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IRRADIATED MTR FUEL ASSEMBLIES SIPPING TEST

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ABSTRACT

The 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. Many Irradiated fuels have been stored at the reactor pool along the various years of the facility operation. 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. 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 analyzed. 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 inside the pool or for shipment abroad.

I. INTRODUCTION

The IEA-R1 is a pool type, light water moderated and graphite reflected research reactor. It was designed and built by Babcock & 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 one achieved in South America. Although designed to operate at 5 MW, the IEA-R1 reactor has been operating at 2 MW since its beginning. In these 40 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 provide irradiation services to the scientific community and also to the industry.

Since startup more than 185 core configurations have been installed and around 150 fuel element assemblies used. From these 150 fuel assemblies, 40 are located in the dry storage, 80 inside the storage pool and 30 inside the core.

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.

The spent fuel assemblies in the dry storage are those of the first load (first cycle), which presented corrosion and fission products release at the earlier stages of reactor operation, and their burnups are almost zero.

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 at the spent fuel racks.

Pool water radiochemistry analysis 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 for aluminum, the old fuel assemblies show pitting corrosion. Some of these fuels present pitting corrosion nodules, visible by bore eye inspection, and these corrosion nodules may reach the fuel plate meat, exposing fission products to the pool water. This is due to galvanic corrosion because the spent fuel racks are made of stainless steel and the fuel cladding is made of aluminum.

This paper summarizes the work done at IPEN in

which the objective was to identify the leaking fuel assemblies at the storage pool and their ^{137}Cs leaking rate.[1]

II. 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, (ID number IEA-41 through IEA-80) and the HEU fuels of the second cycle (ID 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 not be so strong if the stainless steel rack would not be in contact with side plate or fuel plate.

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.

III. SIPPING TEST

Procedure. In order to perform the sipping test, the irradiated fuel assemblies were withdrawn from the spent fuel storage rack, had 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 1. 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.

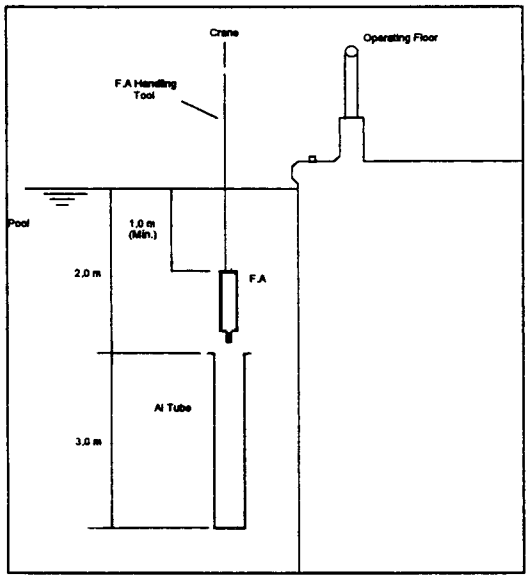

Figure 1 . Loading the F.A into the Al Tube.

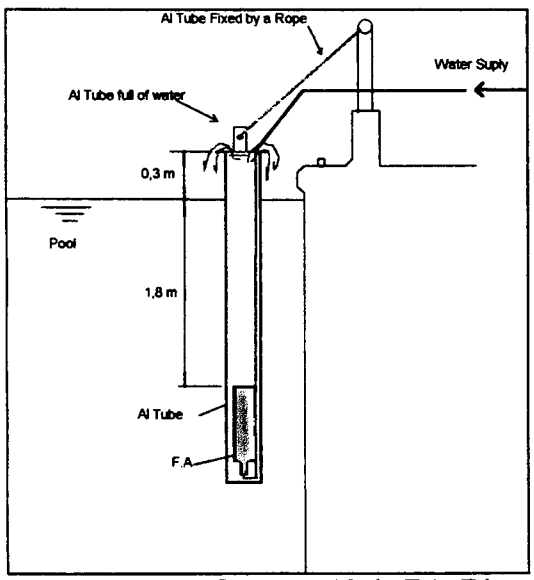

Figure 2 . Sipping Sequence with the Tube Edge out of the Pool.

