SPENT FUEL ASSESSMENT, ISOLATION OF LEAKERS, STUDIES WITH CORROSION COUPONS AND PREPARATIONS FOR SHIPMENT ABROAD

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Introduction.

IEA-R1 is a pool type, light water moderated, and graphite reflected research reactor. It is located at the "Instituto de Pesquisas Energéticas e Nucleares" IPEN, which is part of the Brazilian Nuclear Energy Commission. This reactor was designed and built by Babcok & Wilcox Co. in accordance with specifications furnished by the Brazilian Nuclear Energy Commission, and financed by the US Atoms for Peace Program. The first criticality in this reactor occurred on September 16th, 1957, and was the first such event in South America. Although designed to operate at 5 MW, IEA-R1 has been operating at 2 MW since the beginning. This reactor has been used for research in nuclear and solid state physics, radiochemistry and radiobiology, production of some radioisotopes and to provide irradiation services to the scientific community and also to industry.

Since startup, 181 core configurations have been installed and around 150 fuel assemblies (FAs) used. The reactor operated 40 hours per week (8 hours/day) during most of its life time, but since 1996 is operating in one continuous cycle of 64 hours per week. In terms of the fuel used in this reactor, four stages can be visualized:

The first stage corresponds to the first core of the reactor. It was composed of U-Al alloy fuel with 20wt% enrichment. Each fuel had 19 curved fuel plates produced by B&W. These FAs failed in the very early stage of the reactor operation, due to pitting corrosion caused by the brazing flux used to fix the fuel plates to the support plates. These fuels were replaced, in 1958, by new ones, also produced by B&W. This second batch of FAs were identical to the earlier ones (U-Al alloy, 20wt% enrichment, 19 curved fuel plates) but brazing was not used for assembling. The fuel plates were fixed mechanically to the support plates. These fuels performed well up to the discharge burnup used at that time.

The second stage corresponds to complete substitution of the core. Fuel bought from UNC (USA) and made with U-Al alloy, 93 wt% enrichment and 18 flat fuel plates. At this time the core was converted from LEU to HEU. In the middle of this cycle the control rods were changed from rod type to fork type (plate type). The control FAs were made by CERCA (France), using U-Al alloy, 93 wt% enrichment, and flat plates.

The third stage is characterized by restrictions in HEU fuel supply. IPEN bought 5 FAs from NUKEM (Germany). These were the UAl_x-Al dispersion type, with 20wt% enrichment and had 18 flat fuel plates per fuel element assembly. The amount of ^{235}U in the LEU fuel plate was almost the same as the HEU fuel plate and the geometry of the fuel element assembly was the same. At this stage with partial LEU core load, the HEU fuels, still in the core, began to have higher burnup and the number of FAs used in the reactor core increased to meet reactivity needs.

The fourth stage began with IPEN's decision to fabricate its own fuel and to replace gradually the high burnup HEU fuel in the core. IPEN had at that time sufficient knowledge and experience in core engineering, fuel engineering and fuel fabrication to produce MTR fuels for the IEA-R1. The IPEN fuels are U_3O_8-Al dispersion type, with 20wt% enrichment and geometrically identical to the LEU fuel from the third cycle. Table I summarizes the different FAs used in IEA-R1 core.

The dry storage is located in the first floor of the reactor building and consists of horizontal silos in a concrete wall. The spent FAs in dry storage were those of the first load that had presented corrosion and fission products release in the early stages of reactor operation. Their burnups were almost nil, but some of them had more than 1 R/h of dose rate at the FA surface. These FAs were wrapped in plastic bags.

The reactor pool is divided into two sections. The first section is the core pool, where the core and irradiation facilities are located. The second sector is the spent fuel pool, where the spent fuel storage racks are located. The FAs in the pool were the rest, used until recently. Some of these fuels had been in the pool for almost 40 years, and 30 of these, in the spent fuel racks. Some of these fuels revealed pitting corrosion nodules to unaided eyes. The pool water quality was and continues to be excellent. pH is always maintained between 5.5 and 6.5, the conductivity below 2 μS/cm and chloride content, less than 0.5 ppm.

Along the years, radiochemical analysis of the pool water showed low ^{137}Cs activity (less than 5 Bq/l), indicating that there were some leaking FAs. This activity was also low because the water cleaning system was always in operation.

It is possible that these corrosion nodules could have reached the fuel plate meat, exposing fission products to the pool water.

In the 70's the reactor pool wall lining was changed from ceramic to stainless steel and the spent fuel rack, from aluminum alloy to stainless steel. This was done at that time without considering the possibility of galvanic corrosion between the FA and the storage rack.

Over the last 10-15 years, increasing concerns about the large number of SFAs in various storage basins around the world led the US-DOE to offer to take back the fuels of US origin. The IAEA initiated the Co-ordinated Research Project (CRP) on the corrosion of aluminium clad spent research reactor fuels to help evaluate the state of the SFAs and to orient the pool/basin operators regarding maintanance/housekeeping procedures to extend the lives of the SFAs. In this context, IPEN opted to accept the US-DOE offer to take back the SFAs and also joined the CRP. Sections I, II and II of this paper detail the activities related to the assessment of spent fuels, corrosion studies with coupons and the shipment of SFAs to USA.

Section-I

I. FUEL ASSESSMENT

Having observed pits and nodules on the outer fuel plates of some FAs and being concerned about storing about 130 FAs in both dry and wet storage for extended periods, it was important to assess the state of the FAs

I.1. Visual inspection

The visual inspection of FAs was originally carried out with unaided eyes. The FAs were held at a depth of 2 meters in the pool. This inspection helped locate corrosion pits on the outer surface of the external fuel plates and their distribution. The surfaces of the internal fuel plates could not be examined. The inspected FAs were the LEU fuels of the second core, first stage, (I.D number IEA-41 through IEA-80) and the HEU fuels of the second stage (I.D number 81 through 118). Pits were observed on many FAs: (a) At specific regions on the fuel plate along the interface with the side plate. These regions were away from the fuel plate meat region; (b) On some FAs, the corrosion pits were on the fuel plates at regions close to the stainless steel rack; (c) On LEU FAs with curved fuel plates, pits were found on the convex side (side that contacted the stainless steel (SS) rack) and very few pits on the concave side (that did not have contact with the rack). This

provided evidence of pitting corrosion by galvanic action with SS; (d) The lateral support plates showed few corrosion pits in some FAs and most of these, at regions in contact with the rack. Table II summarizes the results of visual inspection.

