In Situ Synchrotron Radiation Diffraction Study of Neutron-Irradiated Yttrium Hydrides during Thermal Treatments

M. Nedim Cinbiz*, Mehmet Topsakal†, Annabelle Le Coq‡, Kory Linton‡

*Idaho National Laboratory, 1955 Fremont Avenue, Idaho Falls, ID 83415, mahmut.cinbiz@inl.gov
†Brookhaven National Laboratory, 33 North Renaissance Street, Upton, NY 11973, mtopsakal@bnl.gov
‡Oak Ridge National Laboratory, 1 Bethel Valley Road, Oak Ridge, TN 37830, lecoqag@onrl.gov
‡Oak Ridge National Laboratory, 1 Bethel Valley Road, Oak Ridge, TN 37830, lintonkd@ornl.gov

INTRODUCTION

Metal or alloy hydrides are being considered as solid-state moderators in advanced low-power high-temperature mobile reactor concepts such as microreactors. These moderators significantly reduce the required reactor size, thereby increasing reactor portability. During high temperature operation, hydrogen retention within the moderator is critical. Because hydrogen has high mobility in the metal hydride at elevated temperatures, it can diffuse inside the moderator under temperature gradients and may leave the hydride [1], or it can be trapped by irradiation-induced defects. Therefore, the moderator must maintain its geometrical stability to avoid transients. Because parent materials form hydrides [2], hydrides and/or hydrogen can be monitored via x-ray [3] or neutron scattering [4] techniques.

In this study, heating-cooling experiments were performed on fresh and neutron-irradiated yttrium hydride moderators ($YH_{1.84}$) in-situ with high-energy x-rays in transmission mode, using a 2D x-ray detector. The high peak resolving power of the synchrotron x-rays allowed for a detailed study of the hydrogen's behavior in the hydride moderator. Results were interpretated based on hydrogen's precipitation and dissolution behavior, including its transport behavior in the moderator.

MATERIALS AND METHODS

This study used yttrium hydrides (YH_x) fabricated via massive hydriding at Oak Ridge National Laboratory (ORNL). The pedigree of the initial yttrium material and hydriding approach are described in the literature [5]. The stoichiometry x for these YH_x specimens was evaluated as 1.84.

Specimens were subjected to neutron irradiation at 608 and 862°C in ORNL's High Flux Isotope Reactor (HFIR). The approximate neutron displacement damage was 0.2 dpa.

Because yttrium hydride is brittle, samples were broken into pieces that measured approximately 0.5 × 0.5 × 0.5 mm and were double-encapsulated using quartz capillaries as shown in Fig. 1. Another quartz capillary was placed to keep the sample in place. For comparison, a fresh $YH_{1.84}$ specimen and a blank capillary were also tested.

Synchrotron x-ray diffraction measurements were performed at the 28-ID-2-D beamline end station at the National Synchrotron Light Source-II (NSLS-II) at Brookhaven National Laboratory. The energy of the incident x-rays was 67.1 keV with a beam size of 0.5 mm wide by 0.3 mm high. Cerium oxide powder was used as a calibrant to refine the sample-to-detector distance and the tilt of the detector with respect to the x-ray beam. The sample-to-detector distance was 0.793 m. The integration process was conducted using the pyFAI package (https://github.com/silx-kit/pyFAI)[6]. Detector acquisition time was set to 0.1 s.

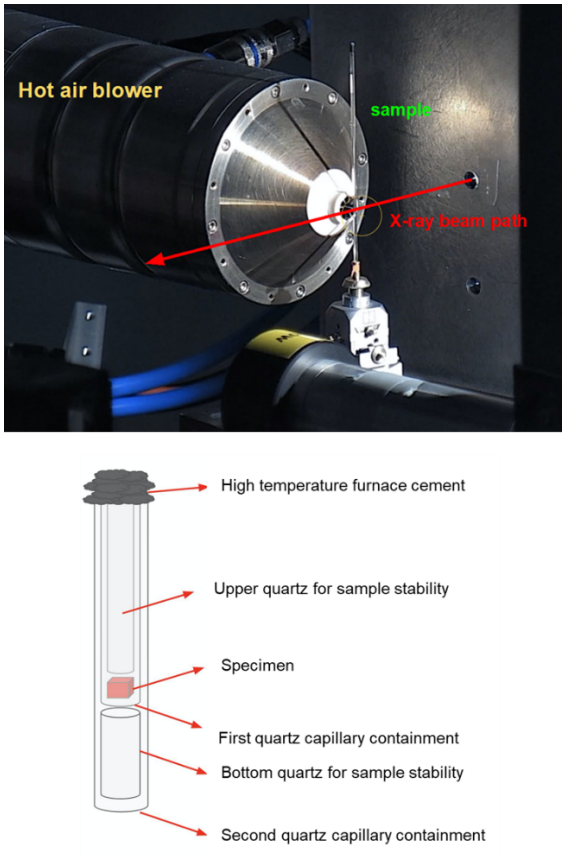

Fig. 1. Experimental setup (up) and specimen placement in quartz capillaries (down).

Neutron-irradiated and fresh specimens were subjected to two-cycle heat treatment (see Fig. 2). The specimens were heated to 600°C at a rate of 10°C/min. Then the samples were soaked at 600°C for 10 min and cooled to 200°C. Specimens were soaked for 10 min. at 200°C. The heat treatment was applied a second time, followed by a cooldown to 25°C.