Gamma-Ray Spectrometry. The gamma-ray spectrometry analysis was carried out with a shielded ORTEC HPGe detector with 130 cm^3 of volume, 2.0 keV of resolution and 25% of efficiency 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 efficiency for the 661.6 keV gamma-ray under fixed geometry conditions was performed using a standard solution of $1.15x10^5$ Bq of ^{137}Cs contained in a plastic bottle identical to the ones used to collect sipping water samples.

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.

Figure 3 shows the laboratory system background gamma-ray spectrum.

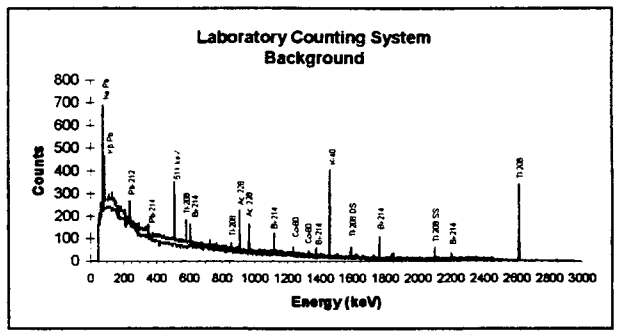

Figure 3 . Laboratory System Background Gamma-Ray Spectrum.

Results. The following observations can be taken from results of sipping:

- 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. 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.

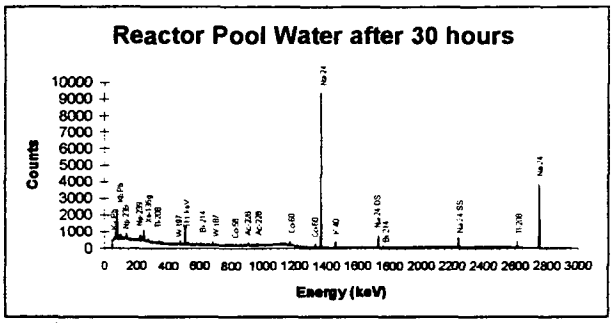

Figure 4 . Pool Water Sample Gamma-Ray Spectrum

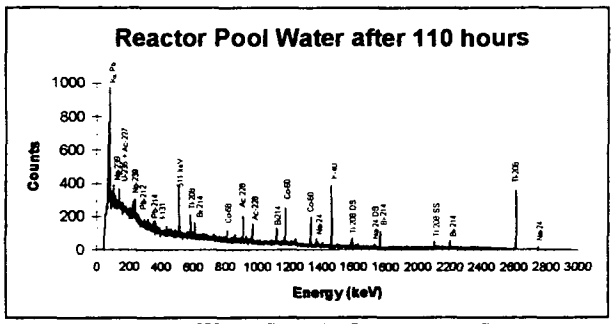

Figure 5 . Pool Water Sample Gamma-Ray Spectrum

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 or very low leaking fuel assembly (IEA-49) and one leaking fuel assembly (IEA-53), showing also the difference in the background spectra.

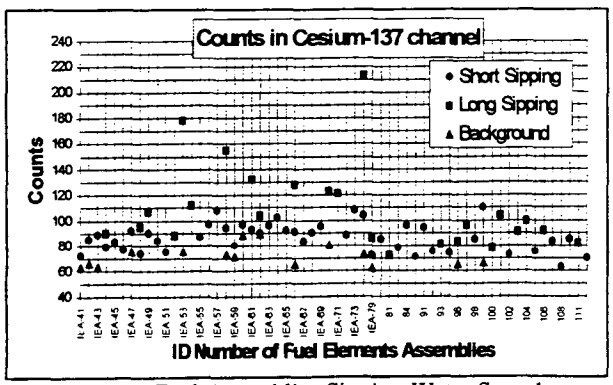

Figure 6 . Fuel Assemblies Sipping Water Samples Counting Results.

