

RESEARCH REACTOR SPENT FUEL MANAGEMENT IN ARGENTINA

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1. INTRODUCTION

In Argentina the radioactive waste and spent fuel management activities are carried out according to the Act N° 25018 ("Radioactive Waste Management Regime") passed by the Parliament in the year 1998, this Act design the Comisión Nacional de Energía Atómica (CNEA) as the governmental organization responsible for the management of the spent fuel and the radioactive waste generated in the country. Moreover, all the activities related to radiological and nuclear safety are regulated and controlled by the Autoridad Regulatoria Nuclear (ARN).

Furthermore, Argentina is a Contracting Party of the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management, adopted in Vienna on September 1997 and entered into force on June 2001.

Argentina is taking part in the IAEA's Regional Project RLA/4/018 on the Management of Research Reactor Spent Fuel (RRSF), together with Brazil, Chile, Mexico and Peru.

2. RESEARCH REACTORS AND CRITICAL ASSEMBLIES

At present CNEA has in regular operation three research reactors (RR): RA-1 (100 kW) located in the Constituyentes Atomic Center, RA-3 (5 MW, being upgraded to 10 MW) located in the Ezeiza Atomic Center and RA-6 (500 kW) located in the Bariloche Atomic Center. Reactor RA-1 is used mostly for research experiments on radiation damage of materials. Reactor RA-3 is mostly dedicated to the production of radioisotopes for medicine and industry and for testing of materials and prototype fuel elements. Reactor RA-6 is used for teaching and research.

In addition there are three critical assemblies: RA-0, located in the University of Córdoba; RA-4, located in the University of Rosario and RA-8, located in the Pilcaniyeu Atomic Center. The last one is at present out of operation.

These reactors and their fuels were fabricated in Argentina, with the exception of the critical assembly RA-4 donated by Germany.

The reactor RA-3 was converted to reduced enrichment in the year 1989 using low enriched uranium (LEU) fuel elements developed in CNEA in the frame of the Program on Reduced Enrichment for Research and Test Reactors. At present the reactor RA-6 is the only reactor in Argentina using highly enriched uranium (HEU) fuel, the feasibility of its conversion to LEU is currently under analysis.

3. MANAGEMENT OF RESEARCH REACTOR SPENT FUEL

3.1. Interim Storage

Table I shows the main characteristics of the fuel elements of the research reactors and the critical assemblies, as well as the inventory and storage condition of the irradiated and spent fuels (SF).

Table I:
Research Reactors, Fuel Elements Characteristics and Inventory of Irradiated and Spent Fuel (*)

Reactor	Thermal Power	Type of Fuel	Cladd and Fuel Material	U-235 %	Qty. in Core	Irradiated Fuel and Spent Fuel Storage	
						AR	AFR
RA-0 (1965)	Critical Assembly	Rod L=680 mm $\phi=9,3\text{mm}$	Al Graphite + UO_2	19.7	183	17 (dry)	-----
RA-1 (1958)	100 kW	Rod L=680 mm $\phi=9,3\text{mm}$	Al Graphite + UO_2	19.7	223	5 (dry)	SF 232 (wet)
RA-3 (1967)	5 MW	MTR (Plate)	Al Al- U_3O_8	19.7	25	SF 7 (wet)	SF 68 (wet)
RA-4 (1972)	Critical Assembly	Annular $\phi(e)=240\text{mm}$ $\phi(i)=200\text{mm}$	U_3O_8 Dispersed in Polyethylene	19.7	7	5 (dry)	-----
RA-6 (1982)	500 kW	MTR (Plate)	Al Al-U alloy	90	30	12 (wet)	-----
RA-8 (1997)	Critical Assembly	Rods L= 1050 mm $\phi=9\text{ mm}$	Zy-4 UO_2 -pellets	1.8	Empty	1500 (dry)	-----

(*) Status at October 25, 2002

Due to the power of the RR and the particular mode of their utilization, only the reactor RA-3 consumes fuel elements (FE), at the current power of 5 MW its consumption is approximately one FE per month, this quantity will be doubled in the near future due to an increase in power to 10 MW. Therefore, in the management of RRSF the principal issue is the spent fuel generated by the reactor RA-3, so the following of the paper will be devoted to this type of spent fuel.

After a cooling period in the reactor decay pool, the SF from the reactor RA-3 is moved to an interim tube-type wet storage facility, located in the Ezeiza Atomic Center, away from the reactor site. This facility consists in a grid of vertical underground stainless steel tubes filled with

water and connected among them through the bottoms and tops. Each tube measures 2.1 m in length and 0.14 m in diameter, having capacity for two RRSF, so that the whole facility has capacity for 396 RRSF. At present there are stored 68 LEU spent fuel elements discharged from the reactor RA-3. A total of 207 HEU spent fuel elements generated by the reactor RA-3 before its conversion to reduced enrichment was shipped to USA in December 2000, under the frame of the US-DOE Foreign Research Reactor/Domestic Research Reactor Receipt Program (1).

The results of the inspection of the RRSF shipped to USA showed the occurrence of surface corrosion on some fuel plates, moreover, the ongoing monitoring program confirmed this phenomenon on aluminum probes installed in this wet storage facility. The occurrence of this phenomenon is related to difficulties in implementing a proper chemical quality control of the water due to the fact that it is difficult to attain an adequate flow of water through all the tubes.

