Experience on Wet Storage Spent Fuel Sipping at IEA-R1 Brazilian Research Reactor

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Abstract

IEA-R1 Research Reactor of Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN-SP) is a pool type reactor of B&W design operating since 1957 at 2 MW of power. Irradiated fuels have been stored at the facility along the various years of operation. Nowadays there are 40 spent fuel assemblies at dry storage, 79 spent fuel assemblies at wet storage and 30 fuel assemblies in core. The oldest fuels are of USA origin, made with U-Al alloy both of LEU and HEU MTR fuel type. Many of these fuel assemblies present corrosion pits along lateral fuel plates. These pits have their origin by galvanic corrosion between fuel plate and stainless steel storage rack although the excellent pool water characteristic would inhibit this occurrence of corrosion. Radiological analysis of pool water have been indicating low activity of ^{137}Cs. According to the decision to send back the old fuels to USA, sipping tests with spent fuel assemblies were performed in order to evaluate their ^{137}Cs leaking rate, if any. This paper describes the procedure and methodology used to perform sipping test with the fuel assemblies at the storage pool, and presents the results obtained for the ^{137}Cs sipping water activity for each fuel assembly. Discussion is made correlating corrosion pits to the activity values measured. A ^{137}Cs leaking rate is determined which can be compared to the criteria established for canning spent fuel assemblies before shipment.

INTRODUCTION

The IEA-R1 is a pool type, light water moderated, and graphite reflected research reactor. It was designed and built by Babcok & Wilcox Co. in accordance with specification furnished by the Brazilian Nuclear Energy Commission, and financed by the US Atoms for Peace Program.

The first criticality occurred on September 16th, 1957, being the first criticality achieved in South America. Although designed to operate at 5 MW, IEA-R1 has been operating at 2 MW since its beginning. In these 39 years of operation IEA-R1 has been used to perform research in nuclear and solid state physics, radiochemistry and radiobiology, production of some radioisotopes and to give irradiation services to the scientific community and also industry.

Since startup to present time (September 1996), 181 core configurations have been installed and around 150 fuel element assemblies used. The reactor operated 40 hours per week (8 hours/day) in most of its life time, but since the beginning of this year is operating in one continuous cycle of 64 hours per week.

Concerning fuel utilization it is possible to analyze the reactor history in four cycles.

The first cycle corresponds to the first core of the reactor. It was composed of U-Al alloy fuel with 20wt% enrichment, having 19 curved fuel plates produced by B&W. These fuel assemblies failed at the earlier stages of the reactor operation, due to pitting corrosion caused by 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. They 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 operated with good performance up to the discharge burnup used at that time.

The second cycle corresponds to a complete substitution of the core. Fuel made with U-Al alloy, 93 wt% enrichment , having 18 flat fuel plates were bought from UNC (USA). At this time the core was converted from LEU to HEU. Some of these fuels are still operating in the core. In the middle of this cycle the control rod mechanical concept was also changed from rod type to fork type (plate type). The control fuel element assemblies were fabricated by CERCA (France), using U-Al alloy, 93 wt% enrichment, and flat plates.

The third cycle is characterized by the restriction of HEU fuel supply. IPEN bought, from NUKEM (Germany), 5 fuel element assemblies of UAl_x-Al dispersion type, with 20wt% enrichment and having 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. With this partial LEU core load, the HEU fuels, that stayed in core, began to have higher burnup and the numbers of fuel element assemblies used in the reactor core had to be increased due to reactivity needs.

The fourth cycle has began with IPEN decision of fabricating its own fuel and to replace, gradually, the high burnup HEU fuels in the core. IPEN had already, at that time, good knowledge and experience in core engineering, fuel engineering and fuel fabrication, so the decision to produce MTR fuels to the IEA-R1 was a natural way to maintain the reactor in operation. The IPEN fuels are of U_3O_8-Al dispersion type, with 20wt% enrichment and geometrically identical to LEU fuel from the third cycle.

Table 1 summarizes the different fuel element assemblies used in IEA-R1 core. It also shows the present position of these fuels in the facility: spent fuel dry storage, spent fuel pool, reactor core.

Table 1 - Fuel Element Assemblies of IEA-R1 Research Reactor

Characteristics			First Cycle		Second Cycle		Third Cycle	Fourth Cycle
			1st core	2nd core	original	modified		
First Year in Reactor			1957	1959	1968	1972	1981	1985(*)/1988
F.A Id. Number			1 to 40	41 to 79	80 to 118	119 to 122	123 to 127	128 and so on
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
Dimensions (mm)	plate th.		1.37	1.37	1.52	1.52	1.52	1.52
	meat th.		0.61	0.61	0.51	0.51	0.76	0.76
	cladding		0.38	0.38	0.505	0.505	0.38	0.38
	active w.		63.5	63.5	63.5	63.5	60.35(min)	60.35(min)
	active l.		597	597	597	597	590 (min)	590 (min)
Grams of ^{235}U per F.A	Standard		159	159	186		180	180
	Control		76	76	90	130		120
	Partial		87	76 / 84				20 / 100
F.A Max. Burnup ($\%^{235}U$)	Standard		0	~ 30	~ 50		~ 50	~ 30(**)
	Control		0	~ 40		~ 50		~ 20(**)
	Partial		0	~10	~ 43			~ 20

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

The dry storage is located at the first floor of the reactor building and is composed by horizontal silos at a concrete wall.

The reactor pool is divided in 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 laid. Figure 1 shows the rector pool with the two sections.

The spent fuel assemblies in dry storage are those of the first load (first cycle) that presented corrosion and fission products release at the earlier stages of reactor operation. Their burnups are almost zero, but some of them have more than 1 R/h of dose rate at the fuel assembly surface. These fuel assemblies have plastic bags wrapping them, and the discussion of canning or not them before shipment has to be done with the shipper and the receiving facility.

The fuel assemblies in the pool are all those used up to now except the first load. Some of these spent fuels are almost 40 years inside the pool and more than 30 years inside the spent fuel racks. Some of these fuels present pitting corrosion nodules, visible by nude eyes, and these corrosion nodules may reach the fuel plate meat, exposing fission products to the pool water.

Pool water radiochemistry analysis, along the various years of sampling, have been showing a low ^{137}Cs activity (less than 5 Bq/l). This means that there are some leaking fuel assemblies. This activity is also low because there is a constant water cleaning system in operation.

The pool water quality is excellent. pH is ever kept between 5.5 and 6.5, conductivity is below 2 μS/cm and chlorides are less than 0.5 ppm. Although these excellent water characteristics the old fuel assemblies show pitting corrosion. This is due to galvanic corrosion because the spent fuel racks are made of stainless steel and the fuel cladding is made of aluminum. In the 70's the reactor pool wall was changed from ceramic to stainless steel liner and also the spent fuel rack was changed from aluminum alloy to stainless steel. This was done at that time not taking into account the possibility of galvanic corrosion between fuel assembly and storage rack.

During the next ten years, all the spent fuels of American origin have to be sent back to the USA. Last June, a US-DOE staff came to IPEN to discuss the shipment of these fuels. One of the points of discussion was the necessity of canning leaking spent fuels. The proposal of DOE staff was to perform sipping test inside the shipping cask, with all fuels loaded. IPEN pointed out that this wouldn't be a good solution because if any cesium release were detected, all fuel assemblies would be sent back to the pool storage and an investigation would have to be done to identify the leaking fuel assembly. IPEN proposed to perform individual sipping test of each fuel assembly before transference to the shipping cask. As the schedule for shipment was very tight, sipping tests had to be done quickly. IPEN performed visual inspection and sipping tests in 60 spent fuel assemblies in two weeks of working, and did the selection of the fuel assemblies that could be shipped without canning.

This work was verified, in late July, by another group from DOE-Savannah River Site that came to IPEN. This group agreed with IPEN methodology and results for the sipping tests. They asked for additional sipping tests to confirm the results, performed some visual inspection of fuel assemblies using underwater camera, and also visual inspection of some fuels at dry storage.

This paper describes this work done by IPEN which objective was to identify the fuel assemblies that could be sent back to US, at the first shipment from South America, without canning[1]. It is not presented at this paper the visual inspections performed together with DOE-SRS group.