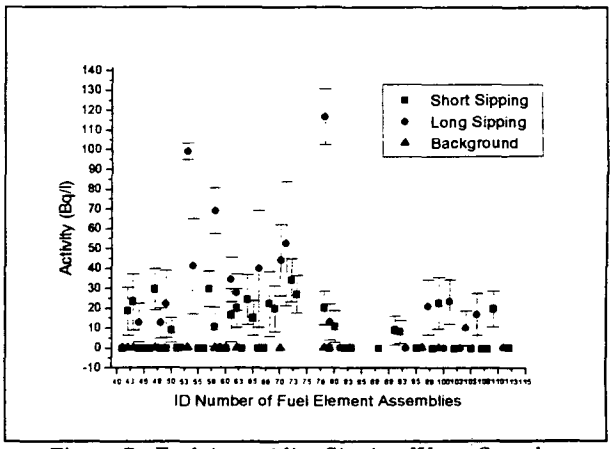

Figure 7 . Fuel Assemblies Sipping Water Samples Activity Results.

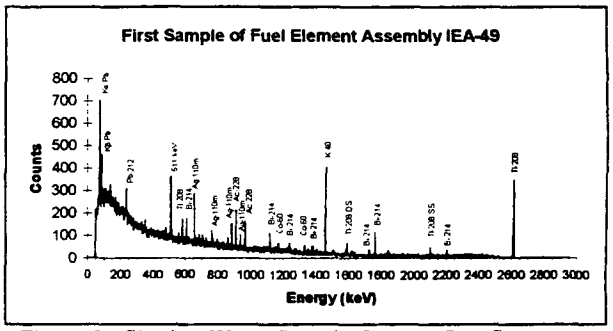

Figure 8 . Sipping Water Sample Gamma-Ray Spectrum.

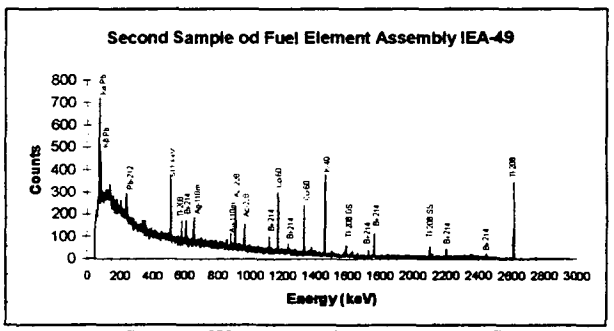

Figure 9 . Sipping Water Sample Gamma-Ray Spectrum.

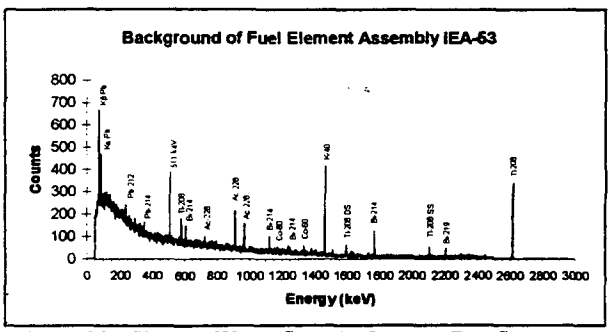

Figure 10 . Sipping Water Sample Gamma-Ray Spectrum.

