

CORROSION SURVEILLANCE PROGRAMME FOR LATIN AMERICAN RESEARCH REACTOR AL-CLAD SPENT FUEL IN WATER

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Summary

The objectives of the IAEA sponsored Regional Technical Co-operation Project for Latin America (Argentina, Brazil, Chile, Mexico, and Peru) 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 research reactor spent fuel during operation and interim storage as well as final disposal, and to establish forms of regional cooperation in the four main areas: spent fuel characterization, safety, regulation and public communication. This paper reports the corrosion surveillance activities of the Regional Project and these are based on the IAEA sponsored co-ordinated research project (CRP) on “ Corrosion of research reactor Al-clad spent fuel in water”. The overall test consists of exposing corrosion coupon racks at different spent fuel basins followed by evaluation.

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) have been 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 will cease, and National solutions in countries without Nuclear Power Programs 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 or macro-activities: (1) spent fuel characterization; (2) safety and regulation; (3) options for spent fuel storage and disposal; (4) public information and communication. Corrosion monitoring and surveillance is one of the activities of the subproject ‘Spent fuel characterization’.

Fuel type and storage

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 Central Storage Facility (CSF), at the Ezeiza Nuclear Center in Argentina, is the only wet, away-from-reactor storage facility. The CSF contains two sectors of in-ground storage tubes for spent nuclear fuel (SNF). The tubes are interconnected via a filtration/de-ionization system and the water is re-circulated. Figure 1 illustrates the CSF. Other in-reactor storage facilities consist of aluminium or stainless steel storage racks as shown in figures 2 and 3.



Figure 1: Central Storage Facility at Ezeiza Nuclear Center, Argentina.



Figure 2. Storage racks in auxiliary pool of Peruvian reactor in Lima, Peru.

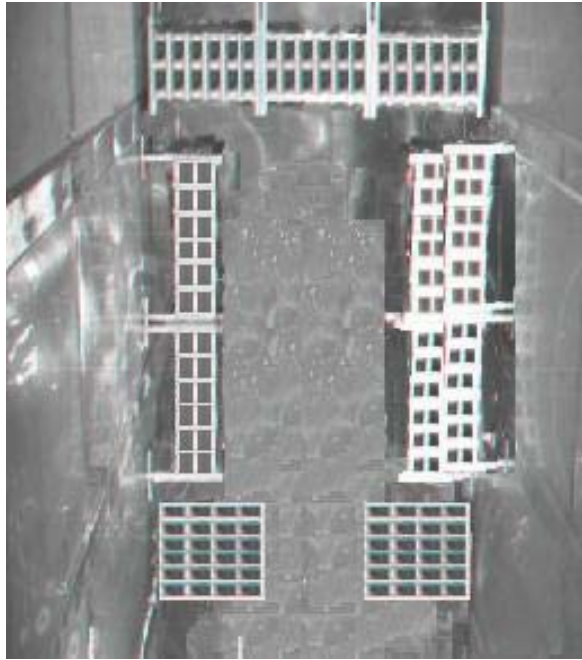


Figure 3. In-reactor storage racks in IEA-R1 research reactor, IPEN, Sao Paulo, Brazil

Two of the countries participating in this Regional Project (Argentina and Brazil) were (and continue to be) participants of an IAEA sponsored co-ordinated research project (CRP) on “Corrosion of research reactor Al-clad spent fuel in water”. The corrosion surveillance activities of the Regional Project are based on this CRP. The objective of this activity is to evaluate the effect of Latin American spent fuel basin parameters on the corrosion behaviour of research reactor fuel cladding. This evaluation will be done by exposing racks containing Al alloy and stainless steel coupons at the different spent fuel basins followed by their withdrawal after periodic intervals for examination. Many procedural changes have been incorporated to maximize results. This test will be complimented with laboratory tests at some of the sites. Prior to initiating the tests, an assessment visit to the different storage sites was made by one of the authors, a workshop was conducted in Sao Paulo, Brazil, the corrosion test coupons and racks were manufactured, equipment was purchased for some of the participants and a test protocol was elaborated. The following sections provide further details about these activities. In the long-term exposure test, initiated this year, 3 racks were immersed in each of the basins. The racks are expected to be withdrawn after one, two and three years of exposure.

