

Material Characterization and Corrosion Control in Wet Storage of Chilean Spent Fuel.

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ABSTRACT

Chile has two MTR type research reactors and the spent fuel will be stored in water previous to the conditioning for final disposal. One of the serious problem presented during wet storage is the phenomenon of corrosion, which depends on the water quality, the structural materials and the storage conditions. Thus, it is necessary to solve how to guarantee the integrity of the spent fuel during its wet storage. The water quality and fuel assembly materials are being characterized with the purpose to define the criteria of surveillance and control of corrosion as a function of time.

The behavior of the 6061 Al and N4 Al alloys is being studied to characterize the susceptibility to pitting corrosion in solutions with chloride and cadmium as aggressive ions. The analyses were performed in a three-electrode electrochemical cell with 6061 Al and N4 Al as working electrodes. Platinum wire was the auxiliary electrode while Ag/AgCl was the reference electrode. To obtain the electrochemical characterization the polarization curves were used and the evolution of the corrosion potential of the aluminum alloys and SS 304 were measured. The electrolyte was deionized water with different concentrations of chloride and cadmium.

At present, the results show that 6061 Al and N4 Al alloys are more susceptible to be attacked by pitting due to the presence of chloride than cadmium.

INTRODUCTION

The spent fuel produced by the operation of the two Chilean research reactors will be locally managed in the future. Thus, one of the major problems is to guarantee the integrity of the aluminum-clad of the fuel during wet storage. To reach this objective, it is necessary to evaluate the resistance of aluminum and aluminum alloys against corrosion under the present storage conditions.

RECH-1 Reactor

The RECH-1 research reactor is a pool type reactor with a nominal thermal power of 5 MW. The reactor is operated by the Chilean Nuclear Energy Commission (CCHEN) at La Reina Nuclear Center. Light water is used as moderator and coolant and beryllium as reflector. The maximum thermal neutron flux is about 6.8×10^{13} n/cm².s. The reactor reached its first criticality in 1974 using HEU (80% of ²³⁵U) fuel assemblies. A total of 58 HEU spent fuel assemblies were shipped to Savannah River Site in two shipments (August 1996 and December 2000) using NAC-LWT shipping casks.

The present core configuration has 34 fuel assemblies, 30 MEU fuel assemblies (45% of ²³⁵U) and 4 LEU fuel assemblies. The LEU fuel assemblies containing U₃Si₂-Al were fabricated by the Chilean Fuel Fabrication Plant. The technical specifications of the LEU fuel assembly

were developed by the Chilean Manufacturer based on the original HEU fuel assembly and approved by the reactor operator.

RECH-2 Reactor

The RECH-2 research reactor is a MTR type research reactor with a design thermal power of 10 MW. The reactor is operated by CCHEN at Lo Aguirre Nuclear Center. This reactor has an inventory of 29 HEU (90% of ^{235}U) fuel assemblies and it was licensed to operate up to 2 MW with HEU fuel. The present status of the reactor is permanent shutdown.

MATERIAL CHARACTERIZATION

A. Material Characterization

RECH-1 Reactor

Component	Material
MEU FE cladding (45% ^{235}U)	N4 Al (B.S. 1470) Gr.1 B.
LEU FE cladding	6061 Al
Liner	ASTM 312 Grade 321
Spent fuel rack	N4 Al (B.S. 1470) 2S Al (Standard)

RECH-2 Reactor

Component	Material
HEU FE cladding (90% ^{235}U)	1100 Al
Liner	AISI S314
Storage pool	AISI 304

Aluminum Alloys Compositions

Alloy	Si	Cu	Mn	Mg	Cr	Fe	Zn	Ti	Al
Al 6061	0.60	-	0.28	1.0	0.20	-	-	-	remainder
Al N4	0.40	0.15	0.10-0.50	1.70-2.40	0.15	0.50	0.15	0.15	remainder

B. Water Quality Control

Determination of pH, conductivity, and chloride, iron and copper ions from the RECH-1 and RECH-2 reactor pools is a standard practice and it has been carried out using the norm CCHEN N° 3.2.1[1].