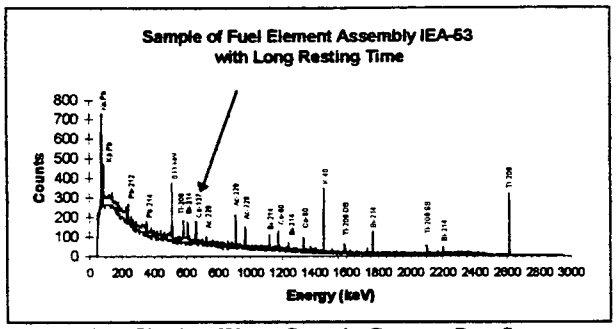

Figure 11 . Sipping Water Sample Gamma-Ray Spectrum.

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.

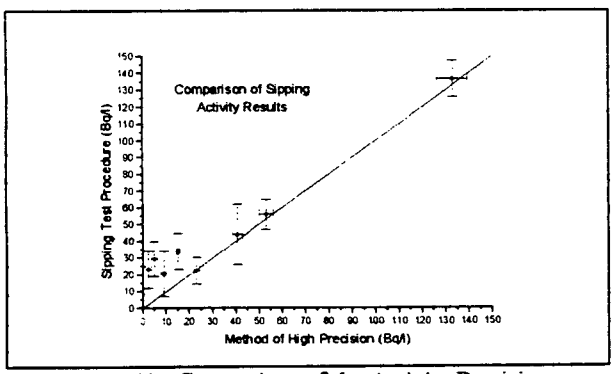

Figure 12 . Comparison of the Activity Precision.

IV. RELATION BETWEEN VISUAL INSPECTION AND SIPPING

It is interesting to correlate the visual inspection pattern with the sipping results. Table 1 shows this correlation. 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 1 . Correlation between Visual Inspection and Sipping Activity of 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*		

V. DETERMINATION OF ^{137}Cs LEAKING RATE

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.

It was observed that there is no evident difference between the results before and after cleaning some pitting corrosion nodules. The differences are more evident upon the sipping resting time. Figure 13 shows the activity along sipping resting time for fuel assembly IEA-53. The results show a straight line with a constant leaking rate of ~0.2 Bq/l.min or ~14 Bq/min. It also compares the activity before and after cleaning and one can see that there is almost no difference at the leaking rate.

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 for canning leaking fuel assemblies.

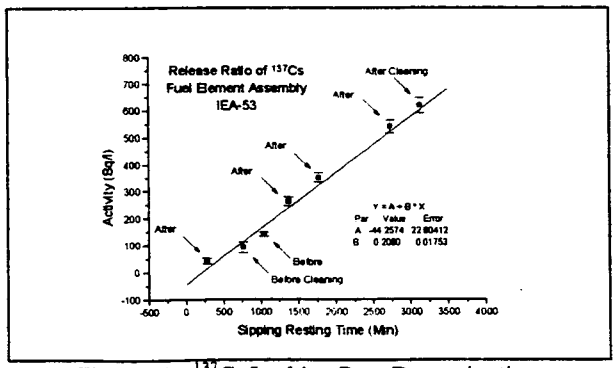

Figure 13 . ^{137}Cs Leaking Rate Determination.

VI. GAMMA-RAY SPECTROMETRY OF A PITTING CORROSION NODULE

A sample of a pitting corrosion nodule from a fuel plate was taken to perform gamma-ray spectrometry analysis. Figure 14 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.

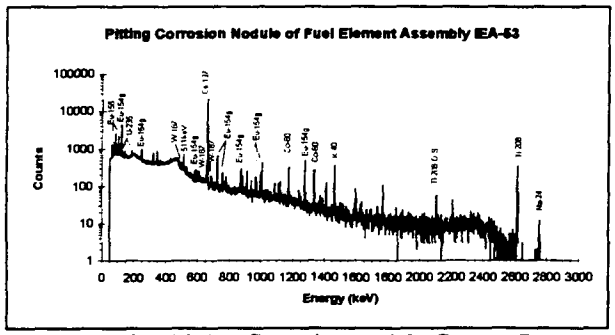

Figure 14 . Pitting Corrosion Nodule-Gamma Ray Spectrum

VII. 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 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.

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