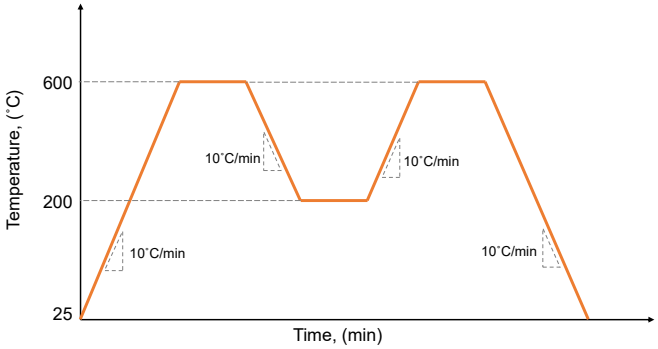

Fig. 2. Schematic of the heating-cooling treatment of neutron-irradiated and fresh samples.

RESULTS

Fig. 3 shows the integrated x-ray diffraction profile of the $YH_{1.84}$ moderator during (a) heating and (b) cooling. The high-resolution diffraction profile indicates that the moderator consisted of minor yttrium (Y) metal phase and yttrium oxides and precipitates (not indicated on the diffraction profile). During heating, all the peaks shifted to higher d-spacing values, illustrating the thermal expansion behavior. As predicted by the Y-H phase diagram, the yttrium phase disappears when temperatures rise above 400°C. The yttrium phase reappears when the temperature falls below 400°C. However, the maximum peak intensity of yttrium decreased compared with that of the pristine uncycled material. By contrast, the hydride maximum peak intensity increased. Therefore, some portion of yttrium metal precipitated as hydride. This behavior might indicate that excess hydrogen bonded to the moderator, or pre-existing hydrogen trapped in the pristine material was used to form YH_x. The protective yttrium oxide–formed outer surface was expected to prevent environmental hydrogen diffusion into the moderator. Thus, hydrogen transport from the quartz to sample was not considered.

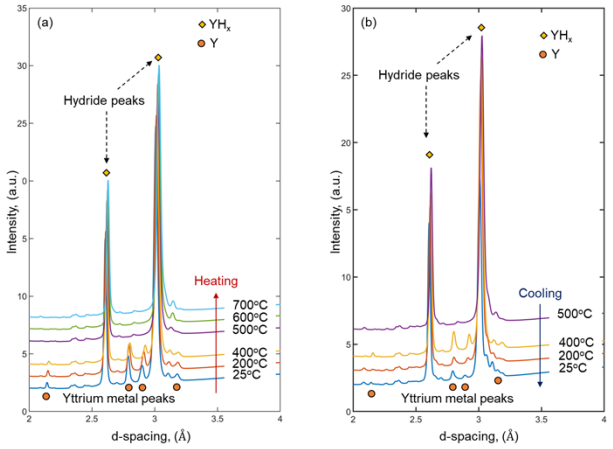

Fig. 3. Evolution of x-ray diffraction peaks of fresh $YH_{1.84}$ moderator during (a) heating and (b) cooling.

Fig. 4(a) shows the evolution of the x-ray diffraction profile obtained from the neutron irradiated $YH_{1.84}$ moderator at specific temperatures during the heating stage. As with the fresh sample, the yttrium phase disappeared during heating, but this occurred at a much lower temperature (200°C). This significant temperature difference suggests that incoming fast neutrons might dislodge hydrogen from its lattice position and knock-out from the moderator. Thus, neutrons may facilitate hydrogen removal from the moderator. Furthermore, the yttrium phase disappeared after the heat treatment, as shown in Fig. 4(b). This behavior indicates that there could also be some irradiation damage recovery effects that enabled excess hydrogen that was trapped in the radiation-induced defects to precipitate or reprecipitate as hydrides.

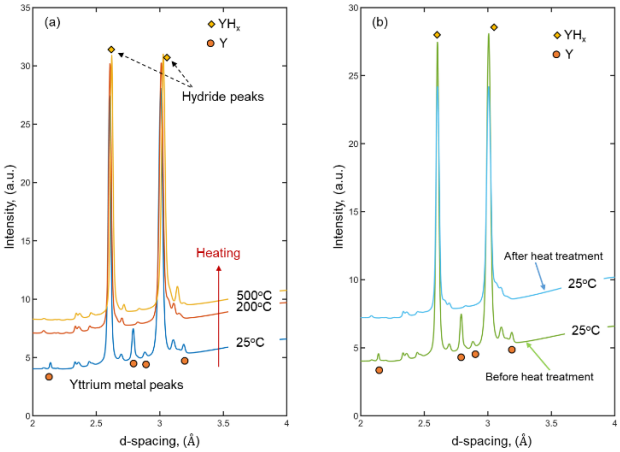

Fig. 4. (a) Evolution of x-ray diffraction peaks of the neutron irradiated $YH_{1.84}$ moderator, and (b) room temperature x-ray diffraction profile showing the disappearance of yttrium peaks after heat treatment.

SUMMARY

High resolving power of in situ synchrotron radiation diffraction revealed unique observations of an irradiated bulk YH_x neutron moderator for advanced reactors. Preliminary data analysis indicated that neutron damage may cause hydrogen to be removed from defects or outside of the moderator, and it also may cause some portion of the hydrogen to be regained as hydride via heat treatments. However, more data analysis and additional synchrotron x-ray diffraction experiments are needed to elucidate hydrogen's behavior in metal hydride moderators.

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