Assessment visits

At the request of the IAEA, preliminary assessment visits were made by one of the authors (LVR) to the spent fuel storage facilities at: (1) Instituto Nacional de Investigaciones Nucleares (ININ), Mexico, D.F., México; (2) Instituto Peruano de Energia Nuclear (IPEN), Lima, Peru; (3) Comision Nacional de Energia Atomica (CNEA), Constituyentes, Buenos Aires, Argentina; (4) Centro Atómico Bariloche, CNEA, San Carlos de Bariloche, Argentina. These visits were considered important, based on experience from the first stage of the IAEA-CRP on “Corrosion of Research Reactor Al-clad Spent Fuels”. The main objectives of these visits were to:

1. Inspect the spent fuel and to obtain information about:

- (a) peculiarities in terms of geometry of storage racks;
- (b) position, material and state of specific reactor/basin components;
- (c) water flow conditions;
- (d) extent of pool surface dust and settling;
- (e) amount of sludge and frequency of clean up; and
- (f) illumination;

and to help select the location for immersion of the corrosion test racks.

2. Collate information to advise local staff both at the site and during the Workshop in Sao Paulo, Brazil, in November 2001.

3. Evaluate laboratory facilities, (for corrosion measurements, chemical analysis and microstructural studies) either at the site or in nearby institutions to which they have access, and accordingly recommend complementary tests or measurements.

Corrosion test rack and coupons

Since the main objective of this activity was to evaluate the effect of Latin American spent fuel storage basin parameters on the corrosion behavior of research reactor fuel cladding, a questionnaire was prepared and sent to the participating countries to report the number of storage sites, the cladding alloys, the materials of construction of the storage basins and the basin water parameters. These data are summarized in Table I and were used to specify the coupon materials. It was also considered essential to expose aluminium alloy coupons fabricated under conditions similar to those used to prepare fuel plates. The surface condition was also similar to that of fuel plates used in Research Reactor fuel elements. The aluminium alloys used to fabricate the test coupons were obtained from the Fuel Fabrication Department of IPEN, Brazil. Identical aluminium alloys are also used in the Fuel Fabrication Section in Chile.

Fifty racks, similar to that shown in figure 2, were manufactured in Brazil and sent to the participants. Each rack consisted of a stand on which circular metallic coupons and circular ceramic disks were stacked. The metallic coupons were either individual coupons or coupled coupons. The ceramic disks (non-porous alumina) were used to separate the individual coupons and the coupled coupons, one from the other. The coupons were stacked in the following sequence, from top to bottom:

- Al 1050
- Al 6061
- Al 1050 (pre-oxidized and scratched)
- Al 1100 - Al 1050 (couple)
- Al 1050 - Al 6061 (couple)
- Al 6061 - Al 6061 (couple)
- Al 1050 - SS 304 (couple)
- Al 6061 - SS 304 (couple)

Table I: Details of cladding alloys, other basin materials and water parameters.

Country (no of basins)	Basin I.D.	Fuel cladding alloys	Other basin materials	Basin water parameters
Peru (2)	Main pool; Aux. pool	AlMg1 AlMgSi 1	SS316L;Al99.5	pH- 6.0,Cond.- 1.3 μ S/cm,Fe- <0.05mg/l,Cl- <1.00mg/l
Chile (4)	Rech- 1(2);Rech-2 (2).	AlNP4 (BS 1470- grade 1B);Al- L115 (Al- 1100)	AlNP4;Al-2S; SS321;EN- 58B; SS321Al- ASTM:B209; SS:F314; SS304;S314	pH-5.63- 6.5;Cond- 0.78 μ S/cm;Fe- 0.1 μ g/l;Cu- 1.4 μ g/l;Cl- 3.5 μ g/l
Argentina(6)	CSF-new; CSF-old; RA1- reactor pool; RA3-decay pool; RA6- decay pool; RA6-reactor pool	CSF- new-Al6061 CSF- Old-Al99.7 RA3- decay-Al6063 RA1- pool-Al-99.7 RA6- pool-Al99.7 RA5- decay-Al99.7	Non std-SS Non std SS SS304 Al99.7 SS304 SS304	Good Poor V.good Excellent V.good V.good
Brazil (2)	IPEN; CDTN	Al 1100; Al6061;Al6262 AA 5052; SS 304	SS304 Al 1100; AA 5052	V.good; Cond- 8 μ S/cm



Figure 3. Corrosion test rack

The racks had enough space to accommodate a few site specific coupons.