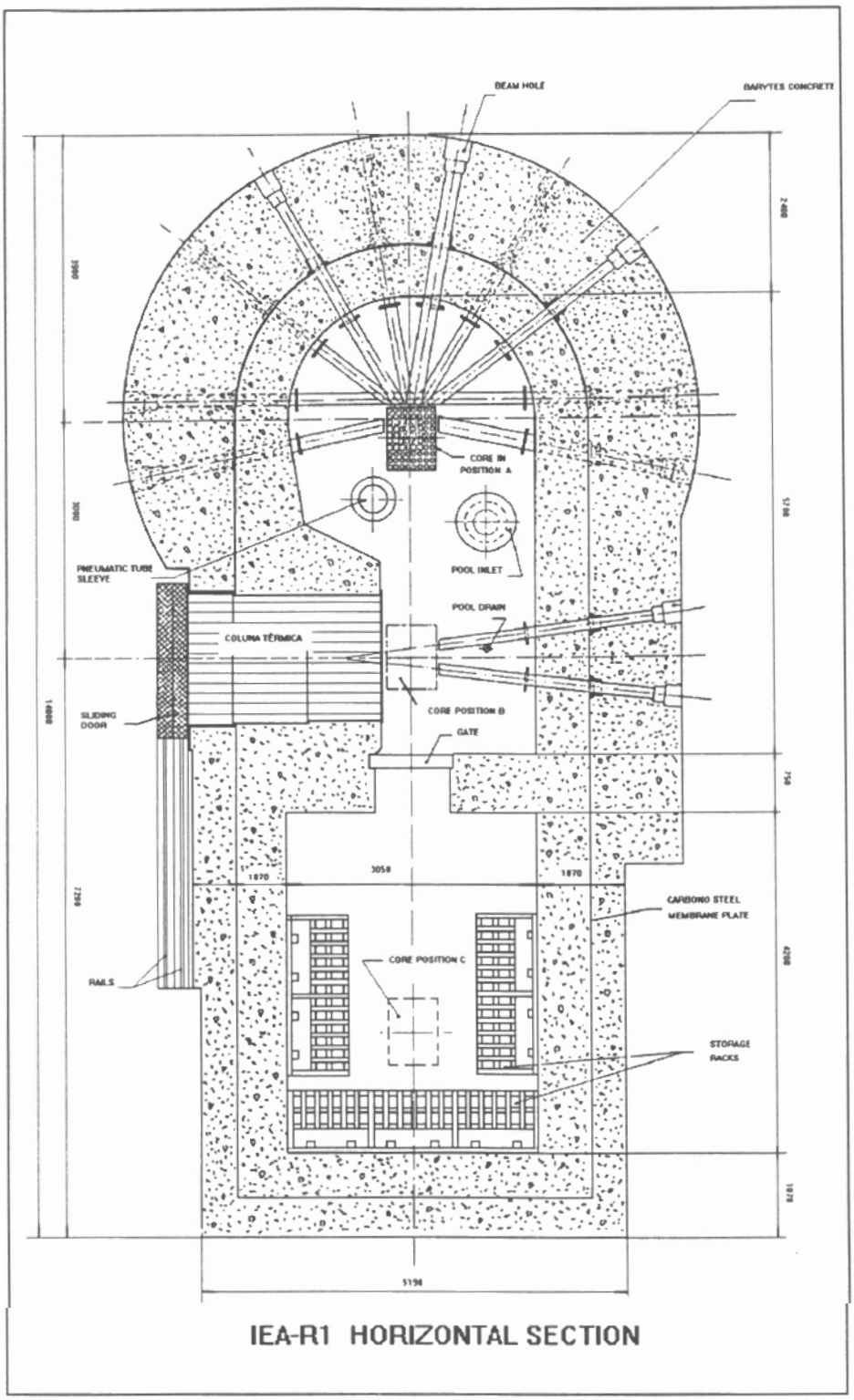

Figure 1

VISUAL INSPECTION

The visual inspection was done by nude eyes with the fuel assembly inside the pool with 2 meters of depth. As IPEN didn't have any underwater camera, this visual inspection was done just to verify if was there any visual corrosion pit at the outer surface of outer fuel

plates and the pattern of this occurrence. The inspected fuel assemblies were the LEU fuels of the second core, first cycle, (I.D number IEA-41 through IEA-80) and the HEU fuels of the second cycle (I.D number 81 through 118).

In many fuel assemblies was observed the occurrence of corrosion pits. Some patterns observed are:

- there are corrosion pits along the interface of external fuel plate with the side plate. The pits seem to be localized at the fuel plate side. These regions normally are out of the fuel plate active width, although there is no way to confirm if the corrosion pit is deep enough through the fuel plate meat;

- there are some fuel assemblies with corrosion pits along the fuel plates in defined regions of the plate where there is contact between the fuel plate surface and the spent rack frame surface;

- some fuel assemblies show corrosion pits along the height of the external fuel plate. Observing the position of these fuel assemblies in the spent fuel rack it is noticed that they are located at corner positions in the rack where there are close contact of the fuel assembly with the rack frame;

- LEU fuel assemblies, having curved fuel plates, show corrosion pits at the convex plates (that have contact with the rack frame) and show very few corrosion pits at the concave fuel plates (that have no contact with the rack frame). This is a very strong evidence of pitting corrosion by galvanic phenomena that is occurring to the fuel assemblies;

- the lateral support plates show few corrosion pits in some fuel assemblies and most of them in the region where exists contact with the rack frame;

- as the visual inspection did not used any equipment, it was not possible to see any corrosion occurrence at the internal fuel plates of the fuel assemblies. The visual observation is based only on the external fuel plates.

From these visual observations it was concluded that the main reason of the pitting corrosion existence is the galvanic pair existing between fuel assembly and support rack. The material of fuel plate cladding is Al 1060, the side plate is Al 6061 T6 , and the support rack is SS AISI 304. Also there is some galvanic pair between fuel plate cladding and side plate, but this effect could be not so strong if the stainless steel rack would not be in contact with side plate or fuel plate.

Table 2 shows a resume of the visual inspection.

IPEN had already some experimental results of pitting corrosion in a fresh fuel assembly laid inside the spent fuel rack. It was observed that the external fuel plates showed corrosion pits at points where there was contact between fuel plate and the stainless steel support rack frame. The side plate showed corrosion pits, at the same position of contact with the support rack frame but with less intensity, as shown by the fuel plate (side plate made of Al 6262 T6, and fuel plate cladding made of Al 1060). There were no corrosion pits at the internal fuel plates.

These observations can also be applied to the spent fuel assemblies, where it was noticed that fuel plates have more corrosion pits than the side plates. Perhaps it is fair to

conclude, as observed in the fresh fuel, that also the internal fuel plates do not have (or at least have few) corrosion pits.

The inspection of the fuel assemblies with underwater camera was made afterwards together with the DOE-Savannah River Site group. The pattern of the pitting corrosion nodules, their sizes and location were investigated. Nevertheless these results and observations will not be discussed at this paper. At this paper the emphasis will be given to the sipping tests and the leaking pattern of the fuel assemblies.

Table 2 - Visual Inspection Resume

	LEU Fuel Assemblies	HEU Fuel Assemblies
Corrosion Pits along Fuel Plates	*42 43 48 49 53 55 58 61 62 64 66 69 70 78 79*	*103 106*
Few Corrosion Pits at Fuel Plates	*41 44 45 46 47 50 51 52 54 56 57 59 60 63 65 67 68 71 72*	*95 97 99 100 102 104 105 109*
No Remarkable Observation	*73*	*80 81 83 84 88 91 92 93 96 98 101 107 108 111 112*

SIPPING TEST

- **Procedure**

In order to perform the sipping test, the irradiated fuel assemblies were withdrawn from the spent fuel storage rack, have a rigid plastic pipe connected to its bottom nozzle, and were placed inside an aluminum sipping tube (120 mm of diameter, 3 m length, ~ 33 l of volume), as shown in Figure 2. This first part of the procedure was always done with the fuel assembly positioned, approximately, 2 meters of depth inside the pool water and monitored continuously by the radiological protection staff. Before the tests, the sipping tube was washed with demineralized water to reduce as much as possible any kind of residual contamination of radionuclides (mainly ^{24}Na).

The sipping tube with the fuel assembly inside was then lifted up and the top nozzle of the tube put above the surface of the water. It was then fixed to the pool bridge by a nylon rope. A total of 150 liters of demineralized water was then injected through the plastic pipe and flushed through the fuel assembly in order to wash it, as shown in Figure 2. After that, a background sample of the tube water was collected in a small plastic bottle (100 ml) and submitted to gamma -ray spectrometry analysis.

The fuel assembly was then left at rest inside the sipping tube during a time interval of at least four hours. Once finished the resting time, compressed air was injected through the plastic pipe and flushed through the fuel assembly, during two minutes, in order to homogenize the solution that might contain fission products released by the leaking fuel assembly. A sample of this solution was collected in a small plastic bottle (100 ml) and submitted to gamma-ray spectrometry analysis. All bottles used for sampling were identical. Once again, the work was monitored by the radiological protection staff.

Sipping tests following this procedure were performed on 60 irradiated fuel assemblies. Five aluminum sipping tubes were used simultaneously, and all work was done in two weeks.

A sample of pool water was also collected to serve as general gamma-ray background survey.

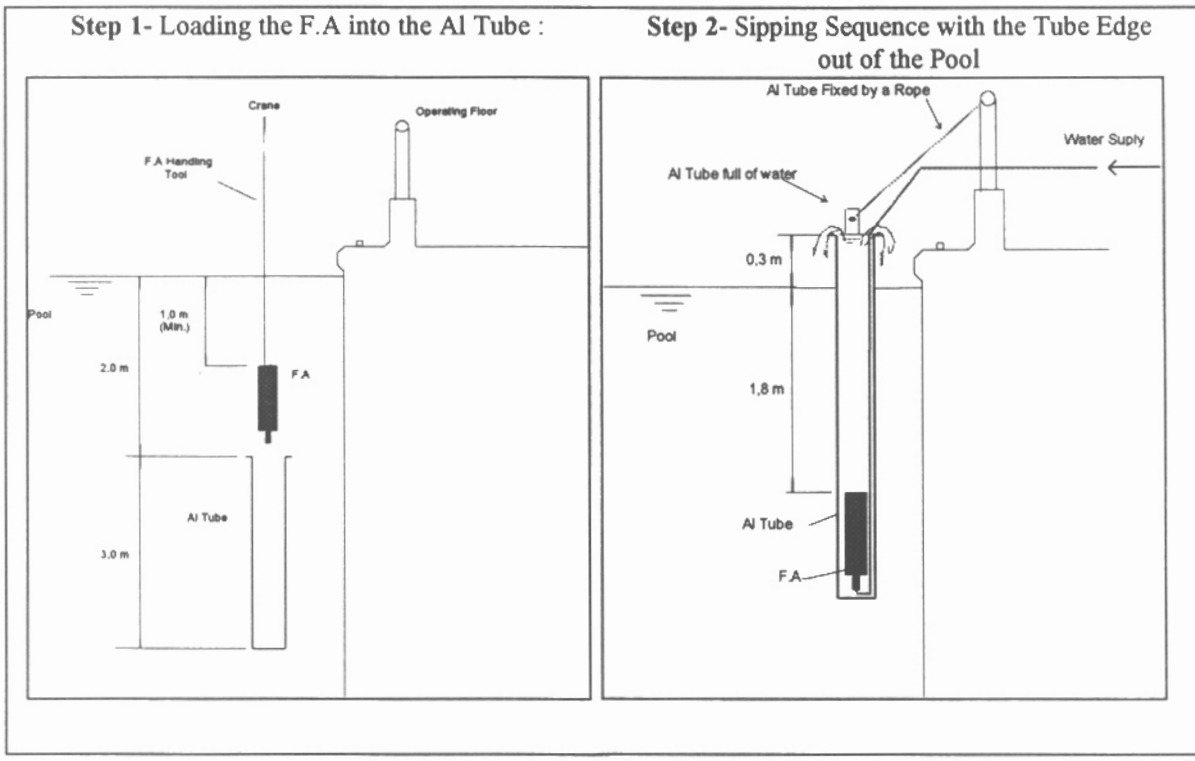

Figure 2 - Scheme of Sipping

- **Gamma-Ray Spectrometry**

The gamma-ray spectrometry analysis was carried out with a shielded ORTEC HPGe detector of volume 130 cm^3 with resolution of 2,0 keV and efficiency of 25% for the 1332 keV line of ^{60}Co. The gamma-ray energy range taken for the analysis was from 50 keV to 2800 keV. The data acquisition was performed with a ORTEC multichannel analyzer system coupled to a microcomputer through a control interface. Gamma-ray spectra were taken in runs of 4000 seconds of live time each.

