SPENT FUEL STORAGE CORROSION MANAGEMENT

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Abstract

The Corrosion Surveillance program was applied for 30 MW GA Siwabessy Multi Purpose Research Reactor on 2006. The objective of this experiment is to understand the Pitting, Crevice, Galvanic and General corrosion of the research reactor material structure, especially for primary water cooling structure materials. The experiment has been started at 2006. The first inspection was done after a year immersion on 2007. The second inspection was done after three (3) years immersion and will be reported in this paper. The coupons were analyzed by visual as well as photographic evaluation of coupon surfaces. Metallographic examination under reflected optical microscope equipment with camera was applied for investigating of pits and surface changing. The water quality is analyzed by measuring conductivity (on-line conductivitimeter), pH (on-line pH meter), Chlor (UV-Viss spectroscopy) and Total Dissolved Solids (TDS, on-line TDS meter). The pH of trapped water between two coupons was measured soon after withdrawing by inserting pH paper. From the results, it is known that the trapped water pH between the coupons is 4, one digit lower than the pool water (pH 5.4–5.5 that was measured digitally with accuracy of ±0.1). No significant differences were found for trapped water pH observed between horizontally and vertically racks, and also for the sample of a year immersion. The conductivity, pH, Chlor and TDS were analyzed as 1.3–1.7 μS/cm, 5.2–5.5, none and 0.7–0.9 μS/cm, respectively. The range permitted and measured inlet temperature for water purification is 23–34^{o}C. From the visual observation, it is known that (a) water quality of primary water is required the SAR, (b) crevice and galvanic effect are the main corrosion; (c) no pitting; (d) no biofilm observed. The experience and scientific results gained is very useful to be implemented for Indonesia Research Reactor Ageing Program and future NPP.

1. INTRODUCTION

Multi Purpose Research Reactor G.A. Siwabessy that has been built and developed at 1983, reached first the critical attainment on July 1987 and ceremonial opened on August 20, 1987. It reached the full energy of 30 MW firstly on March 1992. Therefore, it is almost 25 years old. When the U.S. Department of Energy issued a Record of Decision on Nuclear Weapons Non-proliferation Policy Concerning Foreign Research Reactor Spent Nuclear Fuel, with the purpose to recover enriched uranium exported from the U.S., a decision was taken in Indonesia to repatriate all the US origin spent fuel. BATAN organized the first shipment operation in March 1999. It consisted of 47 spent fuel elements and 1 plate of MTR fuel from RSG-GAS reactor. The second shipment operation was done in 2004 and the third and last shipment was done in June 2009, when 42 spent fuel elements from RSG-GAS returned to the U.S. Since on September 2001 Indonesia started buying Low Enriched Uranium from France and Russia, therefore there is no repatriation anymore. Then, the Indonesian regulatory body recommend transferring all the spent fuel to Interim Spent Fuel Storage Facility, together with some fuel from Radio-metallurgy Laboratory. Therefore, the reliability of Spent Fuel Storage should be addressed.

The water quality control is the important point that should be aware especially for wet storage[1]. The water in a wet storage is subjected to γ-rays, α-rays and a bit of neutron which come from the core during operation, because the wet storage is located just beside the core, separated by very thick aluminium window. Since the existencies of radiation, the radiolyses product will be generated, such as oxidator O_2 and H_2O_2 which induce oxidation reactions of corrosion process. This process may accelerate the corrosion of the spent fuel cladding and also reactor structures materials, and other undesirable effects like forming compound deposits inside the cooling system, in particular, inside the heat exchanger cores, thereby adversely affecting its performance and life-span. As such, it is imperative to monitor, control and maintain the primary water chemistry to the required specifications at all times in order to ensure longevity and performance of the reactor system.

The in-service inspection of the tank liner and other primary cooling structure materials such as heat exchanger were done by using under water camera and ultrasonic, respectively. The results show small progressive deterioration in the structure probably due to inadequate control of water chemistry or originally scratch.

Based on the above description, it is considered to develope the corrosion surveillance program for 30MW GA Siwabessy Research Reactor wet storage. The objective of this experiment is to understand the Pitting, Crevice, Galvanic and General corrosion of the research reactor material structure, especially for primary water cooling structures. The first batch of six coupon assemblies were strategically positioned in the service pool that located just beside the pool but the water quality is precisely similar to pool water, in 2006 and has been withdrawn two sets (horizontally and vertically) in stages for inspection after one (on 2007) and three years of immersing (on 2010). The first report of a year exposuring was presented on International Conference on Research Reactors: Safe Management and Effective Utilization, at Sydney, Australia, 2007 [2] and the latter one will be discussed in this paper.

