326

EFFECT OF COUPON ORIENTATION ON CORROSION BEHAVIOR OF ALUMINIUM ALLOY COUPONS IN THE SPENT FUEL STORAGE SECTION OF THE IEA-R1 RESEARCH REACTOR

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Abstract. Surveillance programmes to monitor the corrosion of aluminium clad spent research reactor fuels have used test racks containing horizontal metallic coupons. Spent MTR type fuel elements are usually stored vertically, with its fuel plates, also vertical. Hence, the influence of coupon orientation on the corrosion behaviour of aluminium alloy coupons exposed to the spent fuel storage section of the IEA-R1 research reactor in São Paulo, Brazil has been studied. Circular coupons of aluminium alloys AA 1050 and AA 6061, oriented both vertically and horizontally, were exposed to the storage section water for a year. Individual and coupled coupons were exposed to simulate general, crevice and galvanic corrosion. The storage section water parameters were periodically measured. Pitting was the main form of corrosion and coupon orientation had a marked effect on the extent of pitting. Vertically oriented coupons pitted less than horizontally oriented coupons.

Introduction

In Latin America, many research reactors (RRs) have been in operation since the late fifties, and a significant amount of spent fuel has accumulated. Most of the spent fuel elements (SFE) were returned to the USA and the Latin American countries with concerns related to spent fuel storage are Argentina, Brazil, Chile, Mexico and Peru. The concerns are based on the fact that in May of 2006, the option to send SFE to USA could cease, and National solutions in countries without nuclear power programmes will be very difficult to implement. These concerns were the driving force for the initiation of the IAEA sponsored Regional Technical Co-operation Project for Latin America. The objectives of this Project are to provide the basic conditions to define a regional strategy for managing spent fuel and to provide solutions, taking into consideration the economic and technological realities of the countries involved. In particular, to determine the basic conditions for managing RR spent fuel during operation and interim storage as well as final disposal, and to establish forms of regional co-operation for spent fuel characterization, safety, regulation and public communication.

This Project is divided into 4 subprojects: (1) spent fuel characterization; (2) safety and regulation; (3) options for spent fuel storage and disposal; (4) public information and communication. Corrosion surveillance is one of the activities of the subproject 'Spent fuel characterization'.

The dominant fuel type used in the Latin American (LA) RRs is plate-type (MTR), LEU, oxide fuel (U_3O_8-Al) clad in Al, followed by TRIGA-type (U-Zr-H) rods. Almost all the spent fuels from LA RRs are stored in racks within the reactor pool, in decay pools or in away-from-reactor wet basins. The in-reactor storage facilities consist of aluminium or stainless steel storage racks.

The main objective of the corrosion surveillance activities of the Regional Project is to evaluate the effect of Latin American spent fuel basin parameters on the corrosion behaviour of research reactor

9972

fuel cladding. This evaluation is being done by exposing racks containing aluminium and stainless steel coupons at the different spent fuel basins for different periods followed by their examination. In all corrosion monitoring programmes carried out so far, test racks containing horizontally oriented coupons have been exposed. It is well known that all MTR type fuel elements are stored vertically in spent fuel basins and the fuel plates are also vertical. Hence, the influence of coupon orientation on the corrosion behaviour of coupons exposed to the spent fuel section of the IEA-R1 research reactor in IPEN, São Paulo, Brazil is being evaluated. The tests were initiated in 2002 and one set of racks was withdrawn in June 2003 after one year of exposure. The other racks are expected to be withdrawn after two and three years of exposure. This paper presents the influence of coupon orientation on the corrosion behavior of coupons withdrawn after one year of exposure

Methods and materials

Aluminium alloys AA 1050 and AA 6061, (compositions shown in table I) are used in the manufacture of MTR type fuel plates and elements. Circular test coupons 100mm in diameter, 3 mm thick and with a central hole 35mm in diameter were prepared from these alloys under conditions similar to that used to prepare fuel plates. The state of the surface of the coupons was also similar to that of the fuel plates. One machined and polished Al 1050 coupon per rack was rinsed, degreased and passivated in water at 95° C for 24 hours. One of the surfaces of this coupon was scratched with a 0.5mm wide scriber to simulate a damaged fuel plate surface. The test rack consists of a stand on which the coupons and ceramic disks are stacked. The coupons are stacked as individual coupons or as coupled coupons. Ceramic disks (non-porous alumina) were used to separate the coupons and the coupled coupons, one from the other. The coupons were stacked in the following order, from top to bottom: Al 1050; Al 6061; Al 1050 (pre-oxidized and scratched); Al 1050 - Al 1050 (couple); Al 1050 - Al 6061(couple); Al 6061 - Al 6061(couple); Al 1050 - SS 304 (couple) and Al 6061 - SS 304 (couple).