The calibration of energy was obtained using a total of 40 peaks from 14 calibration sources. Gamma-ray energies were fitted as a second degree polynomial function of the corresponding peak channels in the calibration spectra.

The calibration of efficiency for the 661.6 keV gamma-ray under fixed geometry conditions was performed using a standard solution of 1.15×10^5 Bq of ^{137}Cs contained in a plastic bottle identical to the ones used to collect sipping water samples. Measurements made with this liquid calibration source gave an efficiency of $\varepsilon = (8.21 \pm 0.25).10^{-3}$.

For each measurement, the plastic bottle containing the collected water sample was placed inside the detector shield and near the detector window by means of a fixed wood support in order to maintain the same steady geometry for counting.

Two kinds of background measurements were carried out. The first one measured a sample of demineralized water in order to determine the natural gamma-ray background. The second one measured a sample of the pool water in order to determine the general gamma-ray background. No 661.6 keV photopeak was found at all in both background spectra (see Figures 3,4 and 5).

The 661,6 keV photopeak of each sipping spectrum, if any, was fitted by gaussian function plus a parabolic curve for the continuous background using the computer code IDEFIX[2]. By means of this procedure, the net number of counts (Area) under the 661.6 keV photopeak was determined and therefore the specific ^{137}Cs activity A of the solution as

$$A = \frac{Area}{\varepsilon . T . I_{\gamma} . Vol} \qquad (1)$$

where ε is the efficiency value mentioned before, T is the live time of measurement, I_{γ} is the gamma-ray absolute intensity and Vol is the volume of the sipping sample.

- **Results**

Table 3 shows the results of the gamma-ray spectra and activities of the ^{137}Cs for the fuel assemblies sipping samples. Table 4 shows the results of the system background and Figure 3 shows the gamma spectrum for the system background. Table 5 shows the results of the pool water background at the region of sipping test.

Some of the items shown in Table 3 have the following meaning:

i) samples

. background - analysis of the gamma spectrum from the water sample taken after water flushing through the fuel assembly and before resting time;

. first - analysis of gamma spectrum from the first water sample taken after resting time (called short sipping ~4 hours of resting time);

. second - analysis of gamma spectrum from the second water sample taken after resting time (called long sipping ~ > 8 hours of resting time).

ii) ^{137}Cs photopeak

. no - there is no defined photopeak at the channel related to the energy of ^{137}Cs gamma-ray (661.6 keV);

. counts - number of counts (for the established counting time - 4000 seconds) at the channel related to the energy of ^{137}Cs;

. photopeak area - calculated area for the photon peak related to the energy of ^{137}Cs;

. specific activity - activity of the sample calculated by means of equation (1).

TABLE 3 - FUEL ELEMENTS ASSEMBLIES SIPPING

Number	Sample	Resting Time (min.)	Sampling Date	Sampling Time	Measure-ment Date	Starting Time of Measure	Counting Time (sec.)	Photopeak of Cs - 137	Counts in Cs-137 channel	Photopeak Area	Specific Activity of Cs - 137 (Bq/l)
IEA-41	Background	-	02/07	09:00	02/07	09:17	4.000	No	63	-	-
	First	280	02/07	13:40	02/07	16:20	4.000	No	72	-	-
IEA-42	Background	-	02/07	09:40	02/07	22:14	4.000	No	66	-	-
	First	255	02/07	13:55	03/07	08:20	4.000	Yes	85	52 ± 34	18,61 ± 12,18
IEA-43	Background	-	02/07	10:17	03/07	09:40	4.000	No	63	-	-
	First	297	02/07	15:14	03/07	11:15	4.000	Yes	89	65 ± 39	23,26 ± 13,97
IEA-44	First	180	02/07	18:14	03/07	19:31	4.000	No	79	-	-
	Second	795	03/07	08:30	04/07	10:19	4.000	Yes	90	36 ± 27	12,88 ± 9,67
IEA-45	First	262	03/07	13:32	04/07	14:41	4.000	No	83	-	-
IEA-46	First	240	03/07	13:52	04/07	18:19	4.000	No	77	-	-
IEA-47	Background	-	01/07	12:16	01/07	19:05	7.200	No	135	-	-
	First	264	01/07	16:40	01/07	17:01	7.200	Yes	165	149 ± 52	29,62 ± 10,38
IEA-48	First	225	03/07	18:50	05/07	08:09	4.000	No	74	-	-
	Second	850	04/07	08:39	05/07	12:36	4.000	Yes	95	35 ± 22	12,52 ± 7,88
IEA-49	First	185	03/07	18:05	05/07	13:47	4.000	No	90	-	-
	Second	820	04/07	08:40	05/07	14:56	4.000	Yes	107	62 ± 47	22,18 ± 16,83
IEA-50	First	247	04/07	14:02	05/07	16:19	4.000	Yes	84	25 ± 17	8,95 ± 6,09
IEA-51	First	235	04/07	14:20	05/07	17:27	4.000	No	75	-	-
IEA-52	First	790	05/07	08:35	08/07	15:27	4.000	No	88	-	-
IEA-53	Backgr.-1	-	04/07	17:55	09/07	12:22	4.000	No	75	-	-
	First	765	05/07	08:40	08/07	16:43	4.000	Yes	178	261 ± 55	93,39 ± 19,88
	First	765	05/07	08:40	12/07	20:02	86.400	Yes	3.672	5984±171	99,13 ± 4,14
	Backgr. -2	-	10/07	16:13	16/07	08:45	4.000	No	69	-	-
	Second	1041	11/07	09:34	16/07	10:38	4.000	Yes	194	422 ± 32	151,00 ± 12,34
	Second	1041	11/07	09:34	23/07	15:30	4.000	Yes	261	382 ± 30	136,69 ± 10,74
IEA-54	First	740	05/07	08:45	08/07	18:05	4.000	Yes	112	115 ± 67	41,15 ± 24,01
IEA-55	First	345	05/07	15:33	09/07	07:02	4.000	No	87	-	-

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Number	Sample	Resting Time (min.)	Sampling Date	Sampling Time	Measure-ment Date	Starting Time of Measure	Counting Time (sec.)	Photopeak of Cs - 137	Counts in Cs-137 channel	Photopeak Area	Specific Activity of Cs - 137 (Bq/l)
IEA-56	First	320	05/07	15:39	09/07	08:18	4.000	No	97	-	-
IEA-57	First	310	05/07	15:43	09/07	09:27	4.000	Yes	108	83 ± 25	29,70 ± 8,99
IEA-58	Background	-	17/07	11:45	18/07	10:05	4.000	No	73	-	-
	First	304	17/07	16:49	18/07	12:42	4.000	Yes	94	30 ± 27	10,73 ± 9,67
	Second	1080	18/07	10:52	18/07	14:42	4.000	Yes	155	193 ± 32	69,06 ± 11,64
IEA-59	Background	-	02/07	09:10	02/07	15:09	4.000	No	71	-	-
	First	275	02/07	13:45	02/07	17:58	4.000	No	80	-	-
IEA-60	Background	-	02/07	10:00	03/07	16:12	4.000	No	88	-	-
	First	310	02/07	15:10	03/07	17:23	4.000	No	97	-	-
IEA-61	First	200	02/07	18:10	04/07	11:44	4.000	Yes	93	46 ± 37	16,46 ± 13,25
	Second	815	03/07	08:25	04/07	13:11	4.000	Yes	132	96 ± 31	34,35 ± 11,14
IEA-62	Background	-	02/07	15:30	02/07	19:40	4.000	No	89	-	-
	Background	-	02/07	15:30	17/07	17:33	4.000	No	67	-	-
	First	167	02/07	18:17	03/07	14:52	4.000	Yes	90	56 ± 27	20,04 ± 9,68
	First	167	02/07	18:17	17/07	18:45	4.000	Yes	92	79 ± 46	28,27 ± 16,48
	Second	780	03/07	08:31	03/07	13:08	4.000	Yes	104	78 ± 26	27,91 ± 9,34
	Second	780	03/07	08:31	17/07	19:56	4.000	Yes	121	113 ± 32	40,43 ± 11,52
IEA-63	First	240	03/07	13:37	04/07	19:31	4.000	No	96	-	-
IEA-64	First	253	03/07	14:20	05/07	18:38	4.000	Yes	102	68 ± 35	24,33 ± 12,55
IEA-65	First	243	03/07	14:25	05/07	19:49	4.000	Yes	92	42 ± 24	15,03 ± 8,60
IEA-66	Background	-	03/07	14:42	17/07	10:07	4.000	No	65	-	-
	First	200	03/07	18:02	08/07	07:58	4.000	No	91	-	-
	First	200	03/07	18:02	17/07	14:11	4.000	No	81	-	-
	Second	834	04/07	08:36	08/07	09:24	4.000	Yes	128	111 ± 82	39,72 ± 29,37
	Second	834	04/07	08:36	17/07	15:27	4.000	Yes	150	125 ± 25	44,73 ± 9,05
IEA-67	First	266	04/07	14:00	08/07	10:36	4.000	No	83	-	-
IEA-68	First	237	04/07	14:07	08/07	12:06	4.000	Yes	90	62 ± 45	22,18 ± 16,12
IEA-69	First	225	04/07	14:25	08/07	14:02	4.000	Yes	95	55 ± 32	19,68 ± 11,47