Surface preparation and treatment of the aluminium alloy coupons was identical to that given to Al-clad fuel plates. All the coupons were machined, polished to semi-bright, identified, degreased and given the following surface treatments .

- a. Pickling in 10 wt% sodium hydroxide solution at 70-80° C for 30 seconds ;
- b. Cleaning by brushing in running filtered water at room temperature for one minute;
- c. Rinsing in running deionized water at room temperature for one minute;
- d. Neutralization in 5 vol% nitric acid solution at room temperature for one minute ;
- e. Rinsing in filtered running water at room temperature for one minute;
- f. Pickling in 10 wt% sodium hydroxide solution at 70-80° C for 30 seconds;
- g. Repetition of steps (b) to (e);
- h. Immersion in deionized water at room temperature for 5 minutes;
- i. Rinsing in deionized water for one minute;
- j. Drying in hot air at 180° C for one minute.

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 scriber to simulate a damaged fuel element surface.

The workshop

At the Workshop on Characterization, held in Sao Paulo, Brazil, in November 2001 the activities related to corrosion included presentations covering different aspects of corrosion of aluminium in spent fuel storage basins and a practical demonstration about: (a) corrosion test rack handling procedures prior to immersion in the spent fuel basin; (b) immersion of rack and sampling of

water; (c) monitoring water parameters and corrosion performance of the rack while immersed; (d) removal and examination of the rack and coupons; (e) transfer of rack to the corrosion laboratory and (f) detailed examination of exposed coupons. A demonstration of laboratory immersion tests, electrochemical measurements and interpretation of results as well as recording of data was also given.

The test protocol

A Test Protocol was prepared and sent to the participants. This described details about handling coupons and racks prior to assembling, during the test, after disassembly and during evaluation. The test Protocol is in Appendix 1.

Equipment for corrosion testing and evaluation

To enable laboratory corrosion tests to be carried out in Peru, an electrochemical system was acquired. An Image Analysis Work Station was acquired and installed in IPEN, Brazil to enable all participants to study corroded surfaces and to quantify extent of surface pitting as a function of basin water parameters.

General comments

A number of procedural changes have been incorporated in the corrosion surveillance programme of the Regional Project. These include:

1. Preliminary assessment visits to the different fuel storage sites and selection of positions for immersing or placing the test racks.
2. Use of rolled aluminium alloy sheets or plates for the preparation of test coupons. Sheets and plates used to manufacture fuel plates were utilized.
3. Surface preparation and treatment identical to that used in fuel plate manufacture.
4. Revised test protocol to maintain rack handling and evaluation of corroded coupons uniform at the different sites.
5. A practical demonstration of all the stages in conducting and evaluating corrosion tests prior to initiating the tests.
6. Prohibiting interim examination of the racks. It is permitted only in specific cases provided the rack and coupons are maintained wet.
7. Prohibiting disassembly of the rack during interim inspections, if any.
8. Prohibiting return of racks to the basin for continued testing.
9. Quantitative determination of pitting corrosion as a function of basin parameters with the aid of an image analyzer.
10. Complementary laboratory tests to determine the effect of site-specific water parameters on corrosion behavior of Al alloys.
11. Testing the effect of Al coupon orientation on corrosion behavior in a storage basin. This was motivated by the fact that fuel plates in a fuel element are vertically oriented. Hence, two racks have been suspended horizontally in the IEA-R1 pool to have the coupons oriented vertically.

Appendix 1.

THE TEST PROTOCOL

THE PARTICIPANTS OF THE REGIONAL PROJECT WILL BE PROVIDED WITH 3 CORROSION RACKS FOR EACH SPENT FUEL BASIN SITE. THESE RACKS WILL CONTAIN COUPONS ASSEMBLED SEQUENTIALLY AND IN A CERTAIN ORDER FROM BOTTOM TO TOP FOR EACH ALLOY.

The word coupon is recommended. Please avoid use of specimen, sample, disk etc.

Definitions:

Corrosion rack or rack is the assembled set of coupons, spacers, central support tube and hanger.

Basin, pool, pond - is the contained volume of water used for storage of spent fuel.

