High Resolution Neutron Imaging of Spent Fuel Cladding Sections

Robert Zubler[1], Pavel Trtik[2], Johannes Bertsch[1] Liliana I. Duarte[1]

[1]Paul Scherrer Institut, Laboratory for Nuclear Materials, Nuclear Energy and Safety,
5232 Villigen PSI, Switzerland

[2]Paul Scherrer Institut, Laboratory for Neutron Scattering and Imaging, Research with Neutron and Muons,
5232 Villigen PSI, Switzerland

Corresponding author: Robert Zubler < robert.zubler@psi.ch>

1. Introduction

At the Paul Scherrer Institute (PSI) in Switzerland the spallation neutron source, SINQ, is operated mainly for research with thermal and cold neutrons.

At SINQ a neutron microscope is installed, with one of the highest spatial resolutions for neutron imaging world-wide and being the only instrument of this performance with regular access for the scientific community [1]. The set-up of the high performance neutron-sensitive scintillator was developed in house [2]; the nominal pixel size of acquired images can be routinely as low as 1.3 µm.

Neutron imaging has become an important tool to investigate hydrogen distribution and quantification in zirconium alloy components. While zirconium has a very high neutron transmission, hydrogen strongly scatters neutrons. Thus, local hydrogen concentrations can experimentally be identified and quantified [3, 4]. In the present work, besides hydrogen distributions for Zry-4 tube samples without and with a liner (so-called duplex cladding), the hydrogen distribution profiles and quantification also in spent fuel cladding sections have been revealed along the radial direction.

The hydrogen and hydrides quantification in spent fuel claddings is crucial to evaluate the potential risk for their integrity. In this contribution, we show that high resolution neutron imaging promises to reveal hydrogen concentrations also in irradiated claddings. In the case of testing active samples, questions arise concerning safety, the sample handling, radiation protection as well as the interdependencies between sample transfer cask, shielding, and beam line set-up and detector geometry.

2. Preparation of beam campaign and samples

In November 2018, the first campaign with active cladding sections was performed at the PSI SINQ facilities, using the high-resolution neutron microscope of the neutron-imaging group (NIAG). The combination of radioactive samples and the high-resolution neutron imaging was a world premiere.

The chemical composition of the tested Zry-4 and DX-D4 (duplex, with liner) samples and their dose rates are listed in Table 1. The samples stem from fuel rods irradiated in the Swiss pressurized water reactor of Gösgen, KKG. Both cladding sections have seen 5 reactor cycles, and have an average burn up of about 60 MWd/kgU and 70 MWd/kgU for Zry-4 and DX-D4, respectively.

Table 1. Chemical composition of the samples, dose rates and contamination

<table>
<tr><td colspan="2">Composition [wt%]</td><td>Sn</td><td>Fe</td><td>Cr</td><td>Ni</td><td>Zr</td><td>dose rate [μSv/h]</td><td>smear test</td></tr>
<tr><td colspan="2">Zry-4</td><td>1.2-1.7</td><td>0.18-0.24</td><td>0.07-0.13</td><td>-</td><td rowspan="3">Bal.</td><td>1’500 [a)]
95 [b)]</td><td>60 RW [c)]
< 1 RW [d)]</td></tr>
<tr><td rowspan="2">DX-D4</td><td>Liner</td><td>0.50</td><td>0.50</td><td>0.20</td><td>0.030</td><td rowspan="2">10’000 [a)]
830 [b)]</td><td rowspan="2">5 RW [c)]
< 1 RW [d)]</td></tr>
<tr><td>substrate</td><td>1.50</td><td>0.21</td><td>0.10</td><td>0.007</td></tr>
</table>

[a)] loaded Active Box in contact, [b)] loaded Active Box at 10 cm distance, [c)] sample contamination, [d)] Active Box contamination (outside)
RW = “Richtwert”, guidance value from Swiss radiation protection regulation

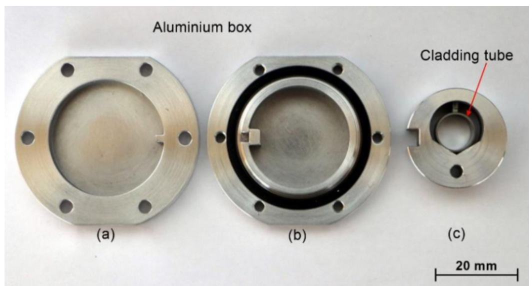

Figure 1. Aluminum Active Box with a diameter of 45 mm;
(a) backside, (b) front side and (c) inside sample holder