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Number	Sample	Resting Time (min.)	Sampling Date	Sampling Time	Measure-ment Date	Starting Time of Measure	Counting Time (sec.)	Photopeak of Cs - 137	Counts in Cs-137 channel	Photopeak Area	Specific Activity of Cs - 137 (Bq/l)
IEA-70	Background	-	04/07	17:40	12/07	17:18	4.000	No	80	-	-
	First	780	05/07	08:37	09/07	10:40	4.000	Yes	123	123 ± 50	44,01 ± 17,94
IEA-71	First	752	05/07	08:42	09/07	13:30	4.000	Yes	121	147 ± 87	52,60 ± 31,17
IEA-72	First	390	05/07	15:30	09/07	14:54	4.000	Yes	88	95 ± 30	33,99 ± 10,78
IEA-73	First	332	05/07	15:35	09/07	16:23	4.000	Yes	108	75 ± 26	26,84 ± 9,34
IEA-78	Background	-	17/07	12:00	18/07	15:53	4.000	No	73	-	-
	First	300	17/07	17:00	18/07	17:38	4.000	Yes	104	56 ± 24	20,04 ± 8,61
	Second	1072	18/07	10:56	18/07	19:44	4.000	Yes	213	326 ± 38	116,65 ± 14,05
IEA-79	Background	-	17/07	12:16	18/07	21:13	4.000	No	62	-	-
	First	295	17/07	17:11	18/07	22:35	4.000	No	72	-	-
	Second	1065	18/07	11:00	19/07	00:01	4.000	Yes	86	36 ± 25	12,88 ± 8,95
80	First	293	08/07	14:13	09/07	17:31	4.000	Yes	85	30 ± 22	10,73 ± 7,88
81	First	1048	09/07	08:45	09/07	18:45	4.000	No	72	-	-
83	First	274	08/07	14:16	10/07	08:39	4.000	No	78	-	-
84	First	307	09/07	15:17	10/07	09:58	4.000	No	96	-	-
88	First	260	08/07	14:20	10/07	11:22	4.000	No	71	-	-
91	First	269	09/07	15:25	11/07	19:03	4.000	Yes	94	25 ± 20	8,95 ± 7,16
92	First	221	08/07	14:28	10/07	14:39	4.000	Yes	75	23 ± 16	8,23 ± 5,73
93	First	1029	09/07	08:53	10/07	17:49	4.000	No	81	-	-
95	First	254	10/07	15:24	15/07	16:00	4.000	No	74	-	-
96	Background	-	08/07	15:53	17/07	08:12	4.000	No	65	-	-
	First	1023	09/07	08:56	10/07	19:25	4.000	No	83	-	-
97	First	1013	10/07	09:00	12/07	09:11	4.000	Yes	96	58 ± 38	20,75 ± 13,61
98	First	328	10/07	15:13	12/07	14:07	4.000	No	85	-	-
99	Background	-	09/07	9:53	17/07	12:07	4.000	No	66	-	-
	First	320	09/07	15:13	11/07	15:34	4.000	Yes	110	63 ± 36	22,54 ± 12,90
100	First	997	10/07	09:05	12/07	10:26	4.000	No	78	-	-
101	First	1057	09/07	08:40	10/07	15:54	4.000	Yes	104	65 ± 31	23,26 ± 11,12
102	First	264	10/07	15:16	15/07	11:14	4.000	No	73	-	-

TABLE 3

Number	Sample	Resting Time (Min)	Sampling Date	Sampling Time	Measure-ment Date	Starting Time of Measure	Counting Time (sec.)	Photopeak of Cs - 137	Counts in Cs-137 channel	Photopeak Area	Specific Activity of Cs - 137 (Bq/l)
103	First	993	10/07	09:15	12/07	11:40	4.000	No	91	-	-
104	First	1017	10/07	08:55	11/07	23:45	4.000	Yes	99	28 ± 24	10,02 ± 8,59
105	First	252	10/07	15:27	15/07	17:08	4.000	No	75	-	-
106	First	983	10/07	09:20	12/07	12:59	4.000	Yes	92	47 ± 29	16,82 ± 10,39
107	First	282	09/07	15:21	11/07	16:46	4.000	No	83	-	-
108	First	244	08/07	14:24	10/07	13:10	4.000	No	63	-	-
109	First	260	10/07	15:20	15/07	14:23	4.000	Yes	85	55 ± 25	19,68 ± 8,96
111	First	1007	09/07	09:02	11/07	09:15	4.000	No	82	-	-
112	First	335	09/07	15:10	11/07	10:45	4.000	No	70	-	-

TABLE 4 - BACKGROUND MEASUREMENTS

Number	Measurement Date	Starting Time of Measure	Counting Time (sec.)	Photopeak of Cs - 137	Counts in Cs-137 channel	Specific Activity of Cs - 137 (Bq/l)
1	02/07	09:17	7200	No	135	-
2	03/07	19:31	4000	No	63	-

TABLE 5 - IEAR1 REACTOR POOL WATER MEASUREMENTS

Sampling Date	Sampling Time	Measurement Date	Starting Time of Measure	Counting Time (sec.)	Photopeak of Cs - 137	Counts in Cs-137 channel	Specific Activity of Cs - 137 (Bq/l)
10/07	10:20	11/07	19:03	4.000	No	199	-
		12/07	18:29		No	120	-
		15/07	08:45		No	62	-
		16/07	11:49		No	66	-

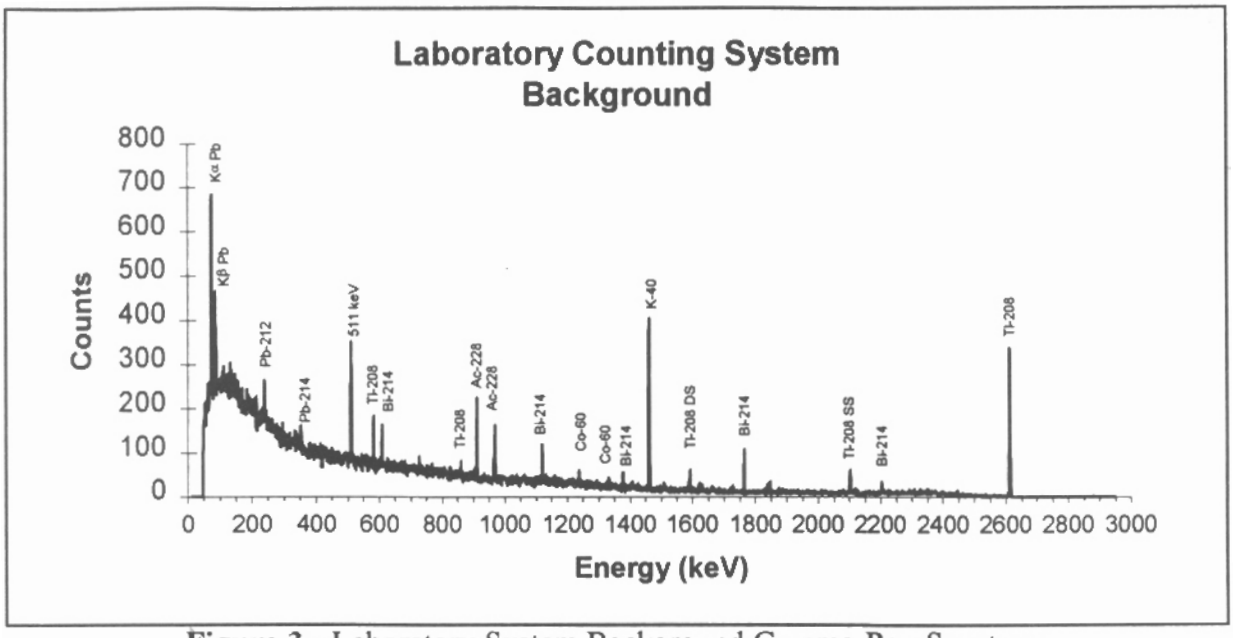

Figure 3 - Laboratory System Background Gamma-Ray Spectrum

The following observations can be taken from the Table 3:

- some fuel assemblies show the ^{137}Cs photopeak;

- for those fuel assemblies that two samples were taken, it is noticed that the activity increases with sipping resting time, indicating that the fuel assembly is really leaking. It is possible to determine a leaking rate of ^{137}Cs from the fuel assembly. For example, for the fuel assembly number IEA-53 this rate is ~0.2 Bq/l.min or ~14 Bq/min;

- for some fuel assemblies the activity is very low and the associated error in the analysis is big. For longer counting time the activity is still the same but the associated error decreases;

- there is influence of the analysis date on the measured activity. This is due to the activation of the pool water that gives a high background level in the gamma spectrum. The sipping tests were done with the reactor in operation and the level of ^{24}Na (half-life of 15 hours) in the water was high enough for a high Compton background. Table 5 shows the pool water background and it is observed that the number of counts at the channel related to the energy of ^{137}Cs decreases with the time of the analysis after sampling. Figure 4 and 5 show the gamma-ray spectra for the pool water, where one can see the influence of ^{24}Na in the Compton background.

- the water volume of the sipping tube is ~33 liters. The volume of the fuel assembly is ~ 2 liters, and the volume lost with the compressed air injection is of ~3 liters. So the activity for the fuel assembly sipping is obtained multiplying the specific activity by 28 liters.