The main reason for pitting corrosion was galvanic corrosion between the FA and the SS support rack. The fuel plate cladding (Al 1060) and the side plate (Al 6061 T6) also formed a galvanic pair, and the corrosion associated with this pair was of reduced intensity.

Many FAs were inspected later with an underwater video camera. This was carried out with the DOE-Savannah River group of personnel. The pitting corrosion nodules, their sizes, distribution and location were investigated.

I.2. SIPPING TESTS

All the FAs in wet storage were tested, one by one to determine whether breach of clading had occured. To perform the sipping test, an irradiated FA was withdrawn from the spent fuel storage rack, a rigid plastic tube connected to its bottom nozzle, and the FA, inserted inside an aluminum sipping tube (120 mm of diameter, 3 m length, ~ 33 l of volume), as shown in Figure 1. This was done with the FA positioned approximately2 meters below the pool surface. Before the test, the sipping tube was rinsed with demineralized water to reduce contamination from residual radionuclides (mainly ^{24}Na). The sipping tube with the FA inside it was then lifted up and the top end of the tube maintained above the surface of the water. It was then fixed to the pool bridge with a nylon rope. (Figure 1b) About 150 liters of demineralized water was then injected through the plastic pipe and flushed through the FA to rinse it. Soon after a sample of the tube water was collected as a background for gamma -ray spectrometric analysis.

The fuel assembly was then left undisturbed in the sipping tube for four hours. Compressed air was then injected through the plastic pipe and flushed through the FA for two minutes to homogenize the solution that could contain fission products released by the (leaking) FA. A sample of this water was collected for gamma-ray spectrometry analysis. A sample of the pool water was also collected to serve as a general background and gamma-rayspectrometric analysis carried out.

The gamma-ray spectrometric analysis was carried out with a shielded ORTEC HPGe detector. Data acquisition was performed with a ORTEC multichannel analyzer system coupled to a microcomputer through a control interface.Details about spectrometric analysis, the detector and the determination of specific ^{137}Cs acivity can be found elsewhere [1].

The main observations from the gamma-ray spectrometric analysis were: (a) some

fuel assemblies showed the ^{137}Cs photopeak; (b) fuel assemblies for which two water samples were takeni showed an ncrease in activity with time. This indicated that the fuel assembly was leaking. It is possible to determine the leak rate of ^{137}Cs for the FA. For example, for the fuel assembly IEA-53 this rate was ~0.2 Bq/l.min or ~14 Bq/min. Figure 2 shows the specific activity for each fuel assembly in the sipping test. Some fuel assemblies showed leaks, and most of the FAs had specific activities lower than 30 Bq/l (water residence time of 4 hours). Figures 3 and 4 show the gamma spectra for a non-leaking and a leaking FA respectively.

Table III shows a correlation between visual inspection and the sipping test results. The older LEU FAs were in a worse condition than the HEU FAs, and only a few indicated leak of ^{137}Cs.

Water samples from the sipping tests with the highest activity (indicating leaks) were analyzed by x-ray fluorescence spectrometry for Cs and U. Cs and U were not detected and only ions corresponding to the corrosion of aluminium and stainless steels were observed. This was due to Cs levels being below detection limits. ($\sim 3 \times 10^{-11}$ g Cs/l corresponds to 100 Bq/l in the water) U was not detected by gamma ray spectrometric analysis or by X-ray spectrometry.

Attempts were made to determine the ^{137}Cs leak rate. This was done with FAs that contained corrosion pit nodules. The oxide covering the pit (the nodule) was removed and the sipping test repeated. No differences were observed in the ^{137}Cs activity levels in the sipping tests, carried out before and after removal of the nodules on the FA.

FA IEA-53 had the highest leak rate, of 14 Bq/min, and this was less than the value established by DOE-SRS for canning leaking fuel assemblies. The DOE-SRS interim criteria was 13.57 μCi/hr per cask to be shipped, which is equivalent to 35.9 pCi/ml.hr for one fuel assembly (assuming 3.6 gallons of water per fuel assembly). The value obtained at IPEN in the sipping tests when corrected to the volume of SRS criteria, was 0.64 pCi/ml.hr (0.4 Bq/l.min.), well below the criteria limit. The DOE interpretation of Foreign Research Reactor Environmental Impact Statement (FRR.EIS) stipulated that any fuel that contained a thru-clad pit should be canned for shipment and storage. Eventhough the IPEN sipping test data met the DOE criteria, they imposed some restraints due to the corrosion pits and their distribution. According to this some of the FA had to be canned eventhough they passed the criteria. This requirement was subsequently withdrawn.

Gamma ray spectrometric analysis of a sample of a nodule on a fuel plate was carried out. Significant ^{137}Cs activity was found and reduced activities of ^{235}U, ^{155}Eu, and ^{154}Eu. This indicated thru-clad pitting. This also indicated that besides ^{137}Cs, U and Eu isotopes migrated from the meat to the nodule. U and Eu were

retained in the nodule and did not reach the water, especially since the water specimens from the sipping tests carried out before and after removal of the nodule did not reveal the presence of either of these two elements.

The conclusions from fuel assessment were:

(a) Visual inspection of the spent fuel assemblies in storage at the spent fuel pool showed pitting corrosion on the external fuel plates of many fuel assemblies. Those stored for almost 40 years were worse. The pitting corrosion was due to galvanic effects between the aluminum fuel plate cladding and the stainless steel spent fuel rack.

(b) The sipping test method and equipment used in IPEN were efficient for determining fission product leak from FAs.

(c) A ^{137}Cs leak rate of 14 Bq/min for the fuel assembly in the worst state was far bellow the DOE-SRS criteria for canning leaking MTR fuel assemblies.

(d) Gamma-ray spectrometry of pitting corrosion nodules indicated the presence of Cs, U and Eu isotopes. The ^{137}Cs activity was much higher than that in the sipped water specimen. U and Eu isotopes were not detected in the water. This confirmed the presence of thru-clad pits in some FA.

