In-situ observation of irradiated-induced amorphization in zirconium suboxides in nuclear fuel cladding alloys

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Zirconium alloys are used extensively as fuel cladding materials in modern light water nuclear reactors. The corrosion and irradiation mechanisms have been widely studied, and a recent suggestion is that the ZrO suboxide that forms part of the inner protective oxide is rate limiting in the transport processes involved in oxidation [1, 2]. Because of the key role that this suboxide layer plays in the corrosion kinetics, it is important to understand the processes by which the suboxide may be damaged by irradiation in service. We report for the first time the direct observation of irradiation-induced amorphization of the zirconium suboxide layer at the interface between bulk oxide and the underlying metal substrate. In-situ irradiation in a transmission electron microscope (TEM) enables the dynamical observation of the damage processes, and high-resolution TEM results (shown in Figure1) clearly reveal amorphization of the ZrO suboxide under heavy ion irradiation at cryogenic temperatures, while both the bulk monoclinic-ZrO_2 and Zr matrix remain crystalline. These results suggest that the protective interface suboxide is particularly susceptible to radiation damage, which may help explaining the acceleration of corrosion rate of zirconium alloys observed in-service in nuclear reactors. This irradiation-induced amorphization behaviour is discussed in relation to the ordered arrangement of interstitials and the formation of superlattices.

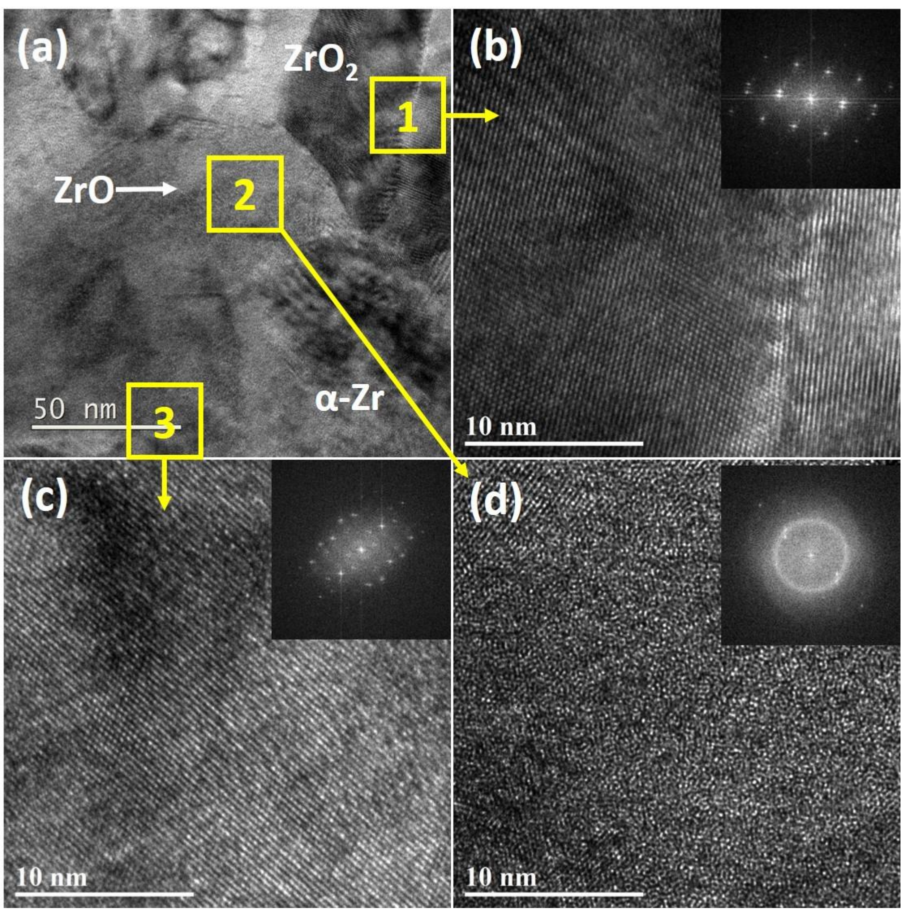

Figure 1. Post-irradiation characterization, (a) Low magnification bright field TEM image showing the monoclinic-ZrO_2, ZrO suboxide and Zr matrix. (b) HRTEM and FFT from the region of monoclinic-ZrO_2 identified in (a). (c and d) HRTEM and FFT from identified regions of Zr matrix and ZrO suboxide, respectively.

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References

1. Hu, J., et al., Understanding Corrosion and Hydrogen Pickup of Zirconium Fuel Cladding Alloys: the Role of Oxide Microstructure, Porosity, Suboxides and Second-Phase Particles, Zirconium in in the Nuclear Industry: 18th International Symposium. 2018. ASTM International.
2. Göhr, H., et al. Long-term in situ corrosion investigation of Zr alloys in simulated PWR environment by electrochemical measurements, Zirconium in in the Nuclear Industry: 11th International Symposium. 1996. ASTM International.