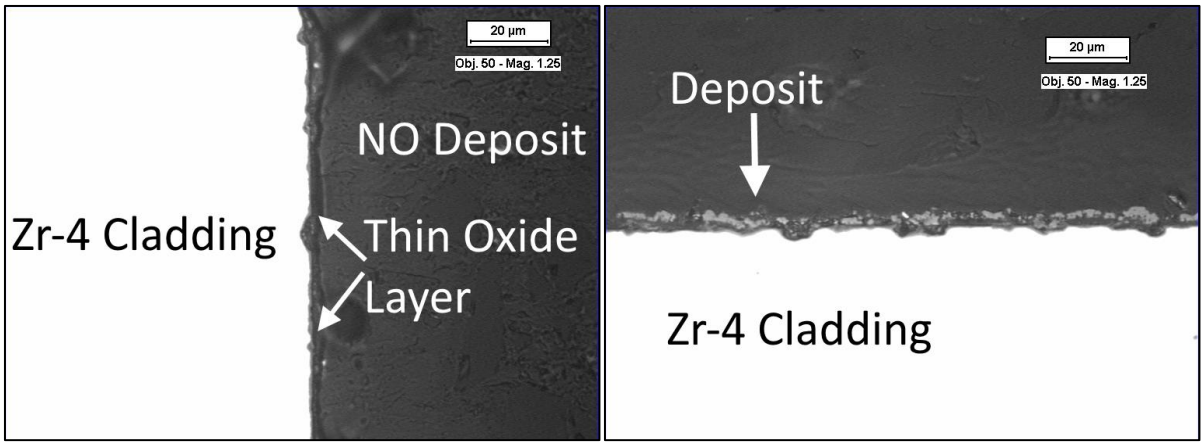

Figure 6 - Microscopic Images of Deposit and Oxide Layers from Unit 1 Elements

Each sample was etched to evaluate cladding microstructure and hydride/deuteride distribution. The samples were taken from different locations along the elements.

The microstructure in proximity to the appendages was heat-affected Widmanstätten (Figure 7) and the microstructure remote of the appendages was as-received cold-worked alpha, as expected. A stronger etch was used to reveal hydride/deuteride platelets (Figure 8). The deposits did not appear to result in preferentially oriented hydrides/deuterides or elevated hydriding/deuteriding.

