

Coatings for safe long term wet storage of spent Al-clad research reactor fuels

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Spent aluminum-clad fuels from research reactors (RRs) are stored for decades in water filled pools. Pitting corrosion of these fuels has been reported despite water quality management programs at the storage sites, and attributed to synergism in the effect of specific basin water parameters on corrosion of aluminum alloys. Corrosion protection of the spent RR fuel in wet storage is therefore considered to be important. Coatings are widely used to protect metallic materials in many industries. The shape of the RR fuel and radioactivity of spent fuels especially preclude coatings formed electrochemically. Hence, chemical surface treatment to form a protective coating is the only option and in this context conversion coatings were considered. The results of preliminary laboratory and field investigations carried out at IPEN in Brazil revealed that chemically formed rare earth element containing oxide coatings increased the corrosion resistance of Al alloys. These investigations were extended to include boehmite (a form of hydrated aluminium oxide) and hydrotalcite (HTC) coatings, which is lithium aluminium-nitrate-hydroxide hydrate that forms on Al alloys immersed in an appropriate alkaline lithium salt solution.

The objective of this investigation was to: (a) prepare and characterize hydrotalcite coatings from baths at different temperatures followed by post-coating treatments such as cerium oxide incorporation and sealing; (b) determine from laboratory tests the corrosion behavior of HTC coated AA 6061 alloy in NaCl; (c) evaluate the extent of corrosion protection offered by the coatings from field studies in which uncoated and coated AA 6061 alloy coupons and 'dummy' fuel elements were exposed to the IEA-R1 reactor spent fuel basin for periods of up to 24 months.

The HTC coating preparation procedure consisted of immersion of AA 6061 alloy specimens for the laboratory tests and coupons as well as full-size plates for the field tests in solutions to: (a) degrease; (b) deoxidize; (c) form the HTC coating; (d) incorporate cerium oxide in the coating; (e) seal the coating.

The HTC coating consisted of intersecting blade-like crystallites and that formed at 98 °C was significantly thicker compared to the coating formed at room temperature (RT). The corrosion behaviour of uncoated and HTC coated specimens was determined from anodic potentiodynamic polarization measurements in 0.01 M NaCl. These measurements indicated marked increase in the corrosion resistance of the HTC coated specimens. Further, the effects of duration of treatment in the coating solution and the cerium solution were also determined. The field tests consisted of: (a) preparing uncoated and coated coupons and plates; (b) stacking of the coupons in racks and assembling of the plates to form full-size 'dummy' fuel elements; (d) immersion of the racks and the 'dummy' fuel elements in the spent fuel section of the IEA-R1 reactor in IPEN, Brazil, for periods of up to 24 months; (e) removal of the racks or 'dummy' fuel elements, rinsing and decontamination; (f) dis-assembly and examination of the coupons and plates.

The room temperature or RT-HTC coated coupons revealed a few pits after 5 months of exposure whereas the HT-HTC coated coupons exposed for the same duration did not reveal any pits. On the basis of these observations the full-size plates were coated with only HT-HTC and not RT-HTC. The HT-HTC coated plates from the 'dummy' fuel elements exposed for 14 months were stained but revealed no pits.

The cerium incorporated HTC imparted much higher corrosion protection. Sealing of the coatings further improved the pitting corrosion resistance of the alloy. The HTC layer imparts pitting corrosion protection by acting as a physical barrier between the solution and the surface. The mechanism by which the cerium in the HTC imparts protection is considered to be 'active corrosion protection'. According to this mechanism, the lower solubility of $\text{CeO}_2 \cdot 2\text{H}_2\text{O}$ allows formation of Ce^{4+} ions in solution which then diffuse to defects in the coating and exposed bare metal. When in contact with the bare metal, these ions reduce to Ce^{3+} and precipitate as $\text{Ce}(\text{OH})_3$ and thus seal the defect in the coating..

The coating procedure for irradiated Al-clad spent fuels would be facilitated if all the treatments were to be carried out at room temperature. Currently studies are in progress to pre-treat, coat and post-treat the Al alloys to obtain highly protective HTC coatings at room temperature. Details of these studies and the extent of corrosion protection offered by this coating will be presented and discussed.

Organization

Country

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