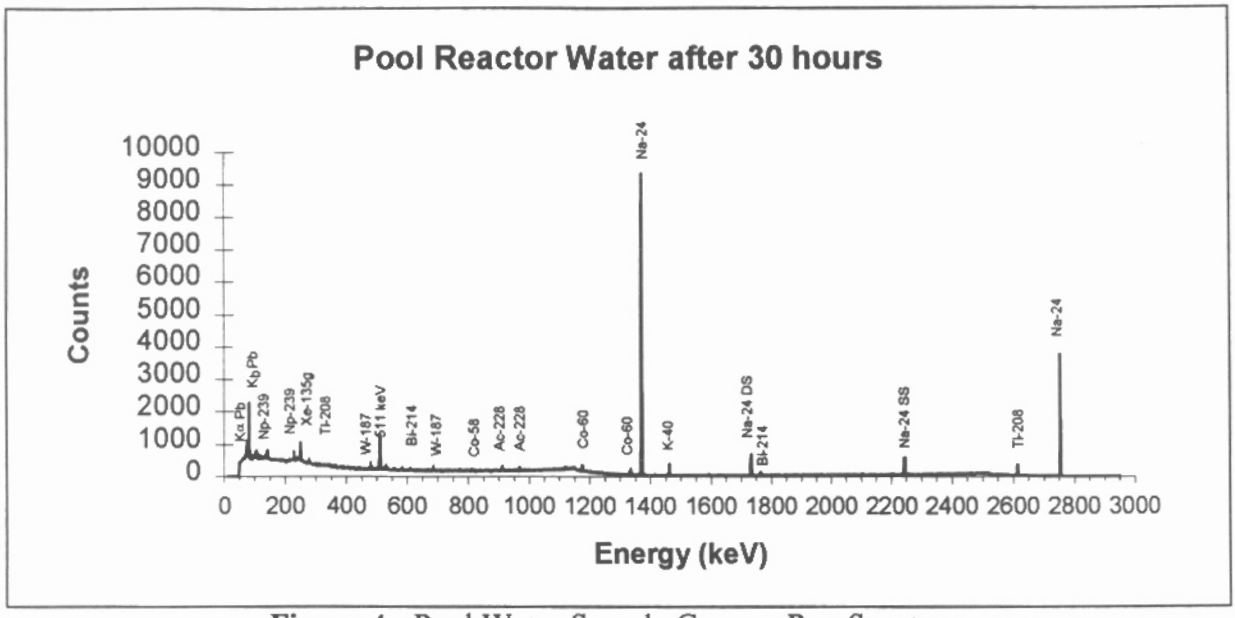

Figure 4 - Pool Water Sample Gamma-Ray Spectrum

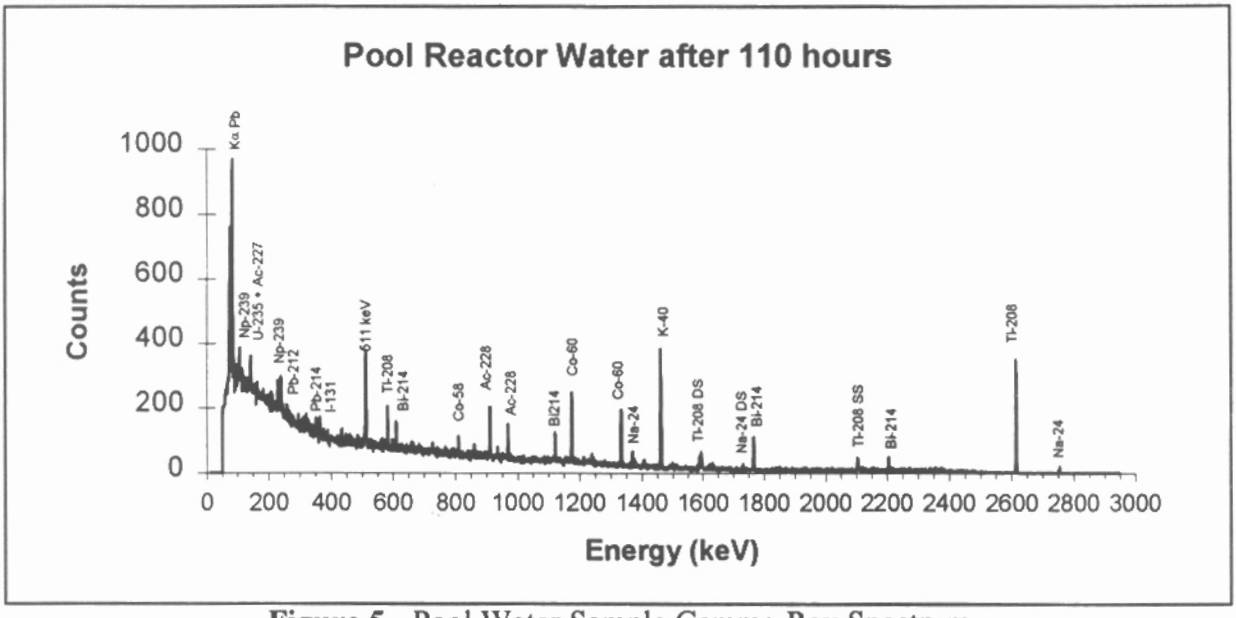

Figure 5 - Pool Water Sample Gamma-Ray Spectrum

Figure 6 shows the counts of the ^{137}Cs energy channel for every fuel assembly analyzed. It can be seen that the background analysis for that fuel assembly with no indication of ^{137}Cs shows a base level of counts between 60 and 85 (for 4000 sec). For those fuel assemblies that indicate the ^{137}Cs photopeak the counts are higher than 90.

Figure 7 shows the specific activity calculated for each fuel assembly sipping test. It can be noticed that some fuel assemblies show a leaking pattern, and that most of fuel assemblies have specific activities lower than 30 Bq/l (for sipping resting time of 4 hours).

Figures 8,9,10,and 11 show the gamma spectra for one nonleaking fuel assembly and one leaking fuel assembly, showing also the difference in the background spectra.