Section-II

II. Corrosion studies with coupons

The International Atomic Energy Agency's (IAEA) concerns about: (a) the state of SFAs, (b) insufficient data about the effects of pool parameters on fuel corrosion; and (c) the need to provide overall guidelines to spent fuel pool managers to undertake practices that would prevent corrosion and further deterioration of SFA,s resulted in the initiation of a Co-ordinated Research Programme (CRP) on "Corrosion of Research Reactor Al-clad Spent Fuel in Water" in 1996, with IPEN as one of the participants. This CRP is expected to go on till 2002. The main activities of this programme are related to exposing racks of Al alloy specimens in different spent fuel basins around the world. Five racks (figure5) were suspended in the IEA-R1 reactor pool, and subsequently withdrawn after different time spans to evaluate the extent of corrosion as a function of alloy composition, crevices, bi-metallic effects and water chemistry. During this period the pool water was monitored for pH, conductivity, chloride ion content and radiometry. Besides the CRP racks, at IPEN, another rack of specimens made from alloys that were site specific (figure 6) was also introduced in the IEA-R1 reactor spent fuel section.

The results of the first CRP rack from the different locations revealed that: (a) the water chemistry varied from very good to poor; (b) the corrosion rates of the specimens varied from very low to extremely high; (c) the crevices played an important role and (d) galvanic corrosion was quite high.

At the meeting held in IPEN in 1998, a Test Protocol was outlined in terms of specimen preparation, handling, length of test etc. The use of site specific alloys was also encouraged. New specimen racks were distributed to the participants for removal at regular. The results so far have indicated that: (a) pitting is the main form of corrosion; (b) crevice corrosion and galvanic effects also predominate; (c) reduction in conductivity and chloride ion content of the pool/basin water is essential to maintain low corrosion rates; (d) dust sediments on aluminium alloy specimens contribute to pit initiation.

The observations made from the IPEN rack of specimens after 16 months exposure to IEA-R1 reactor pool water in the spent fuel section can be summarized as:

- The uncoupled AA6061 and AA6262 specimens had fewer pits than uncoupled AA1060 specimens.
- The top surfaces of uncoupled specimens had more pits than their under sides, due to dust settling
- Crevice surfaces of 1060/1060, 6061/6061and 6061/1060 couples were stained but not pitted. (Figures 7)
- The surfaces of the aluminium alloys within the crevices of 6061/304SS and 6262/304SS couples were heavily pitted, indicating increased corrosion of aluminium coupled to stainless steels. The steel surfaces were only stained and revealed rust marks. (Figure 8)
- The 1060 surface within the crevice of 1060/304SS couple was stained but not pitted.

Section-III

Preparation and shipment of SFA

The following aspects require attention in preparation for shipment of SFAs.

(a) Knowing the local licensing and/or institutional approval procedures;
(b) Obtaining detailed information about the SFA: (i) HEU/LEU type: (ii) Quantity of fuel requiring stabilization etc;
(c) Providing the shippers, descriptions of fuel irradiation histories, discharge dates, mass, radiation levels or dose rates, heat loads, fuel manufacturers, drawings of the FAs and other pertinent information.

(d) Providing the transporters descriptions of the facilities that could be used to prepare and ship the SFA - physical description, maximum storage capacity, crane capacities, floor loading limits, dimensions of building access doors etc.

Characterization of the IPEN SFA in wet-storage involved the evaluation of corrosion and mechanical damage, and cesium release rates as mentioned earlier. The SFA were visually examined with the aid of an an under water camera and the level of ^{137} Cs release determined from sipping tests. A total of 66 high-enriched uranium (HEU) and low-enriched uranium (LEU) fuel assemblies were characterized. The results showed that 25 assemblies (HEU) did not contain corrosion pits into the fuel meat region. The ^{137} Cs release rates from all 66 of the assemblies were well-within the SRS interin criteria of 13.57 μCi/hr per cask shipment which is equivalent to 35.9 pCi/ml/hr based on the volume of water used in SIP tests.

MTR LEU fuel assemblies stored in dry storage were also visually examined. Some of these had leaked radionuclides in-reactor within months after initial service in 1957 due to corrosion attack at the brazed joints between the fuel plates and support plates of the assemblies. These assemblies had been in basin (wet) storage for 24 years followed by 15 years in dry storage at near room temperatures. 6 of the 40 SFAs were examined.

In December 1997 Edlow International Co. was awarded the contract by the U.S DOE to transport the SFA´s from research reactors in Brazil, Uruguay and Venezuela. The Edlow Team consisted of Edlow International Co. (EIC), Nuclear Cargo + Services (NCS), Transnucleaire (TN) and Science Applications International Corp. (SAIC). A Brazilian Company named TRION was contracted to provide the transport equipment, to design and constuct the saw tool to cut the control assemblies and to provide all necessary customs documents for the SFA´s return to USA. It was decided that the consortium would provide 4 transport casks (two GNS-11 and two GNS-16) and IPEN would provide the contract signature, exportation license, appendix A, Transport and Security Plans elaboration and safe guard documents.

In July 1998 the Brazilian authorities approved the export license, the Transport/ Security Plans and, validated GNS-11 and GNS-16 casks for SFA´s transportation in Brazilian territory. Later in july'98, six containers with 4 transport casks and other equipment arrived at the Santos Port in Brazil. Soon after, the US-DOE did not validate the GNS-16 casks to transport fuel assemblies with cladding defects greater than pin holes and hairline cracks. Therefore everything was postponed.

In Abril'99, Edlow International company was again given the contract to transport the assemblies. All the documents and /or approvals that had previously been

obtained were to be used. Between June and September 1999 the German and American authorities provided the revised certificates of GNS-16 and GNS-11 casks for failed assemblies transport. The Brazilian regulatory body provided, also, the validation of those casks to be used in Brazil.

In September'99, when authorization for loading the casks had been given by DOE, the IPEN reactor operation team started cutting the 19 control fuel assemblies. **Also in september** four containers with two GNS-11 German casks and equipment arrived at IPEN. With the help of a 25 ton crane, the equipment was placed on a iron platform located in front of the access hall and with the aid of a mobile lift, moved inside of the reactor building and positioned under the access door to the 3° level. Part of the equipment, like the transfer cask, primary lid, water tank and so on were lifted by the reactor's mobile 10 ton crane to the 3° level. The cask was moved inside the reactor building and positioned in the first level, under the access door to the 3° level.