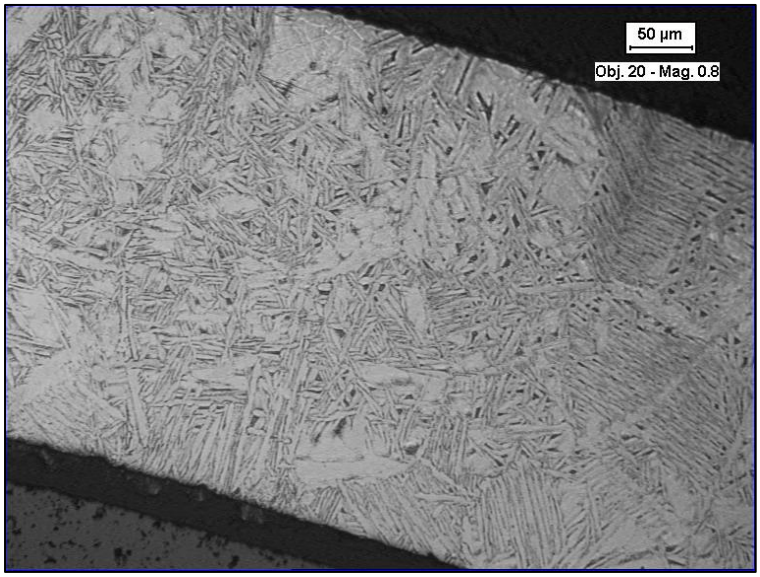

Figure 7 - Example of Heat-Affected Widmanstätten Microstructure

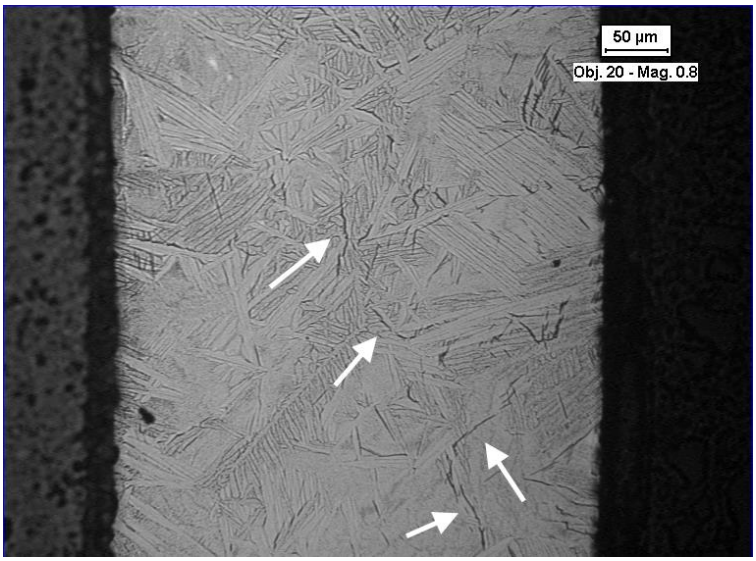

Figure 8 - Example of Hydride/Deuteride Distribution

2.4 Scanning Electron Microscope Examination

Five rectangular cladding samples with deposits were cleaned to remove residual fuel particles and mounted with the deposit facing towards the detector. Each sample was examined using backscatter and secondary electrons in a shielded scanning electron microscope (SEM; Figure 9 and Figure 10). The secondary electron examinations highlighted the topographical details of the deposits. Figure 11 shows a noticeable difference in the deposit morphology between the Unit 4 and Unit 1 samples, indicating that the deposits from Unit 4 formed under different conditions than the deposits in Unit 1.

