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Corrosion Protection and Failure Mechanism of ZrO_2 Coating on Zirconium Alloy Zry-4 under Varied LiOH Concentrations in Lithiated Water at 360°C and 18.5 MPa

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Abstract : After the Fukushima-Daiichi accident, the development of accident tolerant fuel cladding materials to improve reactor safety has become a hot topic in the field of nuclear industry. ZrO_2 has a satisfactory neutron economy and can guarantee the fission chain reaction process, which enables it to be a promising coating for zirconium alloy cladding. Maintaining a good corrosion resistance in primary coolant loop during normal operations of Pressurized Water Reactors is a prerequisite for ZrO_2 as a protective coating on zirconium alloy cladding. Research on the corrosion performance of ZrO_2 coating in nuclear water chemistry is relatively scarce, and existing reports failed to provide an in-depth explanation for the failure causes of ZrO_2 coating. Herein, a detailed corrosion process of ZrO_2 coating in lithiated water at 360 °C and 18.5 MPa was proposed based on experimental research and molecular dynamics simulation. Lithiated water with different LiOH solutions in the present work was deaerated and had a dissolved oxygen concentration of < 10 ppb. The concentration of Li (as LiOH) was determined to be 2.3 ppm, 70 ppm, and 500 ppm, respectively. Corrosion tests were conducted in a static autoclave. Modeling and corresponding calculations were operated on Materials Studio software. The calculation of adsorption energy and dynamics parameters were undertaken by the Energy task and Dynamics task of the Forcite module, respectively. The protective effect and failure mechanism of ZrO_2 coating on Zry-4 under varied LiOH concentrations was further revealed by comparison with the coating corrosion performance in pure water (namely 0 ppm Li). ZrO_2 coating provided a favorable corrosion protection with the occurrence of localized corrosion at low LiOH concentrations. Factors influencing corrosion resistance mainly include pitting corrosion extension, enhanced Li+ permeation, short-circuit diffusion of O^{2-} and ZrO_2 phase transformation. In highly-concentrated LiOH solutions, intergranular corrosion, internal oxidation, and perforation resulted in coating failure. Zr ions were released to coating surface to form flocculent ZrO_2 and ZrO_2 clusters due to the strong diffusion and dissolution tendency of α-Zr in the Zry-4 substrate. Considering that primary water of Pressurized Water Reactors usually includes 2.3 ppm Li, the stability of ZrO_2 make itself a candidate fuel cladding coating material. Under unfavorable conditions with high Li concentrations, more boric acid should be added to alleviate caustic corrosion of ZrO_2 coating once it is used. This work can provide some references to understand the service behavior of nuclear coatings under variable water chemistry conditions and promote the in-pile application of ZrO_2 coating.

Keywords : ZrO_2 coating, Zry-4, corrosion behavior, failure mechanism, LiOH concentration
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