Table 1. Nominal composition of aluminium alloys AA 1050 and AA 6061.

Alloy	Cu	Mg	Mn	Si	Fe	Ti	Zn	Cr	V	Al
AA 1050	0.05	0.05	0.05	0.25	0.05	0.03	0.05	0.05	0.05	99.5 min
AA 6061	0.15-0.40	0.08-1.20	0.15	0.40-0.80	0.7	0.15	0.25	0.04-0.35		Balance

Six racks were immersed in June-2002 in the spent fuel section of the IAE-R1 RR, of which, three were suspended with its coupons oriented horizontally and the other three racks were suspended with its coupons oriented vertically. One rack (no 44) with vertically oriented coupons and another (no 47) with horizontally oriented coupons were withdrawn after 12 months of exposure. (Figure 1) Similar withdrawals will be carried out after 2 and 3 years of exposure.

Immediately after withdrawal, the two racks were disassembled and the coupons examined and photographed. The pH of the water in the crevice of the couples was measured. After decontamination, the coupons from the two racks were examined in an optical microscope coupled to an image analysis system. The distribution of corrosion pits on the coupon surfaces was determined. Histograms of number of pits (counts) as a function of pit diameter, were plotted.

Results and discussion

Basin water parameters

Basin water parameters, such as conductivity, pH and chloride ion concentration, were monitored periodically and radiometric analysis of the water was also carried out. The conductivity was determined directly (but intermittently) by the probe in the deionizing circuit. Conductivity was maintained at < 2.0 μS/cm. the pH was always in the range of 5.5-6.5 and the chloride ion concentration was < 0.2 ppm. When the conductivity came close to 2.0μS/cm or the chloride content came close to 0.2 ppm, the deionization resins were regenerated. The water specimens for radiological analysis were collected once a week after the reactor was switched off. Gamma spectroscopy was carried out to determine the nuclides ^{140}Ba, ^{58}Co, ^{60}Co, ^{51}Cr, ^{137}Cs, ^{131}I, ^{133}I, ^{99}Mo, ^{24}Na, ^{239}Np, ^{132}Te and ^{187}W.

Observations made immediately after removal of the racks

During the disassembly of the racks it was observed that the coupled coupons were difficult to separate. Also during disassembly of the rack, the pH of the water in the crevice between the various couples was measured. In all cases, independent of coupon orientation, the pH of the water in the crevice was 5.5, one point below that of the bulk water pH (6.5).

Figure 1. Photograph of rack with vertically oriented coupons being withdrawn from the storage section of the reactor pool.

Microscopic examination of coupon surfaces.

The exposed surfaces of the two aluminium alloys revealed pits, independent of the orientation of the coupon. However, many features were specific to the alloy, the position of the coupon in the rack and the orientation of the coupon.

Horizontally oriented coupons (Rack-47)

Figures 2a and 2b reveal the histograms of pit count versus pit size on the upward facing and the downward facing surfaces of the individual AA 1050 coupon respectively. The upward facing surface revealed a large number of pits, ~90, in the size range 40-50 μm while the downwards facing surface revealed only 6-8 pits in the same size range. The shape of the pits on this coupon varied from irregular to round. (figure 3)