The water quality is analyzed by measuring conductivity (on-line conductivitimeter), pH (on-line pH meter), Chlor (UV-Viss spectroscopy) and Total Dissolved Solids (TDS, on-line TDS meter). The pH of trapped water between two coupons was measured soon after withdrawing by inserting pH paper. The coupons were analyzed visually as well as photographic evaluation of coupon surfaces. Metallographic examination under reflected optical microscope equipment with camera for investigation of pits width and X-met for analyzing the element content.

2. METHODOLOGY

2.1. Sample preparation and immersion

The test coupons were prepared from the same grade materials as used for the constructions of the reactor structure, e.g. aluminum alloy used for fuel cladding, reactor tank and stainless steel (SS) for fuel assembly storage rack. The service pool of wet storage is chosen to be the environment of study. The design of corrosion coupon is described elsewhere [2]. The first batch of six coupon assemblies were strategically positioned in the service pool that located just beside the pool, and the water quality is precisely similar to pool water. Coupons are immersed into the spent fuel storage pool by hanging them up to for about 1 meter above the floor.

2.2. Water Quality analyses

pH, conductivity and *Total Dissolved Solid* (TDS) were monitored by on line. The pH of trapped water between two sample is being measured by inserting the lakmus paper into the space between the sample. Some element such as sulphate, chlor etc. were analysed weekly by using test kit available. The dissolved ions were analysed by using ion chromatography and Atom Absorption Spectrophotometer.

2.3. Sample Coupons Analyses

One set coupons are being withdrawn from the wet storage pool after three years immersing duration. Then, both racks are being monitored from the radiation exposuring level and being recommended that it is safe, then, the racks are dismantled. The surface of each disc is documented by using camera. The present surface inspections are being compared to the origin and a year immersion's result, to be understood the visual surface changing occur and probable pits. White deposit is sampled and analyzed for microbe existence.

3. RESULT AND DISCUSSIONS

Both racks of samples have been withdrawn from the wet storage pool, and being inspected by the radiation safeguard for its radiation exposuring. After being recommended that the radiation exprosuring is similar or under the backgound, then the further analyses procedure was proceeded.

3.1. Water quality

Conductivity and pH of the water inside the spent fuel storage pool were measured as 1.3–1.7 μS/cm, and 5.2–5.5,respectively. There is no Sulphate and Chlor detected. The TDS is around 0.7–0.9 μS/cm, and the temperature at inlet the ion exchange resin for purification is around 23–34^{o}C. From these results, it is known that the water environment is good enough for maintaining the structure material, due to no detected sulphate and chlor ion, which are the active ions that may induce corrosion process.

3.2. pH between disc

Soon after the withdrawing, the pH of trapped water between two discs was measured by inserting pH-paper through the gap. From the measurement, it is known that the pH between two disc is 4, one point below the water surrounded, in wet storage pool (pH 5.4–5.5), which is measured by using digital pH meter with acuracy of ±0.1.

3.3. Sample analyses

Withdrawn both coupons are shown in Figure 1. There is no color change observed. It can be understood that the general corrosion is not the main corrosion process for both position even after three years exposuring duration. After dismantling, it can be seen that there is white debris on the surface on horizontally coupon (Figure 2). The dismantling process was difficult, because of corrosion, both discs are being sticked together.