	RECH-1	RECH-2
Element	Concentration (ppm)	Concentration (ppm)
Fe	0.001	0.005
Cu	0.001	0.005
Cl	0.050	0.046
SO ₄	0.05	0.10
NO ₃	0.1	0.1
Cd	0.02	-
NO ₂	0.1	0.1
Ca	0.025	0.050
Na	0.05	0.05
Si	3.97	2.00
Pb	0.050	0.037
B	-	0.01
Hg	0.500	0.001

C. Radionuclides

Samples of water from the reactor pools are regularly sent for radiological analysis using gamma spectroscopy of high resolution. One of the most important radionuclides is ¹³⁷Cs which is extremely soluble in water. The activity rate of ¹³⁷Cs is directly related with failed spent fuel elements or breach of clad[2].

D. Surveillance and Monitoring of Corrosion

Corrosion rate is normally low for research reactors when water is demineralized and the temperature remains close to the room temperature. However, the corrosion resistance of materials must be guaranteed for long term storage of spent fuel assemblies[3].

To evaluate the corrosion resistance of fuel assemblies it must take into account the following factors: type of alloy, water quality (pH, conductivity, chloride, sulfates, impurities, and heavy elements), galvanic effect, and crevice[2]. On the other hand, corrosion is promoted by mechanical damage of cladding due to removal of thermocouples, blisters caused by hydrogen, deformation due to thermal stress, etc.

The surveillance and monitoring of the mentioned factors were considered in the IAEA Regional Project RLA/4/018: "Management of Spent Fuel from Research Reactors in Latin America". Therefore, in situ tests were established in each participant country in accordance with an approved protocol. Due to the great number of parameters to be considered (water

velocity, presence of mud, gradients of temperature, activity of radionuclides, etc) the development of a protocol was an important part of the project. The protocol provides the way for standardizing the activities and the results obtained by the countries could be comparable.

E. LABORATORY TESTS

With the purpose to characterize the behavior of some materials, which are components of the reactors, tests at the laboratory scale were performed using a three-electrode electrochemical cell. Due to LEU fuel assembly of the RECH-1 reactor was fabricated using 6061 Al, one of the working electrodes is 6061 Al, and N4 Al(1) provided a second working electrode. The surface exposed was ground to 600 grit to have a fresh surface. Platinum wire(2) is used as an auxiliary-electrode and Ag/AgCl(3) as reference electrode. The electrochemical characterization utilizes a potentiostat/galvanostat which provides cyclic polarization and potential of corrosion. The electrolyte is deionized water with different concentrations of chloride and cadmium.

The effect of ion chloride and cadmium in the corrosion of 6061 Al and N4 Al is being evaluated, where deionized water with addition of chloride and cadmium is used. Tests with high concentration (50, 100, and 1000 ppm) and low concentration (1, 5, and 10 ppm) of chloride were done. To study the effect of cadmium it was used high concentrations of cadmium chloride (40, 80, and 160 ppm) and low concentrations of cadmium sulfate (1, 5 and 10 ppm).



CONCLUSIONS

- The storage of aluminum clad spent fuel must be in optimum quality water; otherwise, it can result in aggressive pitting corrosion.
- An increase of chloride concentration reduces the pitting potential and the aluminum alloys are more susceptible to be attacked by pitting corrosion, Figure 1.
- The cadmium ion modifies the anodic region due to the formation of passive film on aluminum in a small band of potential, Figure 2,3.
- The results have shown that 6061 Al and N4 Al alloys are more susceptible to be attacked pitting due to the presence of chloride than cadmium.
- The difference of potentials between aluminum and stainless steel is more than enough to produce galvanic corrosion. The potential of corrosion is higher for N4 Al than 6061 Al; however, after 2,500 sec both alloys present a similar behavior, Figure 4.
- Figure 5 and 6 show oxide (white zone) but not pitting. The EDS analyses confirmed the presence of cadmium on the aluminum surface.