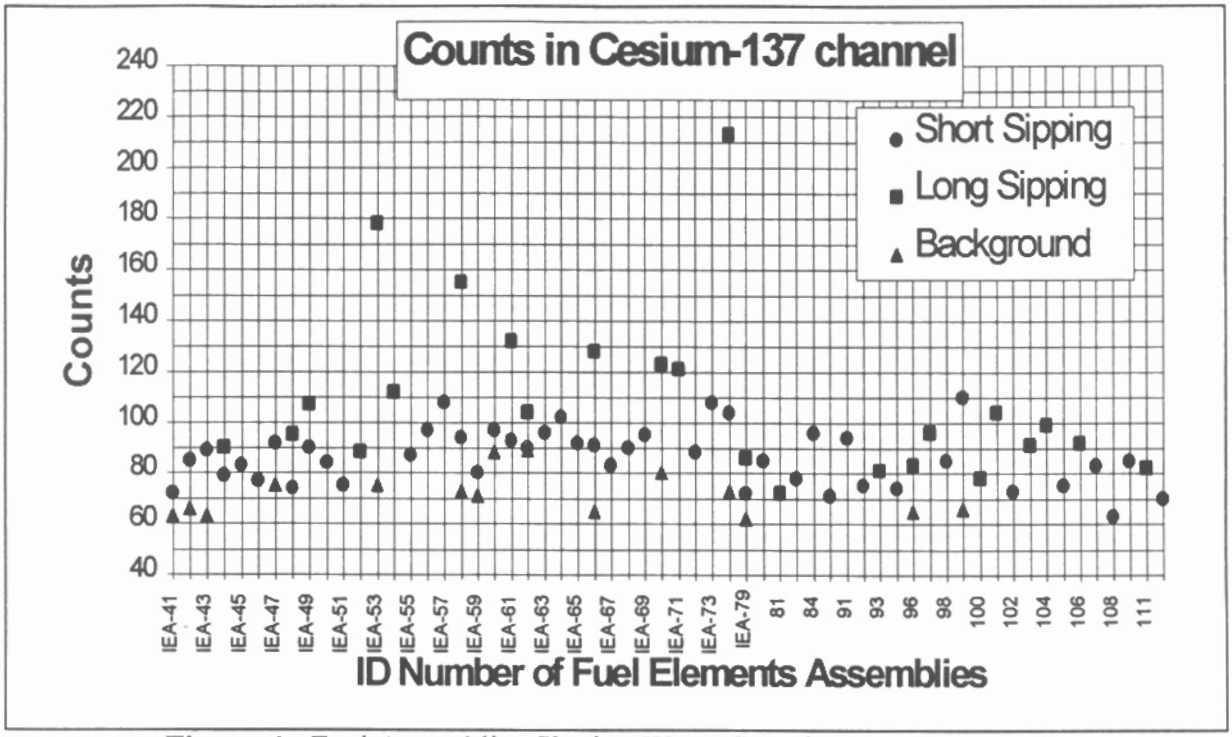

Figure 6 - Fuel Assemblies Sipping Water Samples Counting Results

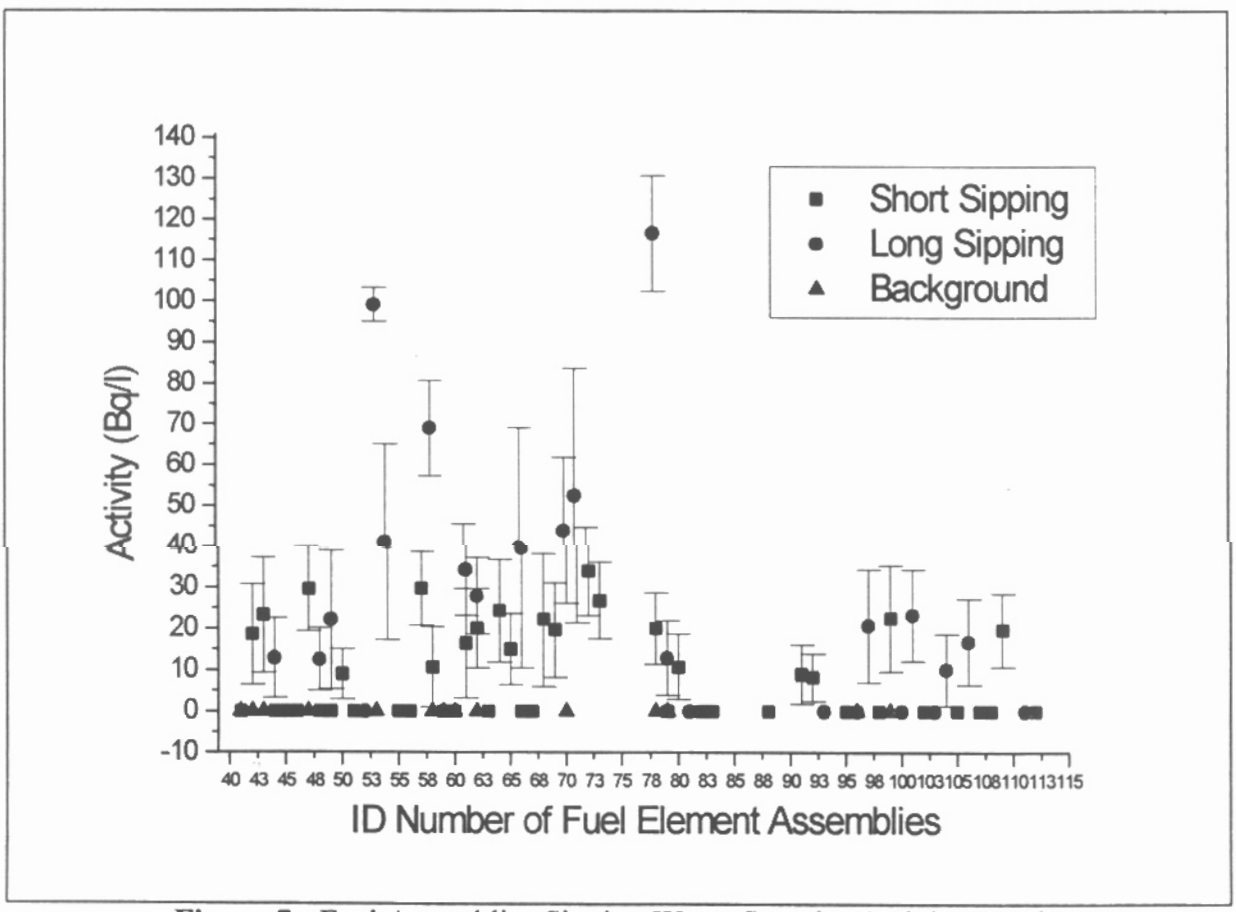

Figure 7 - Fuel Assemblies Sipping Water Samples Activity Results

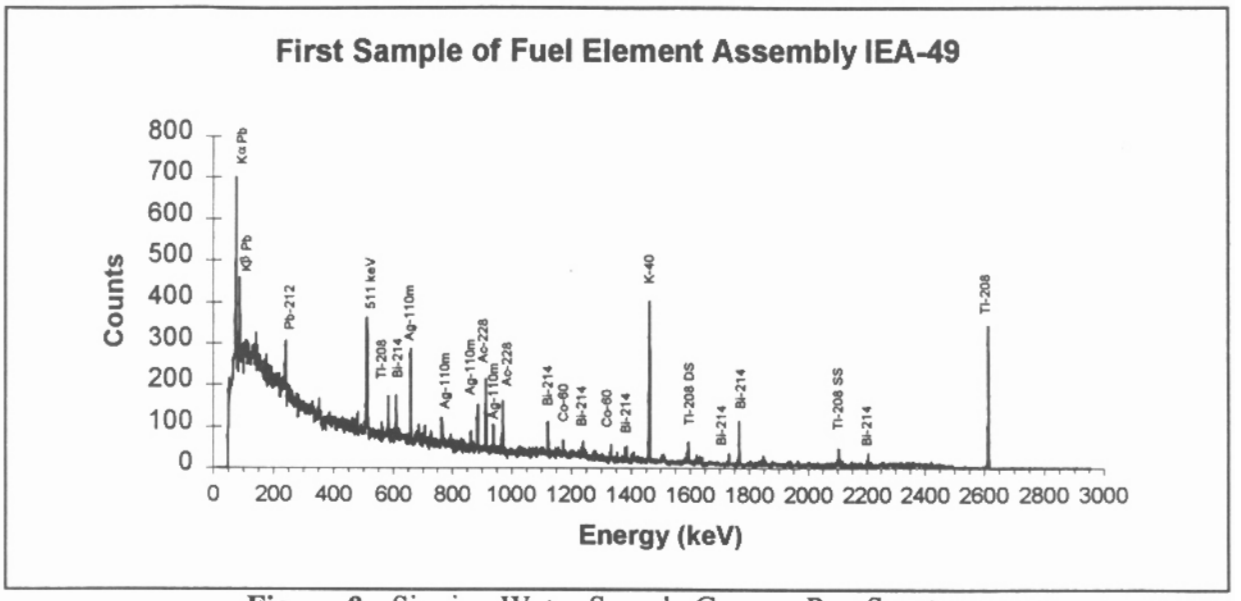

Figure 8 - Sipping Water Sample Gamma-Ray Spectrum

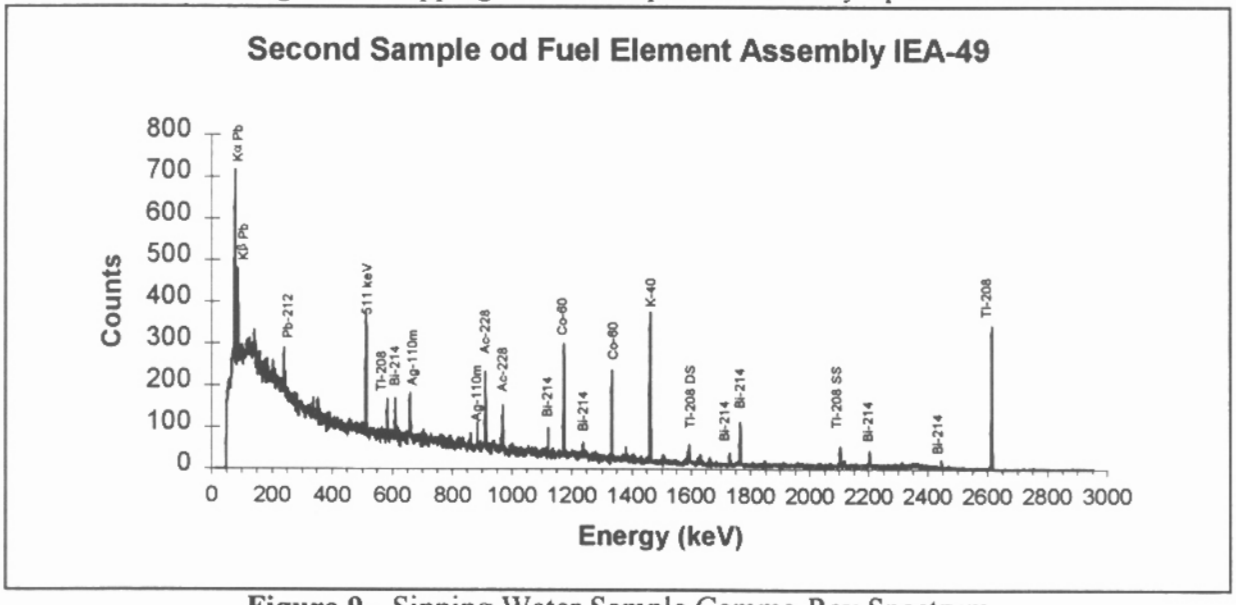

Figure 9 - Sipping Water Sample Gamma-Ray Spectrum

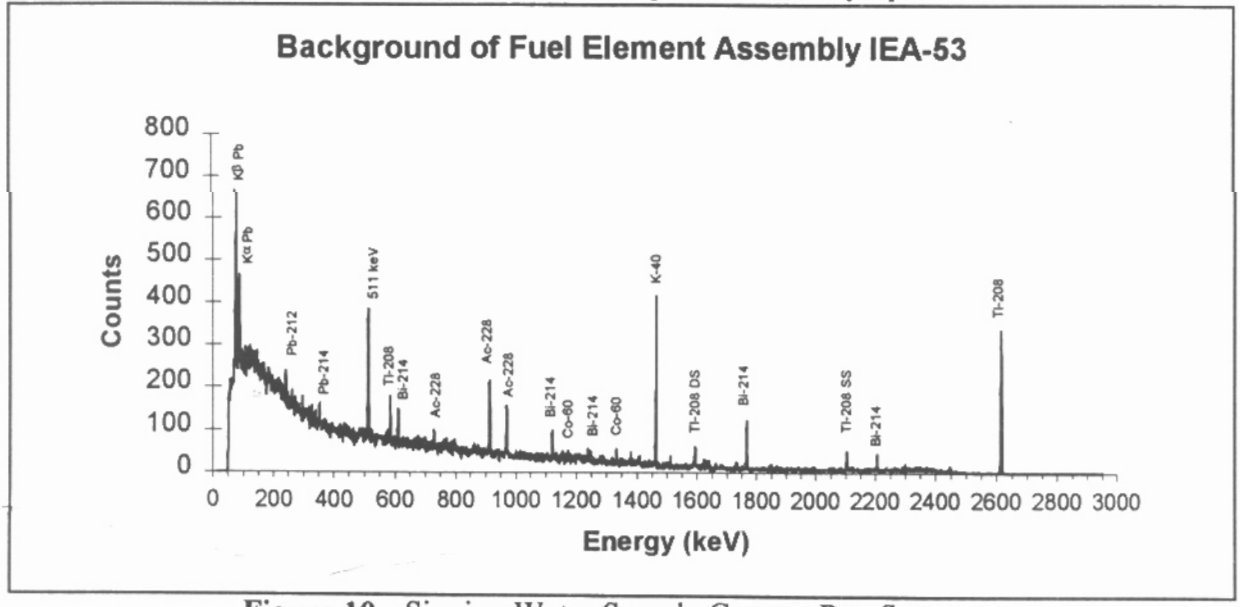

Figure 10 - Sipping Water Sample Gamma-Ray Spectrum

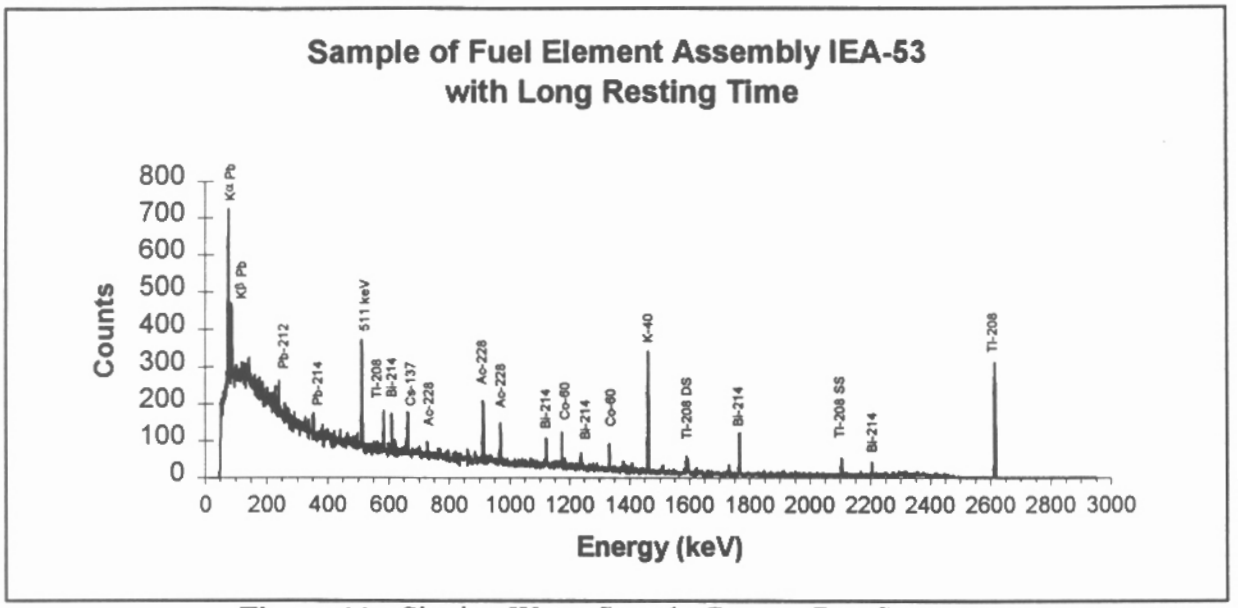

Figure 11 - Sipping Water Sample Gamma-Ray Spectrum

The procedure used for sampling and performing gamma-ray spectra analysis used a 100 ml water sample and 4000 seconds of counting time. This was done due to the high number of fuel assemblies to be analyzed and the short time for doing it. In order to evaluate the precision of the values obtained for the specific activities, it was taken, for some fuel assemblies, water samples of 850 ml. These samples were analyzed in a gamma spectrometry system with higher efficiency than the one used for the sipping tests. Also the counting time was taken to 50000 seconds, which gives minor counting errors. Figure 12 shows the results of this comparison. One can observe in this figure that the system used in the sipping test is always overestimating the results, and giving good results for the specific activity higher than 20 Bq/l. For lower activities the associated deviation is higher. Being this level of activity small, it shows that the sipping test system used is very suitable for determining leaking fuel assemblies.