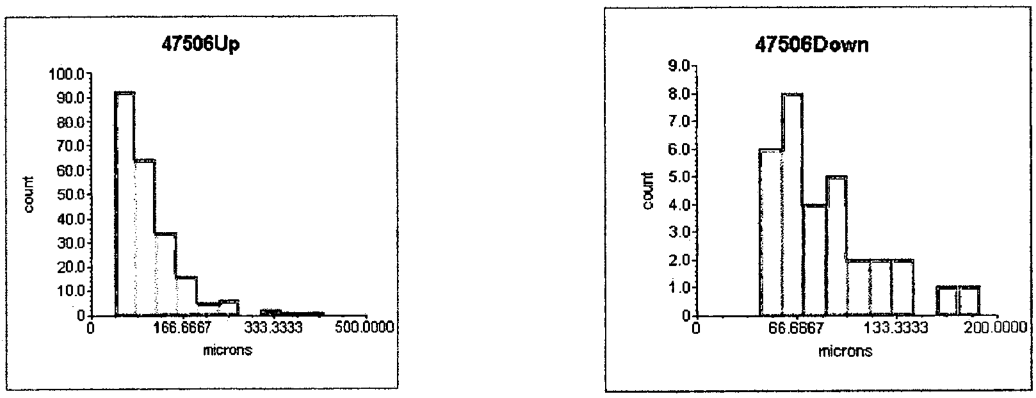

Figure 2. Histograms of pit count versus pit size on the upward facing (47506Up) and downward facing (47506Down) surfaces on horizontally oriented AA 1050 coupon.

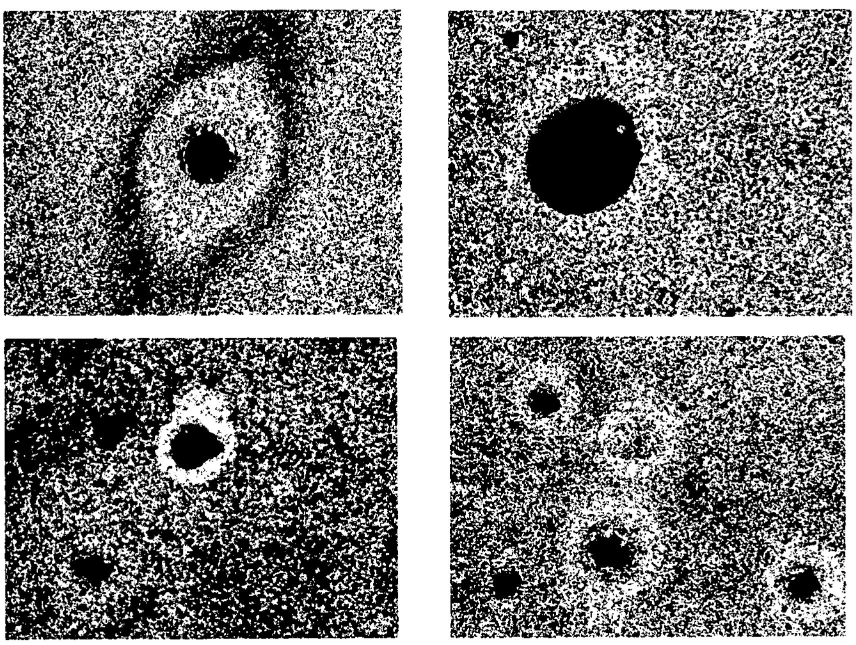

Figure 3. Micrographs showing pits and bright regions on horizontal AA 1050 surfaces.

Most of the pits revealed a bright region around the pit, characteristic of a cathode region around a localized anode region. The shape of this bright region also varied from circular to elliptical. (figure 3) On the exposed surfaces of the other AA 1050 coupons, similar round and irregular shaped pits were observed. On the AA 1050 coupon surface, in contact with the AISI 304 stainless steel coupon, large pits were observed, revealing the deleterious effect of a galvanic junction. The two surfaces of the pre-oxidized and scratched AA 1050 coupon revealed few small pits and no bright regions around the pits. No pits were observed along the scratch.

Vertically oriented coupons (Rack 44)

On the surfaces of the individual AA 1050 coupon, round and irregularly shaped pits were observed. Many pit clusters were also observed. The bright cathode areas associated with pits were shaped like a comet with a tail giving a clear indication of the top and bottom of the vertically oriented coupons. (figure 4)

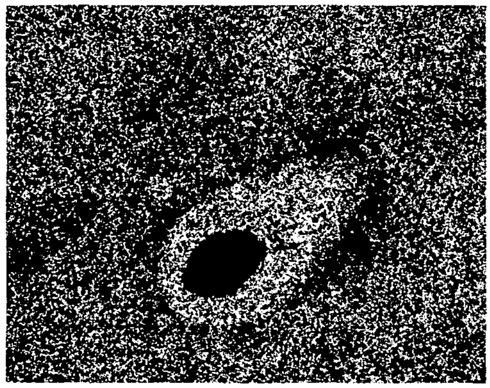

Figure 4. Optical micrograph of vertically oriented AA 1050 surface revealing comet shaped bright region around a pit.