REFERENCES

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3. Vivekanand Kain, S.S Shinde y P. Seetharamiah. " Prediction of Pitting Corrosion Tendency of materials During Extended Storage of Spent Nuclear Fuels". Proc. Int. Conf. On Corrosion CONCORN '97, Dic 3-6, Mumbai, India

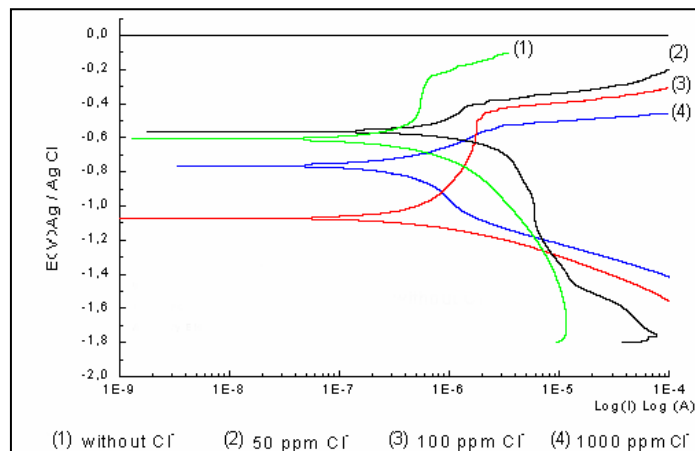


Fig. 1 Anodic polarization curves in different concentrations of chloride

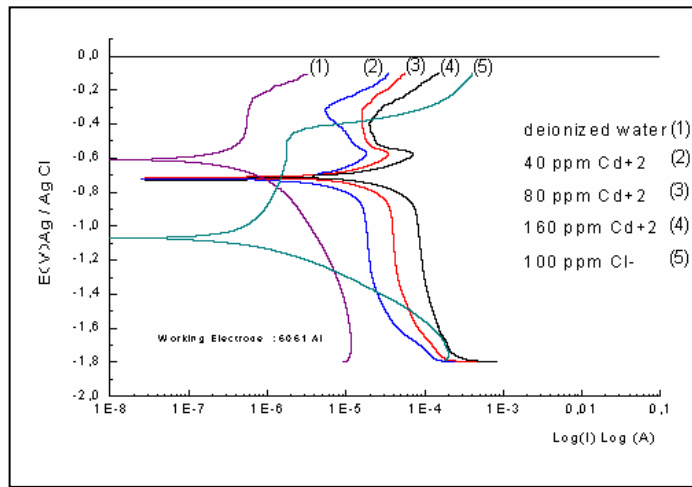


Fig. 2 Anodic polarization curves in 40,80,160 ppm of cadmium and 100 ppm of chloride