On September 21 1999, the primary lid was removed from the GNS-11 cask and lifted to the 3° level. The rotary lid was positioned in the upper part of the cask and a cold test with a dummy element was made. A 4 ton transfer cask was used to transfer the assemblies from wet storage to the transport cask. The assemblies, were lifted one by one from the storage racks with a special tool and positioned inside a plastic tube located on a metalic plataform in the reactor pool at a depth of 2 meters. The transfer cask was immersed inside the reactor pool over the assembly to be transfered (see figure 9). A special tool picked the assembly and drew it inside the cask, which was lifted by the reactor crane and positioned above the rotary lid and transport cask located in the first level. Finally, the assembly was guided to one of the 33 positions of the cask. After a cask was fully loaded and prior to replacing the rotary lid with the primary lid, a water tank was positioned over the cask and filled with 4000 liters of water. After the cask was closed, the water was drained from it and from the water tank. Three water samples were taken from this cask after 0, 4 and 12 hours. A leak test was also performed.

This operation was repeated to transfer the 87 assemblies stored in the wet storage. The 40 assemblies stored in dry storage were taken one by one, by hand and placed within a lead shield. They were then lifted across and positioned directly inside the cask by an operator located at the 3° level of the reactor building. Four GNS casks were loaded with the 127 brazilian spent fuel assemblies. After decontamination the equipment were removed from the reactor building to the containers. The casks were stamped by people from ABACC and supervised by technicians from AIEA. On November 3, 1999 the containers were transported by road to Santos Port and from there by ship to SRS in USA.

References.

1. J.A.Perrotta, L.A.A.Terremoto and C.A. Zeituni, Experience on Wet Storage Spent Fuel Sipping at IEA-R1 Brazilian Research Reactor, Presented at TCM on Procedures and Techniques for the Management of Failed Fuels from Research and Test reactors, Budapest, Hungary, 29-31 October 1996.

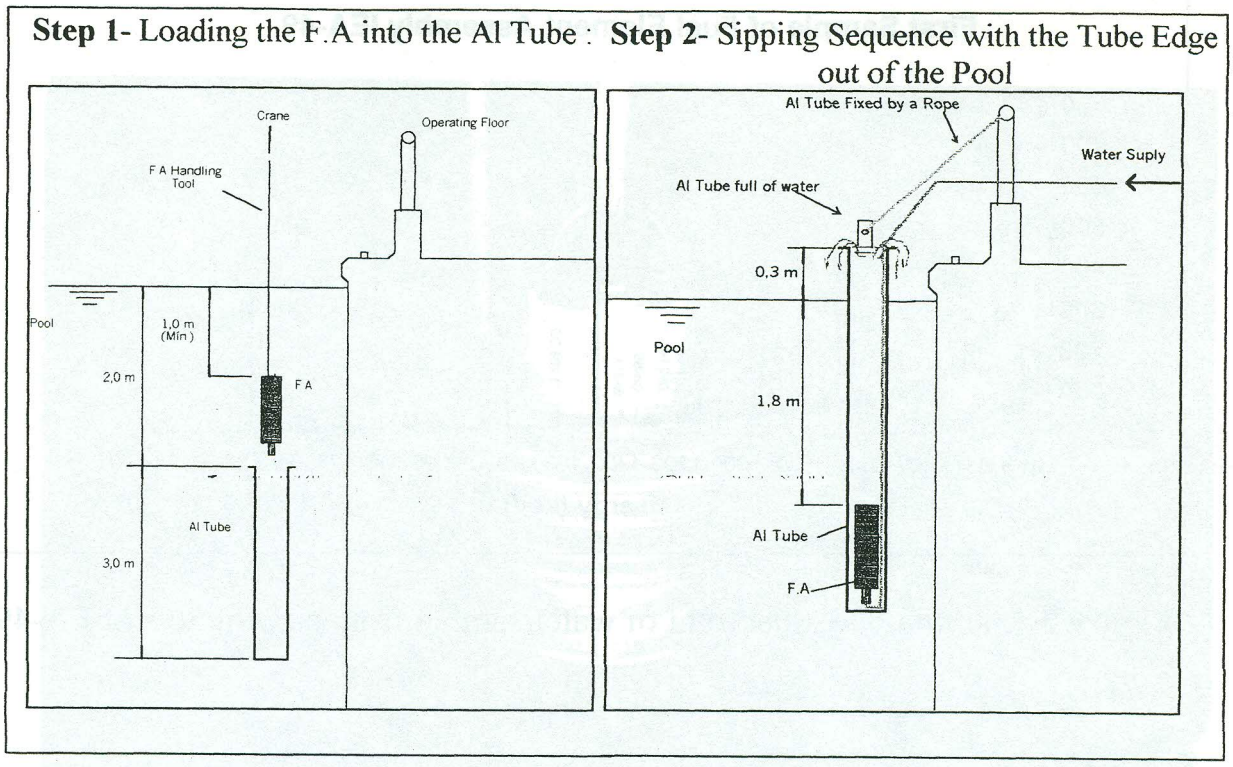


Figure 1 – Schematic illustration of Sipping Test.