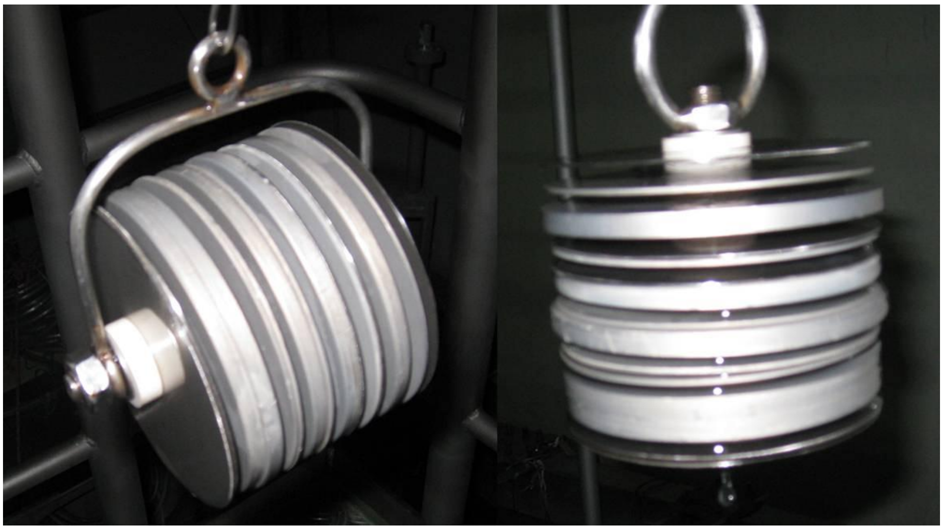

Fig. 1. Coupon after 3 years exposuring, vertically and horizontally.

Fig. 2. White debris on the surface of horizontally coupon.

The galvanic pair discs are being dismantled and each pair are being documented and can be seen as upper for SS 314 and lower row for AlMg2 in Figure 3. Similar pair for galvanic objects are being compared for each exposuring duration, as zero, a year and three years [4]. From Figure 3, it can be understood that the Galvanic corrosion occurs clearly for both pair of SS314–$AlMg_2$ and SS314–Al_2Mg_3 (Figure 3.). The level of galvanic corrosion becomes worse due to the length of exposuring time. The three years exposuring time shows darker than the shorter exposuring time a year. However, the aluminium coupons show more sacrified than the SS coupon. Coupons from the vertically rack shows more severe galvanic corrosion occur. It can be considered that the trapped water in vertically racks seem to be stand longer than the horizontally racks, even there is no significant pH difference for both vertically and horizontally racks. From this results, it can be understood that the wet storage water flow rate doesn't fast enough to suppress the corrosion process for material which is being closed together, as experiment.

The white precipitation that mostly appears on the aluminium surface as Al_2O_3, has made those both galvanic pair becomes difficult to be separated by hands, because of sticked each other. The white precipitate is formed because of potential different between both material that initiated the galvanic corrosion. From the weight measurement, there is only less than 1 μg weight difference for both before and after exposuring coupons into the storage pool water, therefore becoming undetected well. The longer exposuring duration of 3 years does not increase significantly the weight of white precipitation formed.

