

IN-SITU MEASUREMENT OF HYDRIDE CORROSION OF URANIUM USING X-RAY AND NEUTRON SCATTERING TECHNIQUES

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Hydride formation is a well-documented corrosion phenomena for uranium and the thermodynamics of the uranium-hydrogen system are well understood. Many unanswered questions remain regarding the micro-scale, temperature-dependent, surface (or interface)-dominated kinetics of uranium hydride (UH_3) nucleation and growth. Characterization of hydrides in uranium using conventional techniques has historically been nearly impossible for precipitates below a certain size threshold and, similarly, characterization of hydrogen effects on the metal lattice has been challenging due to the high density (19.1 g/cc) and low crystallographic symmetry (orthorhombic) of α -uranium. However, recent experimental campaigns have leveraged the unprecedented spatial resolution available at high energy x-ray beamlines and the penetration depth attainable with neutron scattering to elucidate processing-structure-property links related to hydride corrosion of uranium. For these experimental campaigns, specimens with tailored microstructures of interest were manufactured by combining appropriate casting, deformation, and heat treatment schedules. Small diameter ($\sim 500 \mu\text{m}$) specimens were produced for x-ray scattering experiments conducted at the 1-ID-E beamline of the Advanced Photon Source (APS); these specimens were exposed to high purity hydrogen gas, targeting a hydrogen:metal ratio of 0.05-0.1, prior to insertion in the beamline. X-ray diffraction successfully quantified hydride and metal phase fraction, metal lattice parameters, and strain gradients within metal grains while computed tomographic imaging enabled visualization of UH_3 precipitates within the primarily α -uranium metal specimen. The data reveal that both microstructural characteristics and macrostructural features (e.g. manufacturing-induced defects like scratches) influence hydride formation propensity. Complementing the high spatial resolution of the APS experiments, neutron scattering experiments using larger diameter specimens ($\sim 6 \text{ mm}$) coupled with in-situ hydrogen exposure of uranium via a bespoke high accuracy gas dosing system, enable direct quantification of uranium hydriding kinetics as a function of uranium metal processing schedules and gas characteristics (e.g. pressure, temperature). Time resolved neutron diffraction measurements at the Spectrometer for Materials Research at Temperature and Stress (SMARTS) at the Los Alamos Neutron Science Center directly quantifies phase fraction, establishing a direct link between the quantity of hydrogen gas absorbed by the uranium metal and the quantity of UH_3 formed. Additionally, the measurements reveal that elastic lattice strains evolve as hydrogen uptake in uranium proceeds; the strains are crystallographic direction-dependent, which suggests that hydrogen may preferentially diffuse in certain directions, and the magnitude of the lattice strain increases after the $\text{U} \rightarrow \text{UH}_3$ reaction initiates, concomitant with the strain imposed on the metal lattice by the 70% volume expansion associated with UH_3 formation. These experiments have both highlighted the efficacy of novel characterization methods for hydrogen metal interactions and are helping to answer long-standing questions regarding the metallurgical effects of hydrogen uptake in uranium.

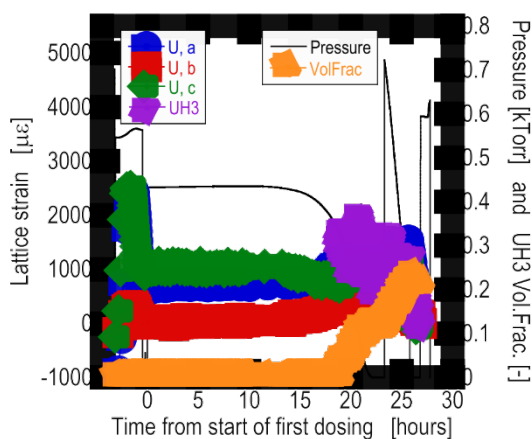
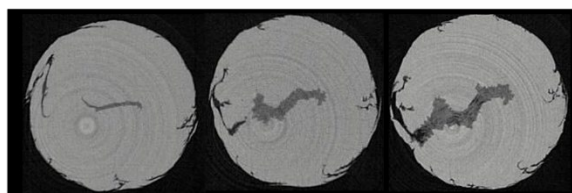


Figure 1. X-ray CT images (2D slices) of UH_3 inside uranium metal sample (top); elastic lattice strains for α -uranium and UH_3 phases, and gas pressure data as a function of exposure time collected on SMARTS (bottom).