1. Pre-assembly

- Unpack rack and disassemble it. Handle with surgical gloves, removing any talcum powder on the outside of the gloves. Weighing of individual coupons is optional. No further cleaning or surface treatment of coupons is necessary. Each rack has one coupon passivated at 95°C for 12 hours in water and scratched (on the side **without** the identification number). (***Please disregard the twin scratches on the side with the identification***)
- Add coupons of site-specific materials, recording all relevant information about the materials and the coupons. Prepare these coupons as per instructions provided.
- The site-specific coupons should have identification numbers. The numbering system chosen should be logical, carefully recorded at the beginning of the test and adhered to throughout the programme.
- Photograph the front and back of each coupon.

2. Assembly

- Assemble coupons in the specified order. The coupon with the scratch should be assembled with the scratch side facing upwards. Site-specific stainless steel coupons should be below all aluminium alloy coupons.
- Note the position of site-specific coupons.
- Tighten the top hanger-nut by hand until tight and then give a further 10° turn.
- Photograph the assembled rack.
- Attach a nylon rope to the hanger on the rack. (avoid use of steel wire or cable as aluminium and steel form a galvanic couple).

3. Immersion in storage basin

- Lower/position the corrosion rack into the water in the vicinity of spent fuel if possible.
- Position below water surface and above basin floor. Do not allow the rack or any of the coupons to touch walls, floor, sludge or metallic components.
- Record the location (depth, distance from walls etc.). Make a sketch to remind you and for the final report.
- Measure the radiation field intensity at periodic intervals underwater near the rack (in R/h, or Sievert/h).
- Sample the water at the immersion depth, as near to the rack as possible.
- Indicate flow conditions near rack, (flowing or static), rate of or frequency of renewal of water in basin.
- Observe if loose particles are present on coupon surface, and if they appear to be causing a corrosion problem, please describe.

4. Exposure interval

- Place three racks at each site as soon as possible.
- Record the date of immersion.
- The racks are to be withdrawn after, one, two and three years.

5. Removal and examination of coupons.

- Withdraw the rack from basin
- Measure pH of water on the external surface of coupons.
- Compare with pH of bulk water.
- Photograph the rack prior to disassembly. Photograph all points of interest including any corrosion of the edges.
- Remove the coupons from rack.
- Photograph front and back of each coupon. A small card with a note about the material, immersion time and coupon identity number should be photographed together with the coupon.

- Count the number of pits.
- Make observations of specific corrosion phenomena for each coupon, including ease of removal (ease of separation of crevice/bimetallic coupons), amount and type of loose deposits, staining, discoloration, pitting, tenacious or loose oxides, raised or embedded particles felt with a gloved finger, etc.
- Measure pH (with pH paper) on the inside faces of crevice and bimetallic coupons, compare with bulk water pH and with that on the outside surface.

6. Post storage detailed examination

- Decontaminate coupons with a *chloride-free* detergent and rinse with deionized water.
- Air dry/wipe.
- To evaluate pitted aluminium coupons: Use 50% phosphoric acid solution, if necessary, to clean/dissolve oxide from pits to conduct metallographic evaluation. Immerse for a short interval only. Remove from the solution as soon as bubbles start to increase. Further exposure will dissolve the base metal.
- Ensure that there are no oxides in the pits before determining the true pit depth.
- Conduct metallographic evaluation on the deepest pit(s). Measure pit depths with a stylus or using the calibrated focusing technique.

7. Final report

- Issue your final report including photographs, data, detailed observations and conclusions following the *Instructions for Authors of IAEA Documents*.

Further notes for evaluating coupons after exposure

Pitting corrosion

- Pits should be photographed.
- Pit depths should be determined, if a stylus device is available, or if a calibrated microscope stage is available.
- Final evaluation should focus on the deepest pits and should be carried out by sectioning and polishing, as in the preparation of all metallographic coupons.
- The section should be through the deepest part of the pit.
- Some attempt should be made to count the pits along with measurement of their diameters and depths. A stocastic approach can be used.

Crevice corrosion

Opening a crevice couple destroys or drastically changes the crevice features. Therefore, it should be opened and evaluated only once.

- Visual and photographic inspection, together with determination of pH using pH-paper is the first step.
- Metallography should be carried out and some evaluation of the area of attack should be made.
- Evaluation of pitting in the crevice should follow the above instructions for pitting.

Galvanic (bimetallic) corrosion

Galvanic couples should be treated in precisely the same way as crevice couples.