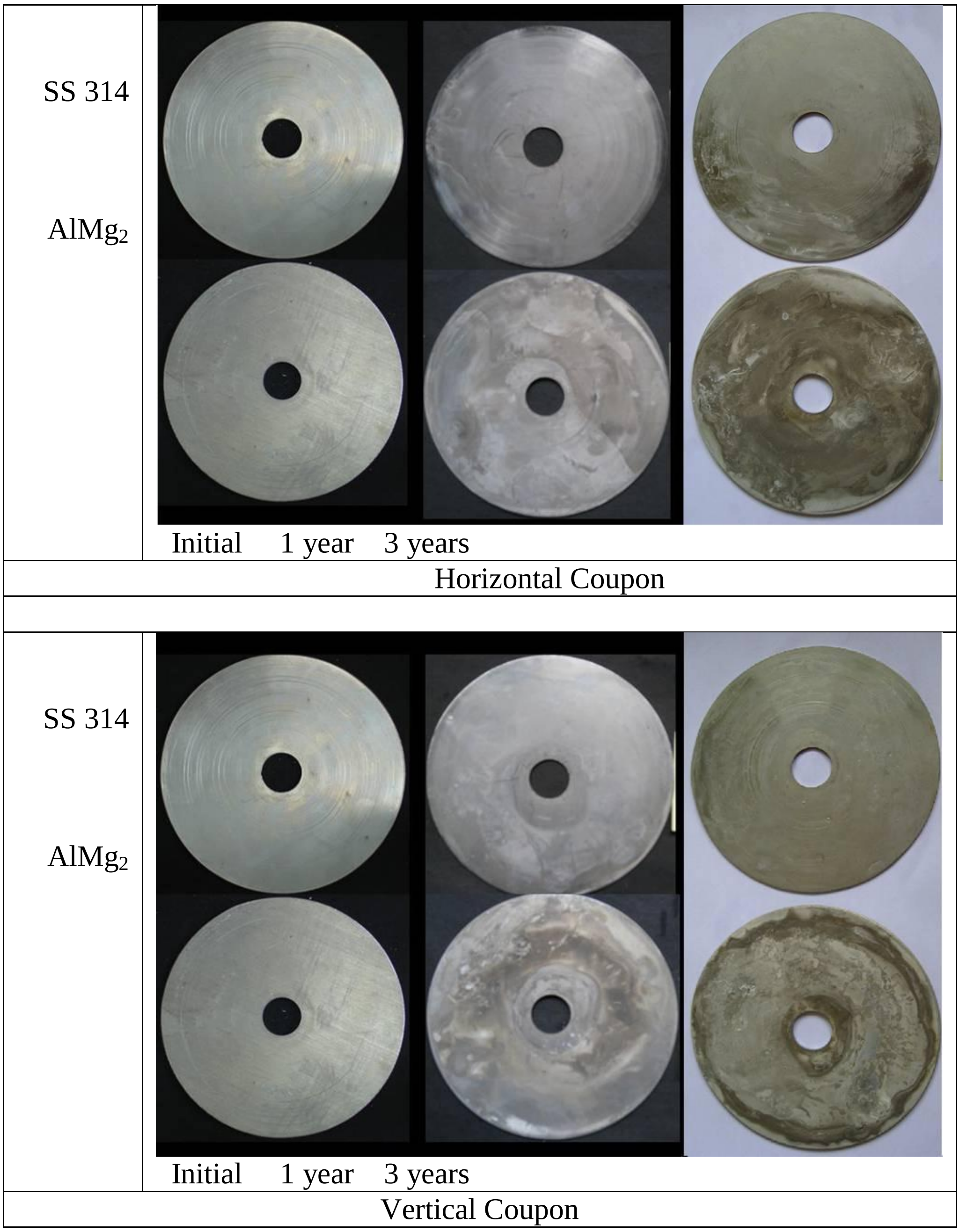

Fig. 3. Galvanic Corrosion between SS and $AlMg_2$ at various exposuring time up to 3 years.

Crevice corrosion either is clearly being observed from $AlMg_2$ and Al_2Mg_3. The white precipitation appears more clear than galvanic corrosion in scale range of less than 10 μg with deviation error of 20%. Longer exposuring duration of 3 years gives more precipitation visually, but still can not be measured quantitatively. Pair of aluminium from different number of magnesium gives white precipitation either. In this case the corrosion process of galvanic and crevice are mix. Visually gives similar white precipitation on both surfaces. Bio-corrosion does not occur and observe in these experimental results. Pitting corrosion also does not detected.