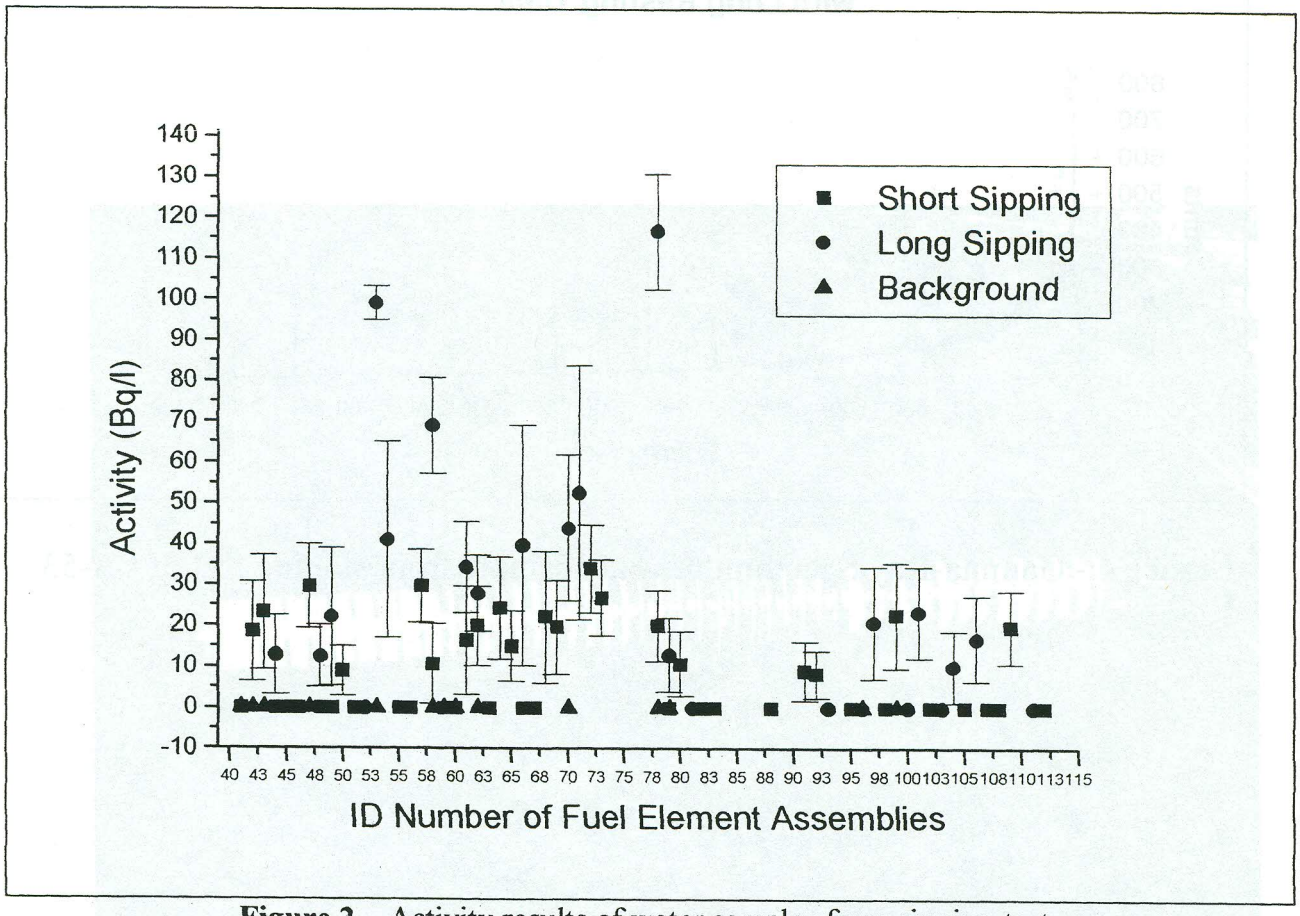


Figure 2 – Activity results of water samples from sipping tests

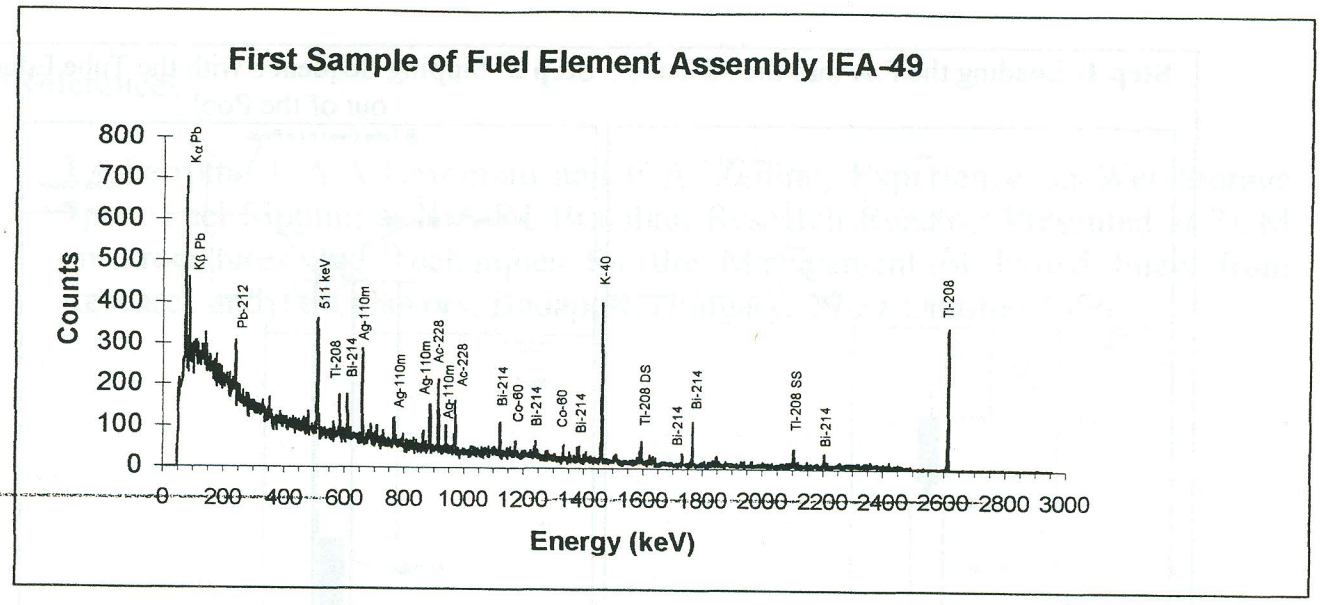


Figure 3. Gamma-Ray Spectrum of water sample from sipping test of FA-49

