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Idaho National Laboratory

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Atom Probe characterization of neutron irradiated commercial ZIRLO® and AXIOM X2® alloys (#2986)

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As part of the Mechanistic Understanding of ZIrconium Corrosion (MUZIC) program, this team has been instrumental to obtain EPRI and Westinghouse sponsorships to prepare the neutron irradiated ZIRLO® and X2® from Vogtle nuclear power plant and ship them from Studsvik's (Sweden) hot cells to Idaho National Laboratory (INL) to be part of the NSUF library. The uniqueness of these sets of neutron irradiated alloys is that they are at the same two extremes of the fuel cycles, allowing us to study the microchemistry evolution as a function of irradiation doses, exposure time and solute content.
Advanced commercial Zirconium-Niobium (Zr-Nb) alloys, such as ZIRLO® and AXIOM®, have been developed by Westinghouse to enhance the corrosion resistance of fuel cladding material for longer service time. This study specifically aims at investigating the neutron irradiation induced Nb redistribution in AXIOM® X2® to understand the effect of neutron irradiation induced microchemistry changes and the irradiation enhanced corrosion resistance observed in ZrNb alloys. The detailed sample information on chemical composition and irradiation is given in shown in Table 1a and 1b respectively. To study the neutron IIP/nanoclusters and Nb concentration in the solid solution, atom probe tomography (APT) is the primary tool to obtain reliable chemical information. APT specimens were prepared in shielded focused ion beam (FIB) at the Irradiated Materials Characterization Laboratory (IMCL) at INL followed by APT analysis at CAES facility using LEAP 4000X.

***Table 1a**: General chemical composition of X2®.*

X2	Concentration (at.%)				
	Nb	Fe	O	Si	Cr
	0.97	0.10	0.68	0.01	0.01

Table 1b: In-reactor conditions. Assume 2 dpa per 10^{25} n/m^2 (E > 1 MeV) for PWRs

Cycle	Measured fast fluences (E>1 MeV, 10^{21} n/cm^2)	Estimated dpa	Temperature (°C)	Outer oxide thickness (µm)
1	3.88	7.6	302	2.7
4	16.5	33	301	5.1

The APT analysis of X2® samples were carried out using the same methodology as used in our previous publications on proton irradiated Zr-Nb model alloys. The peaks in the mass spectrum used to represent major elemental distribution are Zr3+, Nb3+, and Fe2+ ions. For compositional quantification, the counts for Zr2+ and Nb2+ (deconvoluted from 46.5 Da peak due to the presence of ZrH2+) were also considered. Representative 3D reconstructed data from 4 cycle X2® is shown in figure 1.

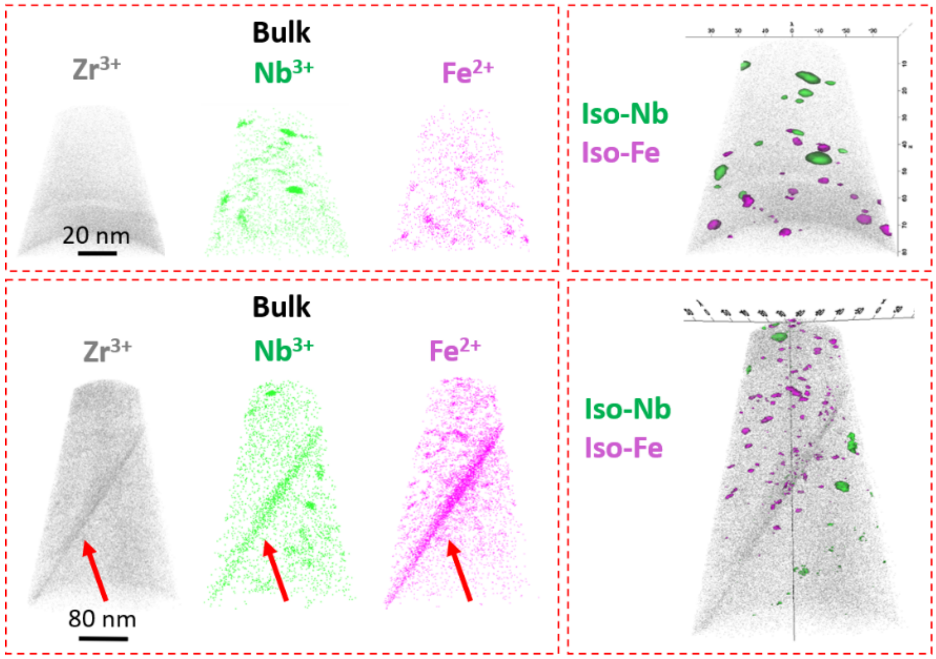

***Figure 1**: 3D reconstruction of two APT tips in the metal matrix of 4 cycles X2®. Red arrows point to the grain boundary that Nb and Fe have segregated to grain boundaries.*

Under irradiation, most of native precipitates are Nb-rich but with a much lower Nb concentration than the one predicted by the phase diagram, indicating that the native precipitates may have lost Nb to the metal matrix. Therefore, it is likely that neutron irradiation results in reduction of the Nb content in the β-Nb precipitates in X2®, an observation similar to high burnup M5®. The evolution of Fe content in the metal matrix is different than the Nb. Although Fe in the metal solid solution precipitates as Fe-rich nanoclusters and segregates to the <c> dislocation loops and grain boundaries, the Fe concentration at high-burnup is still significantly higher than the maximum solubility of Fe in the unirradiated α-Zr. The Fe enrichment in the metal solid solution can be attributed to the irradiation induced dissolution of Fe from the Fe-rich native precipitates. The evolution of precipitates in the metal matrix of in-pile X2® do not show much difference compared to other neutron irradiated recrystallized Zr-Nb alloys, such as M5®. Both Nb-rich native precipitates and irradiation-induced platelets (IIPs) were found in the metal matrix. Their composition tend to reach an equilibrium state of about 40 at.% Nb. The Nb-rich IIPs/nanoclusters density and sizes in X2® follow the same trend reported in literature.

The major hypothesis validated in this study is that the reduced in-reactor corrosion kinetics of Zr-Nb alloys is due to the reduction of Nb content in the Zr solid solution by the precipitation of Nb-rich IIPs/nanoclusters.

Publications from this work:

[1] Nanoscale redistribution of alloying elements in high-burnup AXIOM-2 (X2®) and their effects on in-reactor corrosion
Z Yu, M Bachhav, F Teng, L He, A Couet, Corrosion Science 190, 109652
[2] STEM/EDS and APT study on the microstructure and microchemistry of neutron irradiated ZIRLOTM
Z Yu, M Bachhav, F Teng, L He, M Dubey, A Couet, Journal of Nuclear Materials 573, 154139