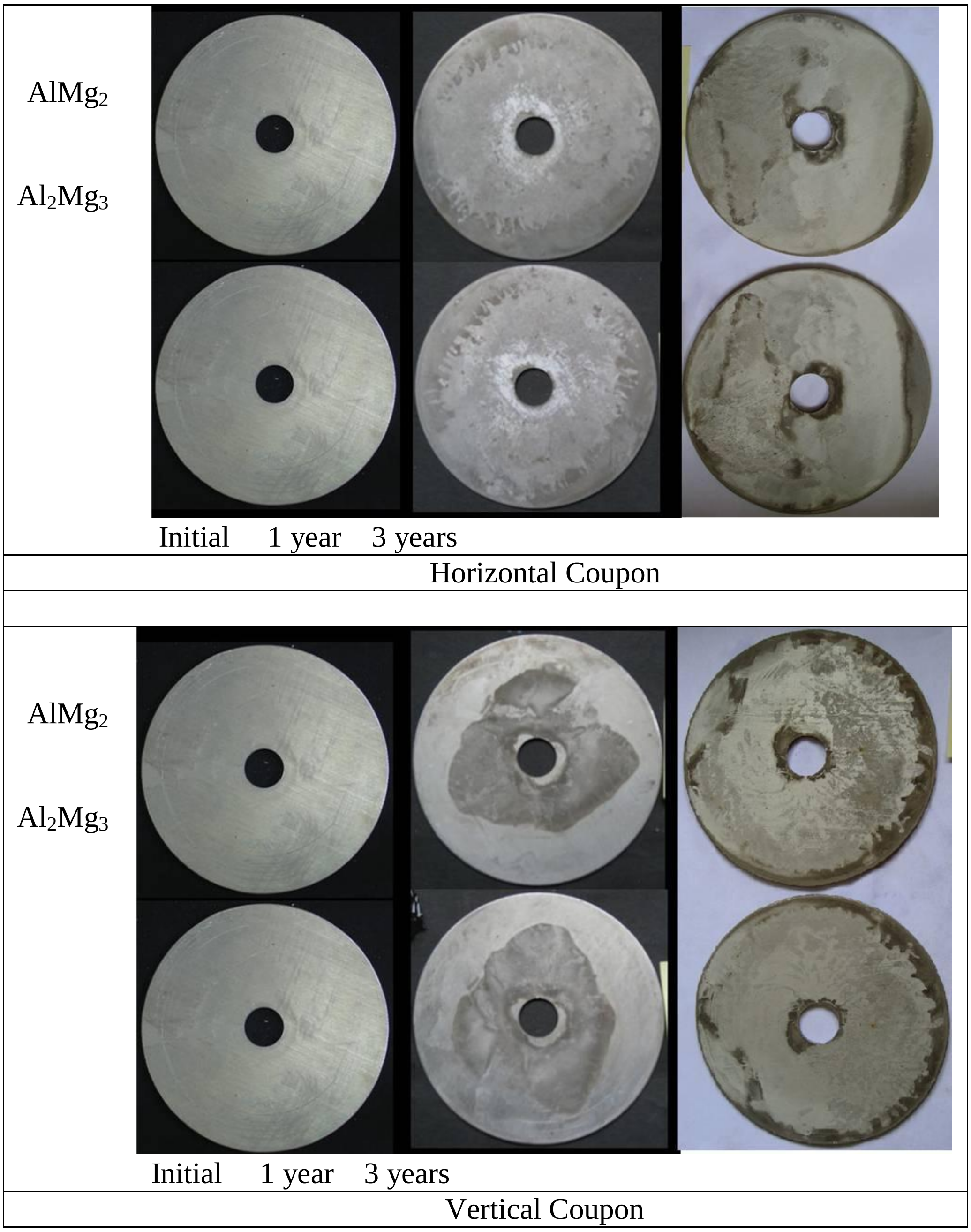

Fig. 4. Crevice Corrosion between $AlMg_2$ and $AlMg_2$ at various exposuring time up to 3 years.

From these results, it can be described that the corrosion of aluminum, induced by crevice or galvanic, dependent upon a vast number of variables. These variables include environment, temperature, water flow velocities, impurities present in the water, and chemistry conditions to which it is exposed. Many of the factors are controlled by design and construction, such as distance between one and other material, that may induce crevice and galvanic corrosion. Pretreatment, soluble and solid impurities, and chemistry of water cooling are the main factors that controlled the corrosion. It is clearly understood that the corrosion in primary water cooling of RSG GAS is very slow compare to secondary cooling water [5].

The white deposit on the coupon's surface, is controlled by the chemistry, including pH, dissolved oxygen, and conductivity. Dissolved oxygen is controlled for essentially the same reasons as for the other metal such as iron. Conductivity is a quantitative indication of the impurities present in the system, and pH theoretically dictates the value of conductivity.

For research reactor plants in which aluminum is used for tank liner and fuel cladding, pH is controlled in an acidic condition because of the corrosion properties of aluminum,therefore pH control is the most important thing that should be aware. pH also has a marked effect on the rate of chemical reaction between the coolant water and aluminum, and also radiolyses product and aluminum.

In case of fuel and spent fuel cladding, the corrosion reduces the thickness and forms an oxide film that may cause as a thermal barrier. From the extensive tests carried out by DOE supporting have revealed that the minimum aluminum corrosion results is with a pH of 5.0, at normal operating temperatures and it is noted that aluminum has a minimum solubility at room temperature at pH near 5.5, 25°C. The aluminum corrosion products tend to reduce the base metal. Therefore, it is desirable to maintain the system pH level in the range of minimum oxide solubility.

From these results, it can be resumed that maintaining pH, water purity and water flow are the main important factors that should be applied for maintaining the reactor research structure materials.

4. CONCLUSION

From the experiment, it is known that the trapped water pH between the coupons is 4, one digit lower than the pool water (pH 5.4–5.5 that was measured digitally with accuracy of ±0.1). The conductivity, pH, Chlor and TDS were analyzed as 1.3–1.7 μS/cm, 5.2–5.5, none and 0.7–0.9 μS/cm, respectively. It can be resumed that (a) water quality of primary water is required the SAR, (b) crevice and galvanic effect are the main corrosion; (c) no pitting; (d) no biofilm observed. The experience and scientific results gained is very useful to be implemented for Indonesia Research Reactor Ageing Program and future NPP.

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