Almost all the pit features and the comet shaped bright areas associated with the circular pits, on AA 1050 were also observed on the exposed surfaces of AA 6061 coupons. Bright regions or comet tails were not observed around irregular shaped pits indicating that the mechanism associated with the formation of round and irregular shaped pits are different. The facing surfaces of the crevice couple coupons, AA 1050-AA 1050, AA 1050-AA 6061 and AA 6061-AA 6061 were stained and did not reveal any pits. The stains on the surfaces of the two alloys were distinct and characteristic of the alloy. The surfaces of the pre-oxidized and scratched AA 1050 coupon revealed a few small pits and no bright regions around the pits. No pits were observed along the scratch.

Pitting behaviour as a function of coupon orientation

Comparison of pit histograms obtained for the horizontally oriented top surface of AA 1050 with that obtained for one of the surfaces of the same alloy oriented vertically (Figure 5) reveals that twice as many pits (size range 40-50 μm) form on the horizontal coupon as compared to that on the vertical coupon.

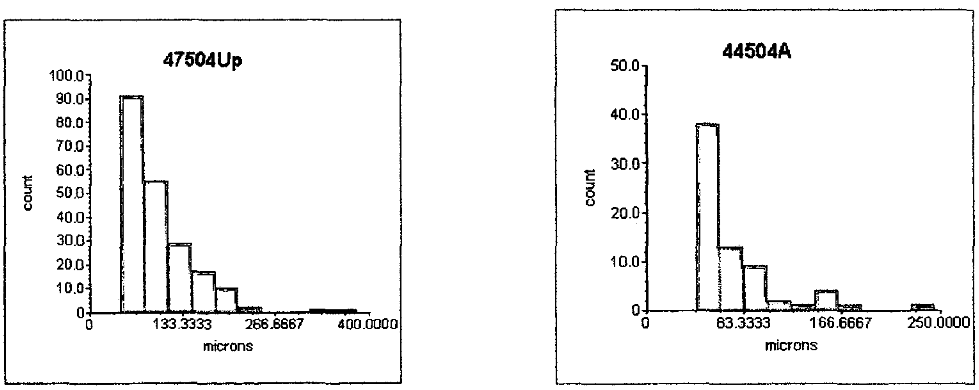

Figure 5. Histograms of pit count versus pit size on the upward facing surface (47504Up) and vertical surface (44504A) of AA 1050 coupon.

Comparison of histograms of the pitted surfaces of vertically and horizontally oriented AA 6061 alloy also revealed similar behaviour. This indicated that among the many parameters that control pit formation, such as alloy composition, metallurgical state and water parameters, settled solids contribute to pit initiation and formation.

Conclusions

1. The top surface of the horizontally oriented coupons pitted more than the bottom facing surface. The extent of pitting on the top surface of horizontal coupons decreased with change in position of the coupon from top to bottom in the rack.
2. The two sides of vertically oriented coupons of both alloys pitted to the same extent.
3. The extent of pitting of vertically oriented coupons was considerably less than that of the horizontally oriented coupons indicating that pit formation is influenced by, among other factors, settled solid particles on the coupon surface.
4. The pre-oxidized AA 1050 coupon revealed very few pits and these were less than 30 µm in diameter. Both the horizontally and vertically oriented pre-oxidized coupons pitted to a lesser extent than the corresponding un-oxidized alloy coupons
5. Coupon orientation had no noticeable effect on crevice or galvanic corrosion. The contact surfaces of AA 1050/AA1050, AA1050/AA6061 and AA 6061/AA 6061 couples and the surfaces of the aluminium alloys in contact with AISI 304 stainless steel, were stained with white to grey aluminium oxide.
6. Overall, the results of this ongoing investigation have indicated that coupon orientation has a marked effect on the corrosion behaviour of aluminium alloy coupons. Since fuel plates of spent MTR type fuel elements are usually oriented vertically, information that can be obtained from the use of vertically oriented coupons in a surveillance test would be more representative of actual corrosion processes taking place on fuel plates.