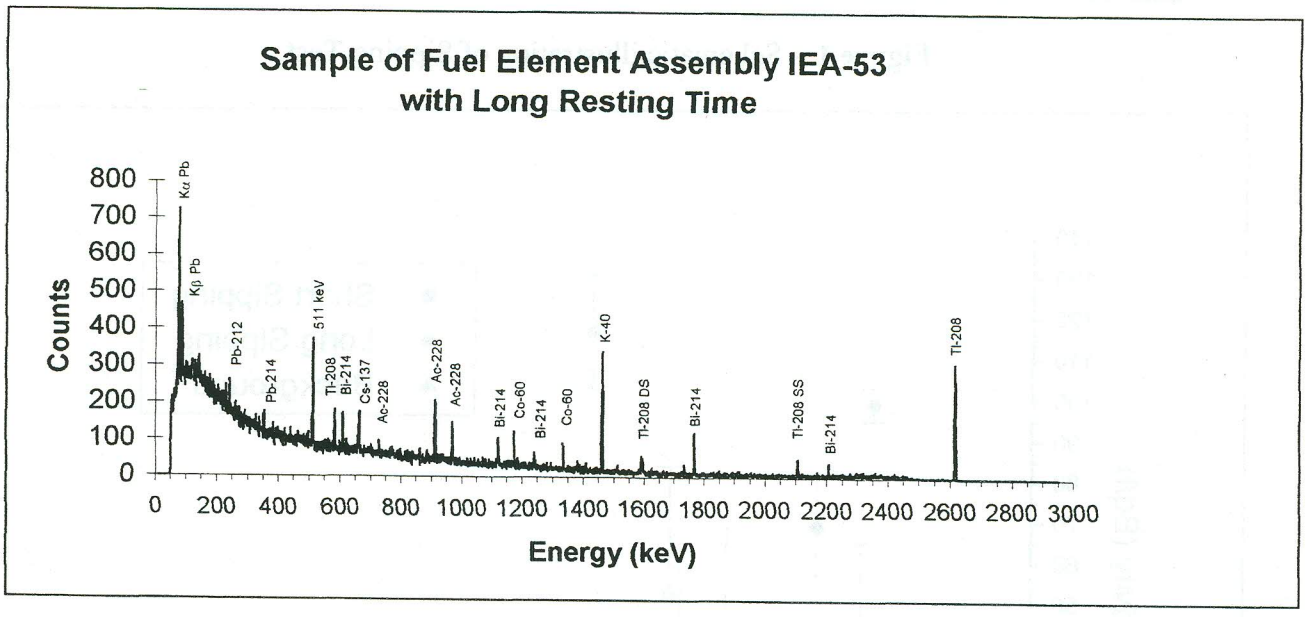
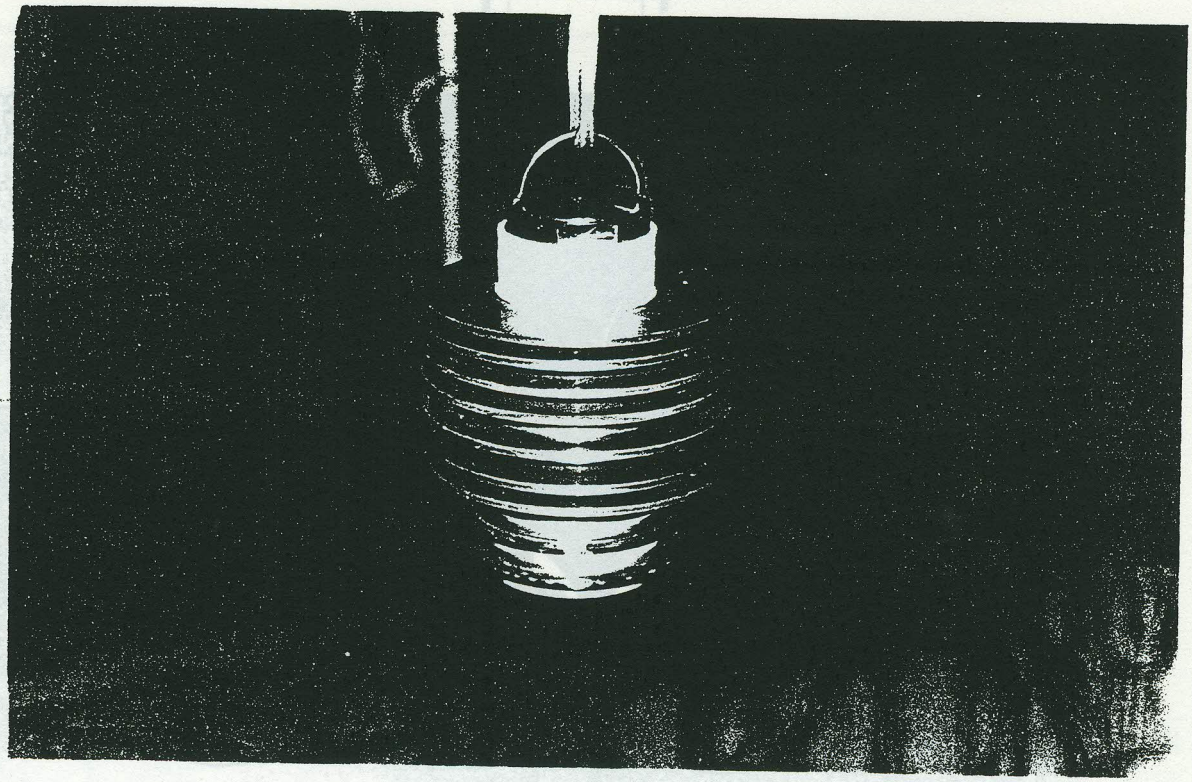
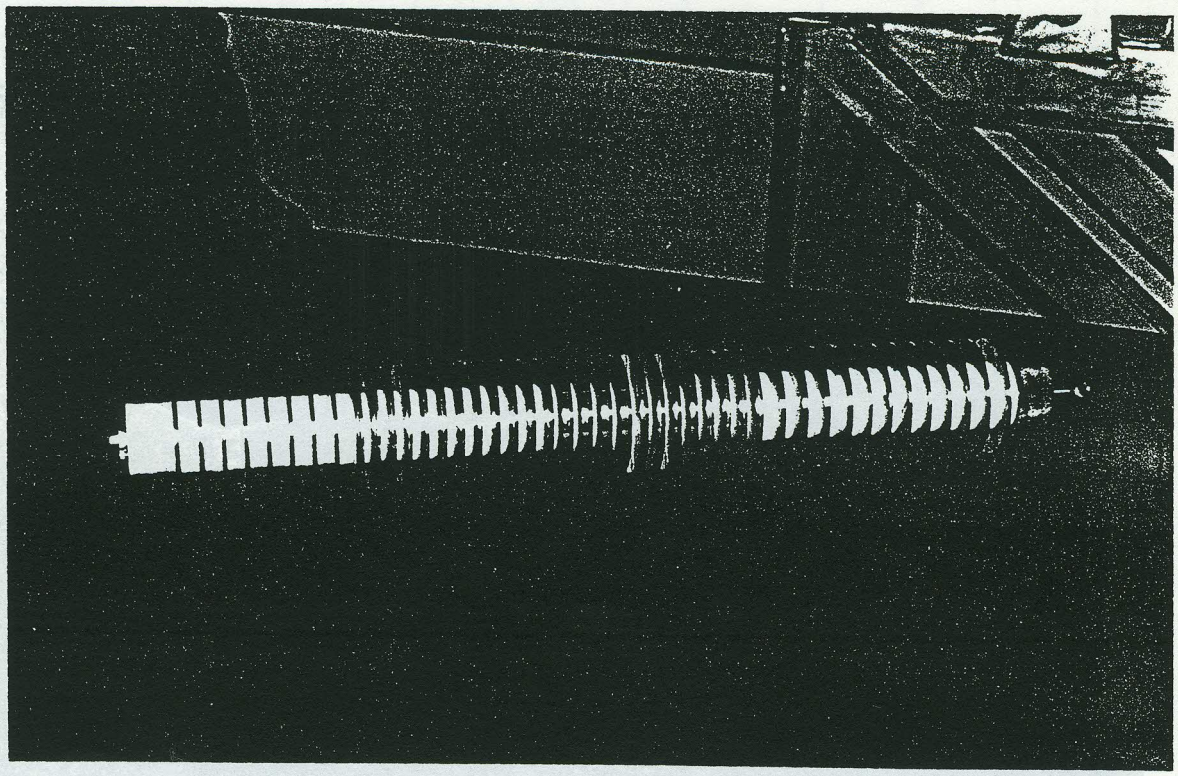
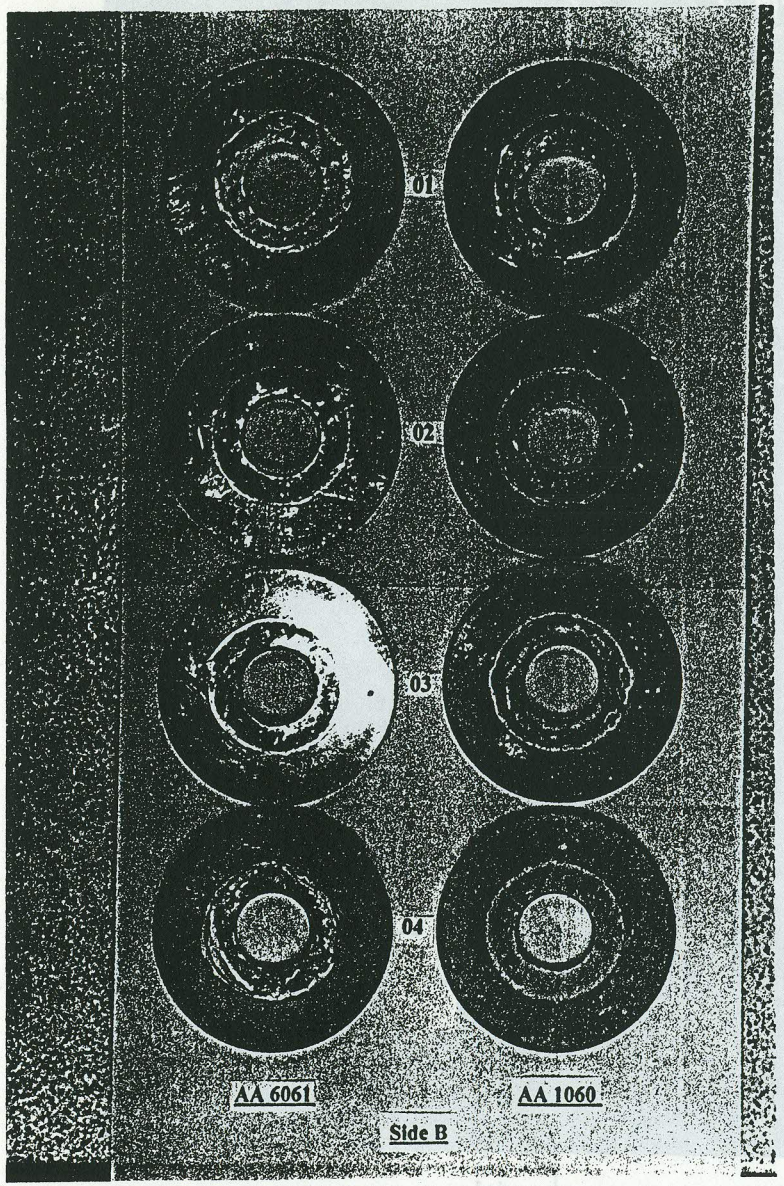