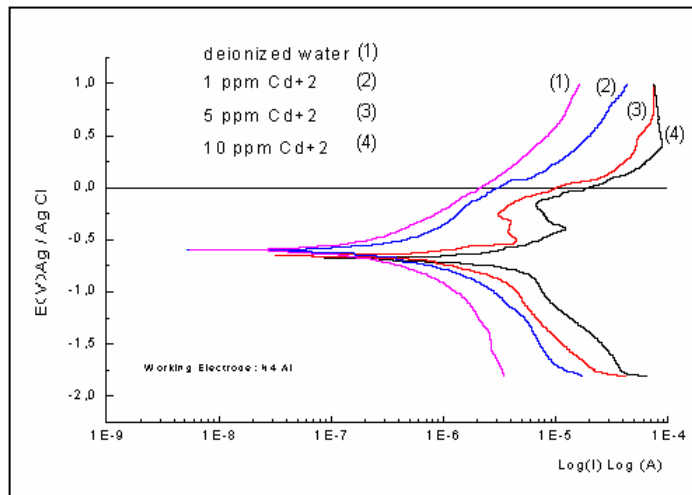


Fig. 3 Anodic polarization curves in 1,5 and 10 ppm of cadmium

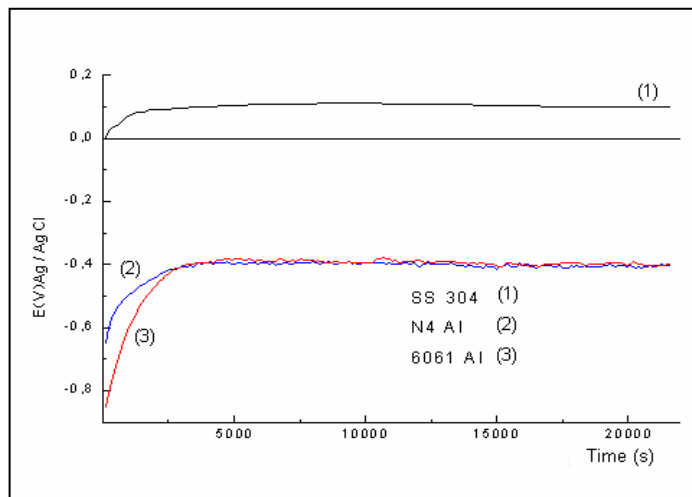


Fig. 4 Evolution of corrosion potential of different materials in 1 ppm of chloride.

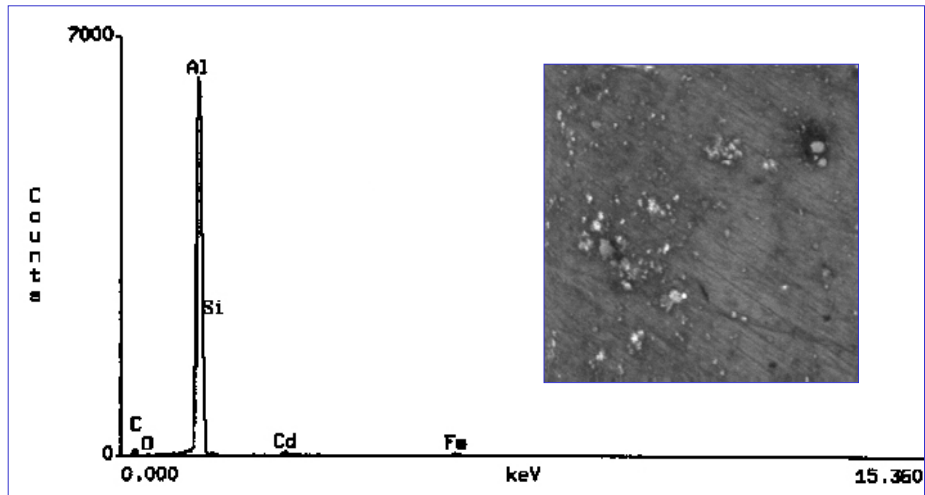


Fig. 5 SEM and EDS of aluminum surface in solution with 40 ppm of cadmium

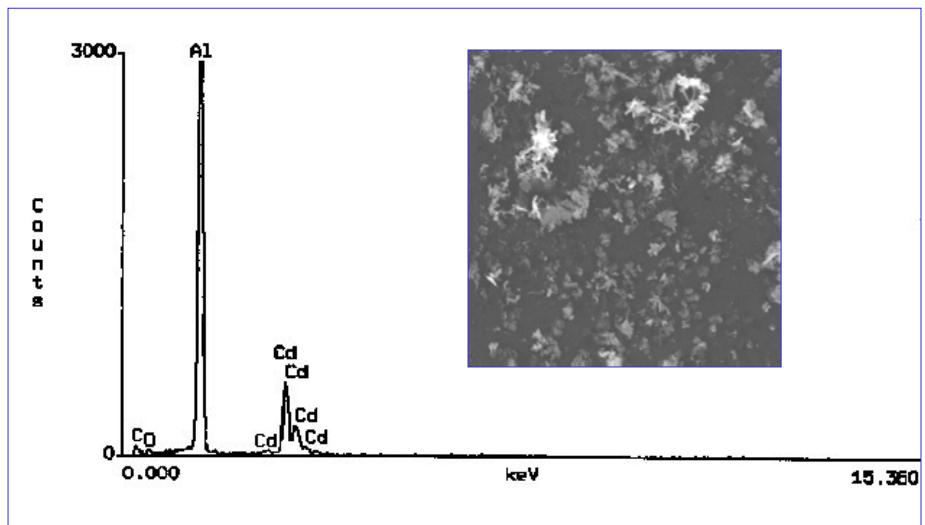


Fig. 6 SEM and EDS of aluminum surface in solution with 80 ppm of cadmium