Figure 9 - Unit 4 Secondary (Left) and Backscatter (Right) Electron Images

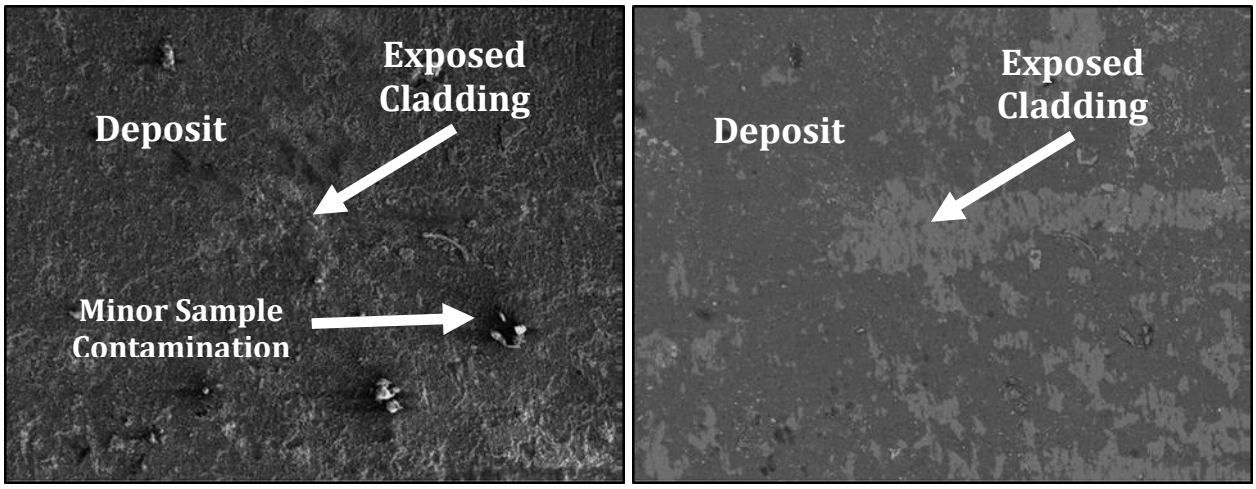

Figure 10 - Unit 1 Secondary (Left) and Backscatter (Right) Electron Images

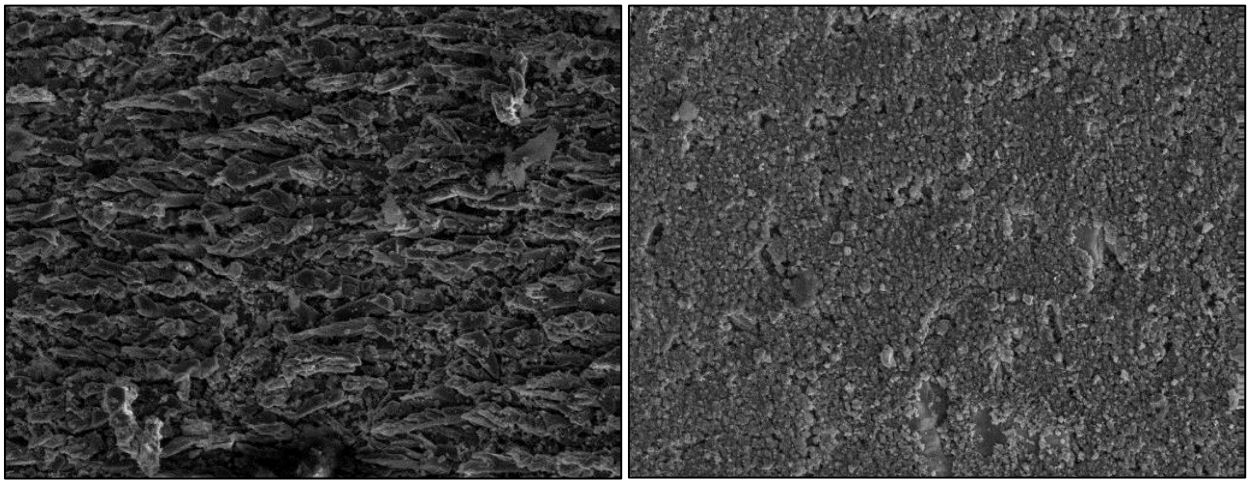

Figure 11 - Different Deposit Morphology Observed on Unit 4 (Left) and Unit 1 (Right) Samples (Secondary Electron)

Backscatter images are sensitive to atomic number and highlight the different elemental composition of the deposits. Figure 9 and Figure 10 backscatter images revealed darker regions, indicating that the deposits had a lower atomic number. The small black spots observed in the backscatter images were attributed to minor contamination.

The deposit material appeared to partially coat the samples, with small distinct regions that were free of deposit. Some of the deposit free features appeared to be a result of deposit removal from handling. However, Figure 10 shows examples of deposit-free patches that do not appear to be associated with scraping from handling; these patches were consistent with the sparse deposition observed during metallographic examination of the Unit 1 deposits.

The shielded SEM includes an energy dispersive X-ray spectroscopy (EDX) detector, which can provide semi-quantitative elemental composition for most elements. EDX confirmed that the deposit free features in Figure 10 and Figure 9 consisted primarily of exposed zirconium. The deposit regions consisted primarily of iron or iron activation products, confirming that the deposit layer was a form of iron oxide.

2.5 Deposit Removal and Activation Analysis

Neutron activation analysis (referred to as activation analysis) is a technique used to characterize neutron activated elements based on their radio-isotopic information. The elemental composition and gamma spectrum are measured and used to determine the elemental composition prior to activation. In addition, the results can be combined with neutron flux and radioactive decay data to estimate the timing of the deposit's activation products.

Activation analysis was performed to determine the timing of the iron oxide deposition on the Unit 4 and Unit 1 elements for comparison with station operating data.

The first task was to provide a suitable sample for gamma spectrometry and measure the radioisotope content of the activation products. The challenge was to focus the gamma spectrometer on the activated deposits, while not flooding the gamma detector with unwanted fission products and other radioisotopes. To achieve this, deposit material was separated from the elements by scraping the deposit into a sample container.

Two scraped deposit samples (from Unit 1 and Unit 4 elements) were transferred to CNL's radioisotope laboratory, where they were weighed and dissolved. The radioisotope inventory was determined using a calibrated gamma spectrometer. The elemental composition of the samples was determined using Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Both samples contained activated Zr-95 and UO_2/fission product contamination, which were expected given the method used for sample preparation. However, sufficient ^{54}Mn (^{54}Fe activation product) was detected to perform the deposition timing calculations.

The final task was to perform an activation and deposition timing calculation, which was performed by Candu Energy using the Monte Carlo N-Particle (MCNP) neutron transport [6] and ORIGEN-S depletion and isotope decay codes [7]. Deposition timing was modeled by combining activation measurements with power history information provided by Bruce Power. The Unit 4 activation analysis results suggested that the iron oxide deposition occurred while the bundle resided in the coolant outlet position (final position prior to discharge). The Unit 1 activation analysis results suggested that the iron deposition was associated with hot-conditioning.

3. Conclusions

Four elements from Bruce A Units 1 and 4 were examined at CNL to characterize deposits observed on the fuel cladding. Thin (<20 µm thickness) iron based deposits were discovered on the cladding of all the elements. The deposits appeared to be non-continuous porous patches and were easily removed from routine handling in a hot cell. The ZrO_2 layer beneath the deposits were typical of normally operated fuel and the cladding and UO_2 fuel appeared unaffected by the deposits. The cladding hermitic integrity for all four elements were confirmed intact. Activation analysis was consistent with iron oxide deposition on the Unit 4 bundles while they resided in the coolant outlet position (final position prior to discharge). The PIE results confirmed that the Unit 1 deposits were consistent with hot-conditioning iron-bearing deposits.

4. References

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[2] IAEA, "Review of Fuel Failure in Water Cooled Reactors", ISBN 978-92-0-102610-1, 2010 June.

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[6] "MCNP5: Monte Carlo N–Particle Transport Code Version 1.40.", Available from the Radiation Safety Information Computational Center under collection number CCC00730.

[7] "SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluations", ORNL/TM-2005/39, Version 6. Available from the Radiation Safety Information Computational Center under collection number CCC00750.