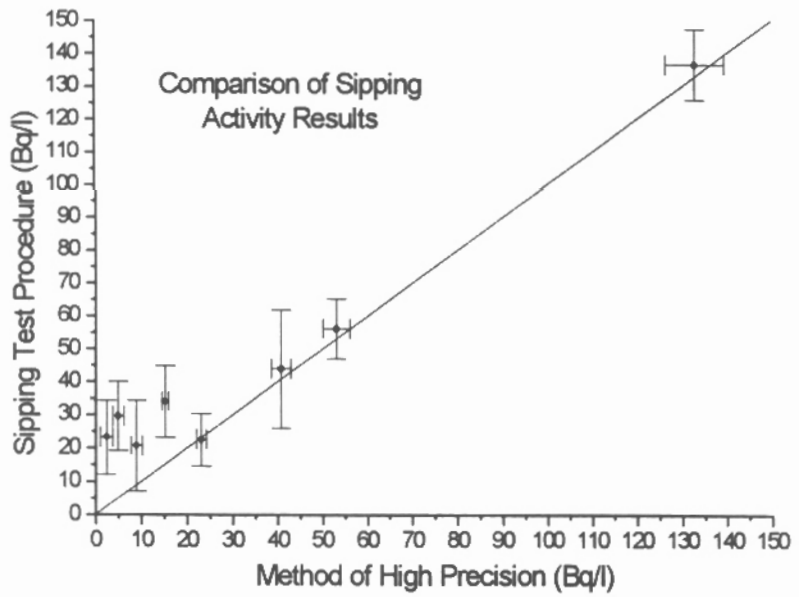

Figure 12 - Comparison of the Activity Precision

RELATION BETWEEN VISUAL INSPECTION AND SIPPING

It is interesting to correlate the visual inspection pattern with the sipping results. Table 6 shows this relation. One can observe that the older LEU fuel assemblies are in worse condition than the HEU fuel assemblies, and only few fuel assemblies have higher indication of ^{137}Cs leaking.

Table 6 - Correlation between Visual Inspection and Sipping Activity for IEA-R1 Fuel Assemblies

	Pits along external fuel plate	**Few pits along external fuel plate**	**No visible remarks**
No Indication of Cs-137	*55*	*41, 45, 46, 51, 52, 56, 59, 60, 63, 67*	
	103	*95, 100, 102, 105*	*81, 83, 84, 88, 93, 96, 98, 107, ,108, 111, 112*
Low Indication of Cs-137 (< 30Bq/l)	*42, 43, 48, 49, 62, 64, 66, 69, 79*	*44, 47, 50, 57, 65, 68*	*73*
	106	*97, 99, 104, 109*	*80, 91, 92, 101*
Medium Indication of Cs-137 (> 30 Bq/l;< 60 Bq/l)	*61, 70*	*54, 71, 72*	
High Indication of Cs-137 (> 60 Bq/l)	*53, 58, 78*		

CHEMICAL ANALYSIS

The sipping water sample, for the fuel assembly with highest leaking activity, was also analyzed by X-ray fluorescence spectrometry to see whether there was Cs and U. Table 7 shows the results. One can see that the only elements observed are those of corrosion ions from aluminum and stainless steel that are inside the pool. Cs and U were not observed. This is easy to understand if one observe the activity of ^{137}Cs in the sipping water. For the value of 100 Bq/l represents $\sim 3 \times 10^{-11}$ g Cs/l. This value is too small to be detect by chemical

analysis. U was detected neither by gamma ray spectrometry analysis nor by X-ray spectrometry.

Table 7 - Chemical Analysis Results (X-Ray Spectrometry)

Element	Fuel Assembly - EC IEA-49	Fuel Assembly - EC IEA-53
Al	< 100 mg/ml	< 100 mg/ml
Si	< 100 mg/ml	< 100 mg/ml
P	< 100 mg/ml	< 100 mg/ml
S	< 100 mg/ml	< 100 mg/ml
Ca	< 100 mg/ml	< 100 mg/ml
Cr	< 100 mg/ml	< 100 mg/ml
Fe	< 100 mg/ml	< 100 mg/ml
Ni	< 100 mg/ml	< 100 mg/ml
Zn	< 100 mg/ml	< 100 mg/ml
Co	ND	ND
Cs	ND	ND
U	ND	ND
In	ND	ND
Ag	ND	ND
Cu	< 100 mg/ml	< 100 mg/ml

ND - Not Detected

DETERMINATION OF ^{137}Cs LEAKING RATE

The DOE-Savannah River Site group came to IPEN to the assessment of the fuel assemblies for shipment. This group agreed with the methodology used by IPEN in performing sipping tests and also with the results obtained. Nevertheless, some additional work concerning fuel assessment was done. One task was to remove some of the external fuel plate pitting corrosion nodules of some fuel assemblies and to repeat the sipping test on these fuel assemblies. The second task was to determine the ^{137}Cs leaking rate, after cleaning some pitting corrosion nodules, from fuel assembly IEA-53 the one that showed the highest sipping activity among all fuel assemblies. Both tasks were done with IPEN methodology and equipment.

Table 8 shows the results for the specific activity in the sipping tests performed with some fuel assemblies after cleaning the outside plate surface and taking out some pitting corrosion nodules. One can observe at Table 8 that there is no evident difference between the results before and after cleaning. The differences are more evident upon the sipping resting time.

One can also observe at Table 8 the results for fuel assembly IEA-53. Figure 13 shows the activity along sipping resting time. The results show a straight line with a constant leaking rate of ~0.2 Bq/l.min or ~14 Bq/min. Figure 14 compares the rate before and after cleaning and one can see that there is almost no difference.

Being the fuel assembly IEA-53 the one which had the highest leaking rate, this value of 14 Bq/min can be compared to the criteria established by DOE-SRS for canning leaking fuel assemblies.

The DOE-SRS interim criteria presented to IPEN was 13.57 μCi/hr per cask

shipment which is equivalent to 35.9 pCi/ml.hr for one fuel assembly (assuming 3.6 gallons of water per each fuel assembly).

The value obtained at IPEN fuel assembly sipping, correcting to the volume of SRS criteria, is 0.64 pCi/ml.hr (0.4 Bq/l.min.) which is far bellow the criteria limit.

Nevertheless DOE interpretation of Foreign Research Reactor Environmental Impact Statement (FRR.EIS) says that any fuel that contain a thru-clad pit may require canning for shipment and storage. So, although the sipping criteria is fulfilled by IPEN fuel assemblies, DOE imposes some restraint upon corrosion pattern. It is the authors concern, that this subject has to be discussed in more details in order to have a well established criteria, since any detected ^{137}Cs sipping activity would be a consequence of a thru-clad pit.