Figure 4 - Gamma-Ray Spectrum of water sample from sipping test of FA-53



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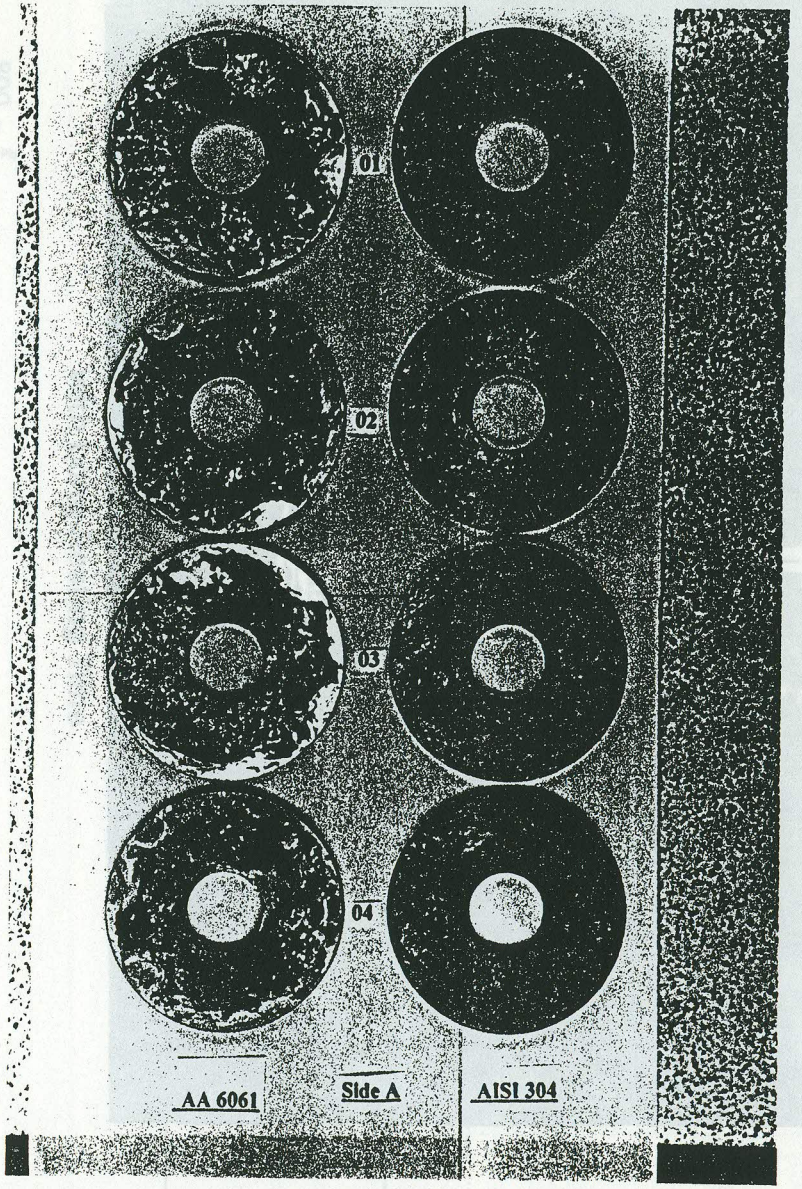
AA 1060
Side B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Side A
AISI 304