Before starting the measurement campaign at SINQ, several issues had to be addressed:

- The sample needs to be enclosed safely, unmovable, with correct position (sample axis in beam direction, sample cutting plane parallel to the detector). A sample positioning reference outside the box is also necessary.
- Flexibility to house various sample diameters.
- The sample holder should disturb the measurements as little as possible. Specific materials are required with thicknesses low enough for a good neutron transmission while sustaining sufficient stability.
- There must be no risk of contamination; the sample box needs to be air tight.
- Sample positioning into the box should be possible with manipulators or long tweezers.
- The holder system or box needs to be easily transportable and the handling with tweezers should be easy.
- The holder needs to fit the settings at the beam line (xyz stage and geometric conditions in front of the detector).

Figure 1, shows the disassembled sample holder system. Aluminum was the material of choice due to its high neutron transparency. The V-like cut-out at the place where the sample is positioned offers the possibility to measure samples with different diameters. The local dimensions and position of the experimental stage and detector had to be taken into account, too. A prototype was designed, manufactured and successfully tested using PSI neutron microscope beforehand with inactive samples.

3. Campaign and results

The two irradiated and defueled samples, Zry-4 and DX-D4, were cut and cleaned in the PSI hot laboratory. The axial section length is 4.5 mm. The sample mounting in new aluminum boxes was performed in a hood with respective safety precautions (shielding, wearing P3 mask). After assembly the dose rates of the Active Boxes were measured, see Table 1. The outside of the boxes was contamination free. The dose rate in 10 cm distance was between 100 and 1'000 µSv. Thus, handling with long tweezers and a special pincer was possible. For the transfer from the hot laboratory to the SINQ, the two boxes with samples were put into a lead container which was then inserted into a steel drum. The transfer was performed by PSI internal services under radioprotection supervision. The package was brought to the SINQ beam line. Unboxing as well as the mounting of the Active Box onto the experimental stage (see Figure 2) was also performed under the supervision of a radiation protection officer.

All measurements were performed during the campaign 19-24 November, 2018. The measurements (image acquisition), the sample positioning were remotely controlled from a computer outside the bunker. The results of imaging with the Active Box (radioactive samples) and without (non-radioactive samples) were evaluated; the image quality is comparably good for both cases. Figure 3 shows the irradiated DX-D4 sample inside the Active Box. The fixing aluminum screw is faintly visible. Using the Active Box brings a loss of beam intensity of about 1.2 %, keeping in mind that the aluminum beam window in the middle of the box is thinner than the outer walls. The signal to noise ratio remains practically the same. The distance between sample and detector is slightly higher with the Active Box, but the influence of the Active Box on the spatial resolution is limited (as can be seen at the good edge profile in the middle part of the images).

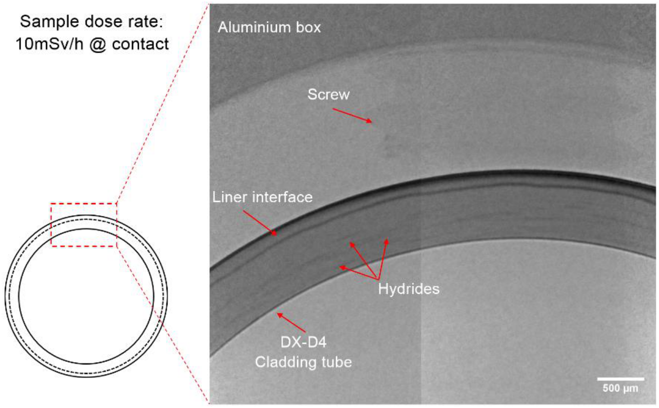

Figure 2. Active box positioned on the sample stage of the PSI neutron microscope in front of the detector's scintillator screen

Figure 3. Neutron image of an irradiated DX-D4 tube section

We could find hydrogen bands in active and inactive samples and their distribution in inactive samples change with different heat treatments [4]. A liner has a clear effect on the hydrides distribution.

After neutron imaging, the irradiated samples were transferred back to the hot laboratory for further investigations, such as EPMA and metallography. During 2019 the SINQ is closed for an upgrade. Further experiments with various irradiated samples and improvements of the test box handling are planned thereafter.

4. Acknowledgment

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