To avoid the above-mentioned phenomenon, there is under study the feasibility of using a pool-type wet storage facility by adapting a pool located nearby the hot cells in the Ezeiza Atomic Center. This pool is at present not in use, it is made of concrete with stainless steel lining, and its dimensions are 2.7 m long, 1.3 m wide and 16 m deep. Complementary, there is under study the feasibility of changing the purpose of the actual wet storage facility by adapting it for dry storage; to this end, the tubes would have to be decontaminated, dried and conditioned accordingly.

If both of the above projects can be accomplished, after the cooling period in the reactor decay pool, the RRSF would be transferred to the pool-type wet storage facility for complementary cooling. From here they would be transferred to the adjacent hot cells where they would be disassembled and the loose fuel plates would be dried and then canned in air-tight canisters. Afterwards, the canisters, containing the plates of up to two RRSF, would be transferred to the tube-type dry storage facility.

3.2. Disposal

Assuming 20 additional years of operation of the reactor RA-3 at 10 MW, at the end of its life will be accumulated about 600 RRSF containing approximately 815 kg of uranium and plutonium (amounting approximately 92 kg of U-235 and 3.7 kg of Pu-239). In this case, there would be no economic incentive to recover the fissile material by reprocessing as could be the case for the SF of the nuclear power plants (NPP) Atucha-1 and Embalse.

The disposal of the conditioned RRSF will be in the deep geological repository to be built for the disposal of the NPP spent fuels or the high-level waste that would be generated in the case of their reprocessing. According to the current planning, the deep geological repository would be in operation in the year 2050.

3.3. Conditioning of RRSF for Disposal

The spent fuels of the reactor RA-3 have a relatively high remnant enrichment at discharge (about 11 % U-235), for reasons of non-proliferation during interim dry storage as well as for criticality control in deep the geological repository, it is considered convenient to make an isotopic dilution of the fuel material by blending it with depleted or natural uranium.

For this purpose could be applied a process like the "Melt and Dilute" developed in the Savannah River Laboratories at the USA. Conceptually, this process comprises the melting of the whole RRSF at a temperature of about 800 °C, the addition of depleted metallic uranium for isotopic dilution and aluminum to reach the eutectic composition in the Al-U alloy. The casting of this melt in a suitable mold produces an Al-U ingot that can be regarded as a conditioned RRSF material inside its container. This container, properly sealed, is ready to be packaged for its disposal in a deep geological repository. The main advantages of this process are its relative simplicity and the minor generation of secondary waste. The disadvantage is that it produces a conditioned RRSF material with poor corrosion resistance, demanding additional engineered barriers to comply with the safety requirements of the deep geological repository.

There are some processes under study to develop a RRSF conditioning method resulting in a glass or ceramic material. In general these processes include the following principal steps:

- Step 1: Decladding of the RRSF plate by controlled selective chemical separation of the aluminum clad.
- Step 2: Dissolution of the meat and blending with depleted or natural uranium for isotopic dilution.
- Step 3: Drying (if needed) and oxidation.
- Step 4: Blending with glass or ceramic powder and sintering.

Steps 1 and 2 can be accomplished using a wet or dry chemical process. The wet chemical process can be done by dissolution in sodium hydroxide. This is a well-known route that is at present applied for the dissolution of irradiated miniplates for the production of Mo-99. However, this route has the disadvantage of producing relatively large amount of secondary liquid waste, needing further conditioning. Therefore, an alternative dry chemical process is under development, it consists in decladding by selective chlorination in which the aluminum clad is removed as a gas (Al_2Cl_6) at a temperature of about 200 °C and the meat remains in solid form. The aluminum chloride is condensed and oxidized. Afterwards, the meat is dissolved, either with sodium hydroxide (wet route) or with chlorine gas (dry route) and blended with a chemically equivalent solution containing depleted or natural uranium. Then the final solution is oxidized at a temperature of about 600°C and the oxides are vitrified.

The use of the dry route reduces the volume to be treated by more than one order of magnitude in comparison with the aqueous-based process (2). This dry chemical process, named "HALOX" (Halogenation-Oxidation), is currently under development in CNEA (3-5) as well as the process of vitrification by sintering (6).

Another alternative process is also under study (7,8), it could be applied either to the meat after decladding or to the fuel plate without decladding, it consists essentially on the bulk oxidation of the material followed by blending with depleted or natural U_3O_8 . The resultant oxide blend is then sinterized to obtain U_3O_8 pellets or blocks. This material has an acceptable leaching rate in water and could be properly canned for disposal in a deep geological repository.

All these studies are carried out in a laboratory scale with non-irradiated miniplates of uranium silicide, although the processes can be applied as well to fuel plates with other uranium compounds. Special attention is given to the handling of the off gas and the volatiles elements generated during the high temperature treatments.

In principle, all the methods outlined above (including Melt and Dilute) are considered for the conditioning of the RRSF. The ongoing research is aimed at the development of the necessary knowledge to select a definitive route. The decision will be based on a combination of the following factors:

- Simplicity of the process and feasibility of its implementation in hot cells.
- Quality of the conditioned RRSF material, in regard principally to leaching rates in water.
- Volume of the conditioned RRSF material, in relation to the starting volume of the fuel plates.
- Cost

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