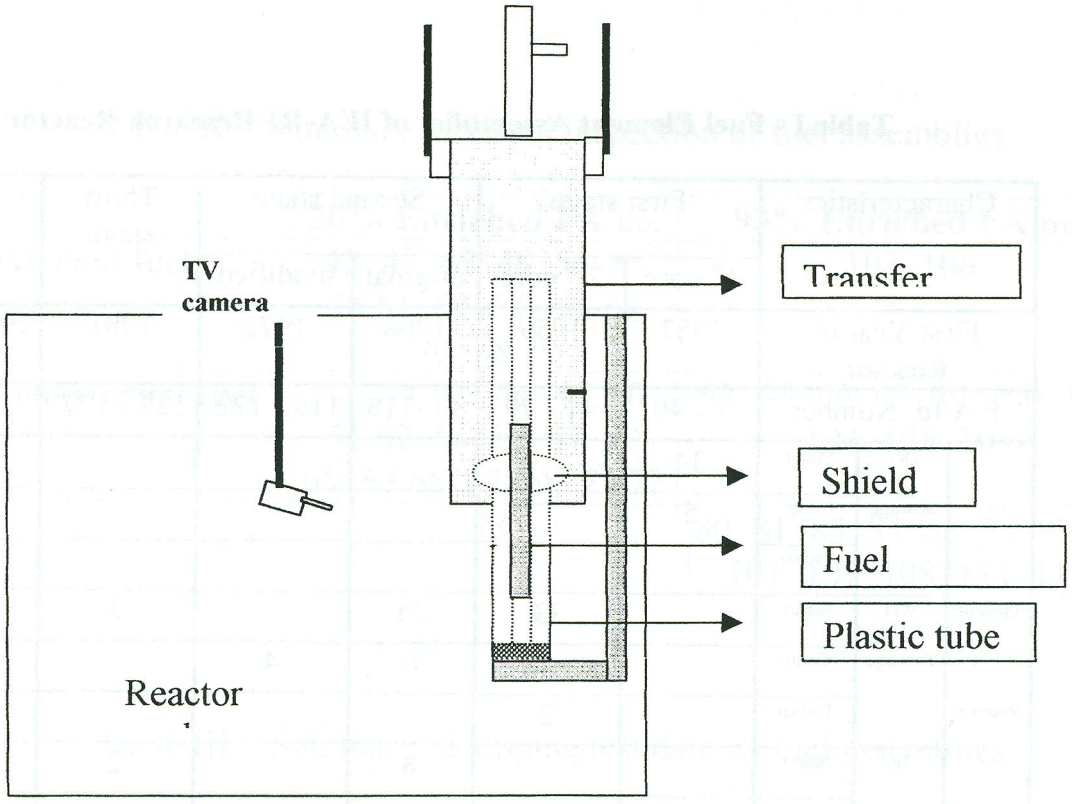

Figure 9. Schematic illustration of the transfer cask.

Table I - Fuel Element Assemblies of IEA-R1 Research Reactor

Characteristics			First stage		Second stage		Third stage	Fourth stage
			1st core	2nd core	original	modified		
First Year in Reactor			1957	1959	1968	1972	1981	1985(*)/1988
F.A Id. Number			1 - 40	41 - 79	80 -118	119 - 122	123 - 127	128 -
Present Position	Dry Storage	Stand.	34					
		Contr.	5					
		Partial	1					
	Wet Storage	Stand.		33	25		3	
		Contr.		4	6	4		
		Partial		2				2
	In Core	Stand.			8		2	16
		Contr.						4
		Partial						
Original Enrichment			20%	20%	93%	93%	20%	20%
Manufacturer			B&W (USA)	B&W (USA)	UNC (USA)	CERCA (France)	NUKEM (Germany)	IPEN (Brazil)
Fuel Type			U-Al alloy	U-Al alloy	U-Al alloy	U-Al alloy	UAl_x-Al	U_3O_8-Al
Number of plates per F.A	Standard		19	19	18		18	18
	Control		9	9	9	12		12
	Partial		10	9 / 10				2 / 10
Type of Fuel Plate			curved	curved	flat	flat	flat	flat
Burnup ($\%^{235}U$)	Control		0	~ 40		~ 50		~ 20(**)
	Partial		0	~10	~ 43			~ 20

(*) - Partial Fuel Element Assembly (**) - Up to September 1996.

Table II – Summary of visual inspection of fuel assemblies

	20% Enriched FA no.	**93% Enriched FA no.**
Pits on external fuel plates	42 43 48 49 53 55 58 61 62 64 66 69 70 78 79	103 106
Few pits onexternal fuel plates	41 44 45 46 47 50 51 52 54 56 57 59 60 63 65 67 68 71 72	93 95 96 97 99 100 102 104 105 109
No pits	73	80 81 83 84 88 91 92 98 101 107 108 111 112

Table III – Summary of sipping test data on fuel assemblies

	20% Enriched FA no.	**93% Enriched FA no.**
No indication of Cs-137	41 45 46 51 52 55 56 59 60 63 67	81 83 84 88 93 95 96 98 100 102 103 105 107 108 111 112
Low indication of Cs-137 (< 30 Bq/l)	42 43 44 47 48 49 50 57 62 64 65 66 68 69 73 79	80 91 92 97 99 101 104 106 109
Medium indication of Cs-137 (> 30 Bq/l: < 60 Bq/l)	54 61 70 71 72	----
High indication of Cs-137 (> 60 Bq/l)	53 58 78	----