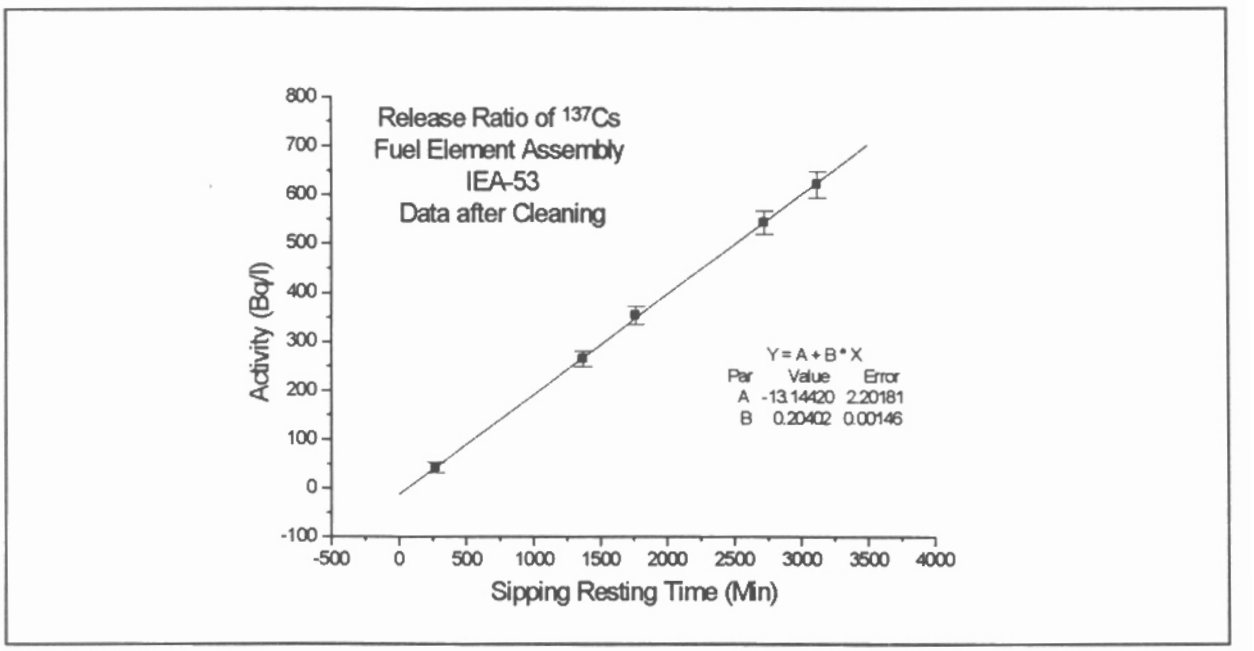

Figure 13 - ^{137}Cs Leaking Rate Determination

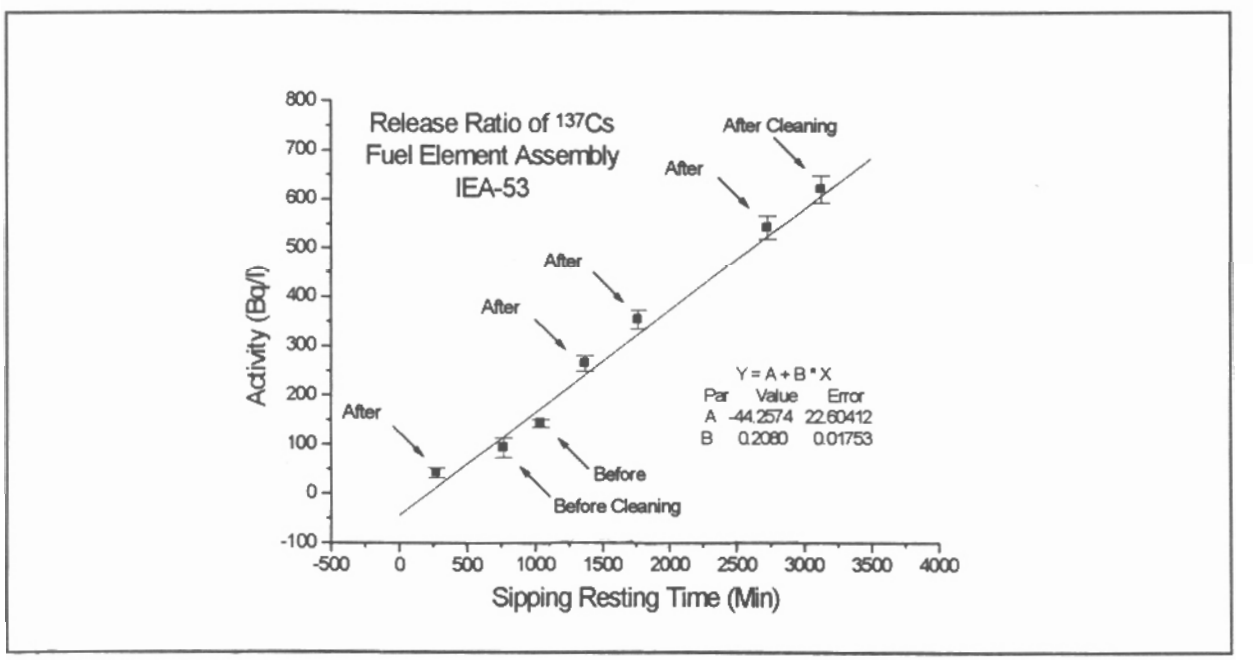

Figure 14 - ^{137}Cs Leaking Rate Determination

TABLE 8 - FUEL ELEMENTS ASSEMBLIES SIPPING

Number	Sample	BEFORE CLEANING					AFTER CLEANING				
		Rest Time (Min)	Photopeak of Cs - 137	Counts in Cs-137 channel	Photopeak Area	Specific Activity of Cs - 137 (Bq/l)	Rest Time (Min)	Photopeeak of Cs - 137	Counts in Cs-137 channel	Photopeak Area	Specific Activity of Cs - 137 (Bq/l)
IEA-41	First	280	No	72	-	-	240	No	82	-	-
IEA-45	First	262	No	83	-	-	240	No	68	-	-
IEA-53	Background	-	No	75	-	-	-	No	68	-	-
	First	765	Yes	178	261 ± 55	93,39 ± 19,88	275	Yes	147	119 ± 29	42,58 ± 10,46
	Second	1041	Yes	228	401 ± 22	142,86 ± 8,10	1370	Yes	418	740 ± 39	264,79 ± 16,12
	Third	-	-	-	-	-	1770	Yes	525	988 ± 43	353,53 ± 18,78
	Forth	-	-	-	-	-	2730	Yes	778	1517 ± 47	542,82 ± 23,58
	Fifth	-	-	-	-	-	3127	Yes	773	1737 ± 56	621,54 ± 27,56
IEA-73	First	332	Yes	108	75 ± 26	26,84 ± 9,34	240	No	77	-	-
92	First	221	Yes	75	23 ± 16	8,23 ± 5,73	240	No	77	-	-
93	First	1029	No	81	-	-	315	No	75	-	-
97	First	1013	Yes	96	58 ± 38	20,75 ± 13,61	240	No	70	-	-
101	First	1057	Yes	104	65 ± 31	23,26 ± 11,12	240	No	75	-	-
103	First	993	No	91	-	-	240	Yes	89	27 ± 19	9,66 ± 6,80
104	First	1017	Yes	99	28 ± 24	10,02 ± 8,59	240	No	83	-	-
106	First	983	Yes	92	47 ± 29	16,82 ± 10,39	255	No	72	-	-

GAMMA-RAY SPECTROMETRY OF A PITTING CORROSION NODULE

A sample of a pitting corrosion nodule of a fuel plate was taken to perform gamma-ray spectrometry analysis. Figure 15 shows the gamma-ray spectrum obtained. One can find a large activity of ^{137}Cs and also activities of ^{235}U, ^{155}Eu, and ^{154}Eu. This indicates that the pitting corrosion nodule penetrates through the cladding up to the plate meat where there is uranium and fission products. Also indicates that besides ^{137}Cs, U and Eu isotopes are migrating from the meat to the nodule. U and Eu are not migrating (or migrating in a very small rate) to the water, though this elements were not found in the gamma-ray spectrometry of the sipping water. This mechanism of U and F.P. migration from the plate meat through the corrosion pit to water (migration/solubility/diffusion/chemical compounds/chemical reaction/etc.) has to be more deeply studied.

The activity of ^{137}Cs in the pitting corrosion nodule sample was determinate to be 2400 Bq or 5600 Bq/g. It is interesting to compare this activity with the meat activity and the sipping water activity.

The ^{137}Cs plate activity, obtained by the code ORIGEN2[3] , taking into consideration the plate burnup and decay time to the date of the sipping test, is ~ 2Ci or $7.4x10^{10}$ Bq. Considering the weight and volume of the U-Al alloy of the plate meat, the specific activity is given by $3.4x10^{9}$ Bq/cm^3 or $7.5x10^{8}$ Bq/g(U-Al).

So, the three values of ^{137}Cs activities are:

- meat - $7.5x10^{8}$ Bq/g ;or ~ $7.4x10^{10}$ Bq/plate ; or ~$1.3x10^{12}$ Bq/F.A

- corrosion pit nodule - $5.6x10^{3}$ Bq/g ; or ~ $2.4x10^{3}$ Bq/nodule

- sipping water sample - 14 Bq/min ; or 0.2 Bq/l.min ; or $2x10^{-5}$ Bq/g.min (max. per fuel assembly)

There is a migration of ^{137}Cs from the meat to the water, with a retention at the pitting corrosion nodule, with a leaking rate of orders of magnitude below the meat activity.

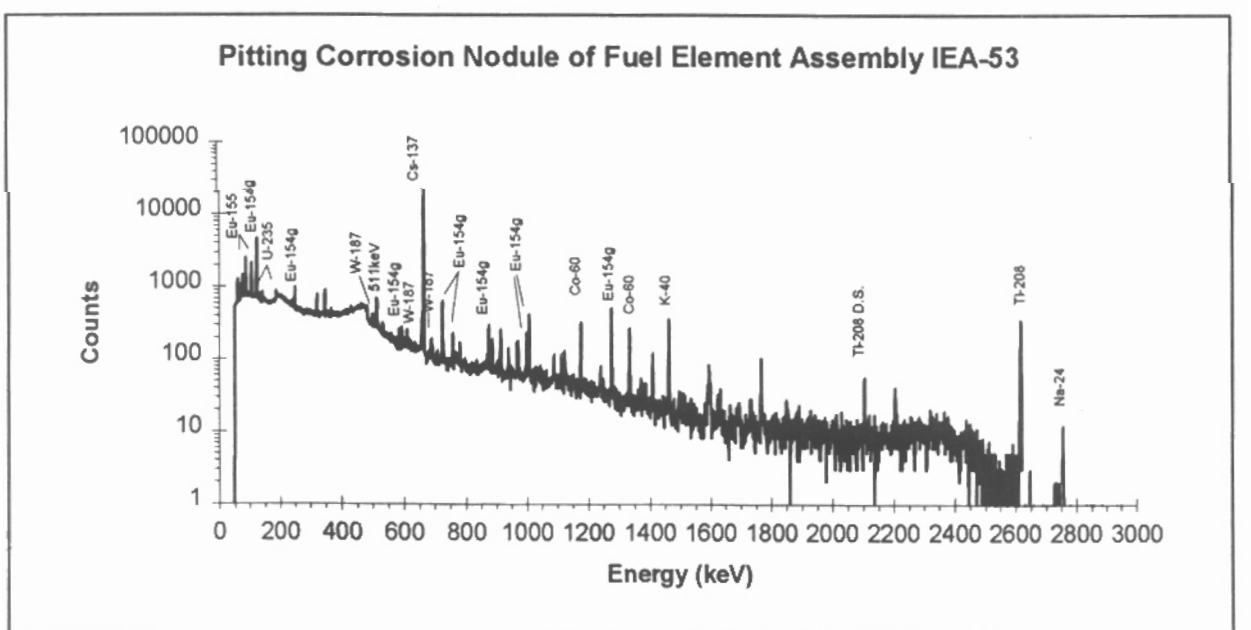

Figure 15 - Pitting Corrosion Nodule-Gamma Ray Spectrum

CONCLUSION

- The visual inspection of the spent fuel assemblies storage at the spent fuel pool showed that pitting corrosion is present in the external fuel plates of many fuel assemblies. Those which are inside water for almost 40 years show the worse pattern. The pitting corrosion observed is due to the galvanic pair existing between aluminum fuel plate cladding and the stainless steel spent fuel rack.

- The sipping test methodology and equipment used by IPEN showed to be efficient in determining fission products leaking fuel assemblies.

- It was determined a ^{137}Cs leaking rate of 14 Bq/min for the worse leaking fuel assembly. This value is far bellow the DOE-SRS criteria presented to IPEN for canning leaking MTR fuel assemblies.

- Pitting corrosion nodule gamma-ray spectrometry shows the occurrence of Cs, U and Eu isotopes. The ^{137}Cs activity is much higher than the sipping water activity and U and Eu isotopes were not detected in water. This confirms that it is a thru-clad pit.

The following items are suggested, by the authors, for future research.

- Increase the understanding of galvanic pitting corrosion between Al alloys and stainless steel in the basin. Also the difference of the pit pattern for different types of MTR fuel plates (U-Al alloy and dispersion fuels- U_3O_8-Al, U_3Si_2-Al, UAl_x-Al).

- Increase the understanding of the phenomena involved in the U and fission products migration through a pitting corrosion nodule to the basin. Also the difference of these phenomena for different types of MTR fuel plates.

- Discuss the visual thru-clad pit criteria for canning leaking fuel assemblies for shipment and storage.

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It is also important to point out the good relationship between IPEN/CNEN-SP and DOE-SRS staffs during the IEA-R1 fuel assemblies assessment for shipment. The authors would like to express their thanks to B.K.Chambers, J.P.Howell, R.L.Sindelar, S.D.Burke, A.S.Busby, and H.B.Peacock for the participation and discussions to achieve the goals of this work.

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[3] RSIC Computer Code Collection - "ORIGEN2 - Isotope Generation and Depletion Code - Matrix Exponential Method" - RSIC Code Package CCC-371 (April 1987).