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EXPERIENCE ON WET STORAGE SPENT FUEL SIPPING AT IEA-R1 BRAZILIAN RESEARCH REACTOR*

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Abstract—The IEA-R1 research reactor of the Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN-SP) is a pool type reactor of B&W design, that has been operating since 1957 at a power of 2 MW. Irradiated (spent) fuels have been stored at the facility during the various years of operation. At present there are 40 spent fuel assemblies at dry storage, 79 spent fuel assemblies at wet storage and 30 fuel assemblies in the core. The oldest fuels are of United States origin, made with U–Al alloy, both of LEU and HEU MTR fuel type. Many of these fuel assemblies have corrosion pits along their lateral fuel plates. These pits originate by galvanic corrosion between the fuel plate and the stainless steel storage racks. As a consequence of the possibility of sending the irradiated old fuels back the U.S.A., sipping tests were performed with the spent fuel assemblies. The reason for this was to evaluate their ^{137}Cs leaking rate, if any. This work describes the procedure and methodology used to perform the sipping tests with the fuel assemblies at the storage pool, and presents the results obtained for the ^{137}Cs sipping water activity for each fuel assembly. A correlation is made between the corrosion pits and the activity values measured. A ^{137}Cs leaking rate is determined and compared to the criteria established for canning spent fuel assemblies before shipment. © 1998 Elsevier Science Ltd.

1. 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. (B&W, U.S.A.) in accordance with specifications furnished by the Brazilian Nuclear Energy Commission (CNEN), and was financed by the U.S. Atoms for Peace Program.

The first criticality occurred on 16 September 1957, and was the first achieved in South America. Although designed to operate at 5 MW, the IEA-R1 has been operating at 2 MW from the beginning. In 39 years of operation the IEA-R1 has been used to perform

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research in nuclear and solid state physics, radiochemistry and radiobiology, to produce some radioisotopes and to provide irradiation services for the scientific community and industry.

From its start-up to present time (October 1996), 181 core configurations have been installed and around 150 fuel element assemblies used. The reactor has operated for 40 h per week (8 h day^{-1}) for most of its lifetime, but since the beginning of 1996 has operated in one continuous cycle of 64 h per week.

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

The first cycle corresponds to the first core of the reactor. It was composed of U–Al alloy fuel with 20% enrichment, and had 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 the brazing flux used to fix the fuel plates to the support plates. The fuels were replaced in 1958 by new ones, also produced by B&W. They were identical to the earlier ones (U–Al alloy, 20% enrichment, 19 curved fuel plates) but brazing was not used during their assembly. The fuel plates were fixed mechanically to the support plates. These fuels operated with good performance up to the discharge burn up used at that time (~20% of the ^{235}U atoms).

The second cycle corresponds to a complete substitution of the core. Fuel made with a U–Al alloy, 93% enrichment, having 18 flat fuel plates were bought from UNC (U.S.A.). At this time the core was converted from LEU (low enriched uranium) to HEU (high enriched uranium). 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 a rod type to a fork type (plate type). The control fuel element assemblies were fabricated by CERCA (France), using U–Al alloy, 93% enrichment, and flat plates.

The third cycle is characterized by the restriction of the HEU fuel supply. IPEN/CNEN-SP bought, from NUKEM (Germany), five fuel element assemblies of UAl_x–Al dispersion type, with 20% 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 in the HEU fuel plate and the geometry of the fuel element assembly was also the same. With this partial LEU core load, the HEU fuels, that stayed in core, began to have higher burnup and the number of fuel element assemblies used in the reactor core had to be increased due to reactivity needs.

The fourth cycle began with IPEN/CNEN-SP deciding to fabricate its own fuel and to replace, gradually, the high burn up HEU fuels in the core. At that time IPEN/CNEN-SP already had a good knowledge and experience of core engineering, fuel engineering and fuel fabrication, and so the decision to produce MTR (Material Testing Reactor) fuels for the IEA-R1 was a natural way to maintain reactor operation. The IPEN/CNEN-SP fuels are of a U_3O_8-Al dispersion type, with 20% enrichment and are 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.

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

The reactor pool is divided in two sections. The first is the core pool, where the core and irradiation facilities are located. The second is the spent fuel pool, where the spent fuel storage racks are laid. Fig. 1 shows the reactor pool with the two sections.

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. burn up (% ^{235}U)						
Standard	0	~30	~50		~50	~30**
Control	0	~40		~50		~20**
Partial	0	~10	~43			~20

*Partial fuel element assembly; **up to October/1996.

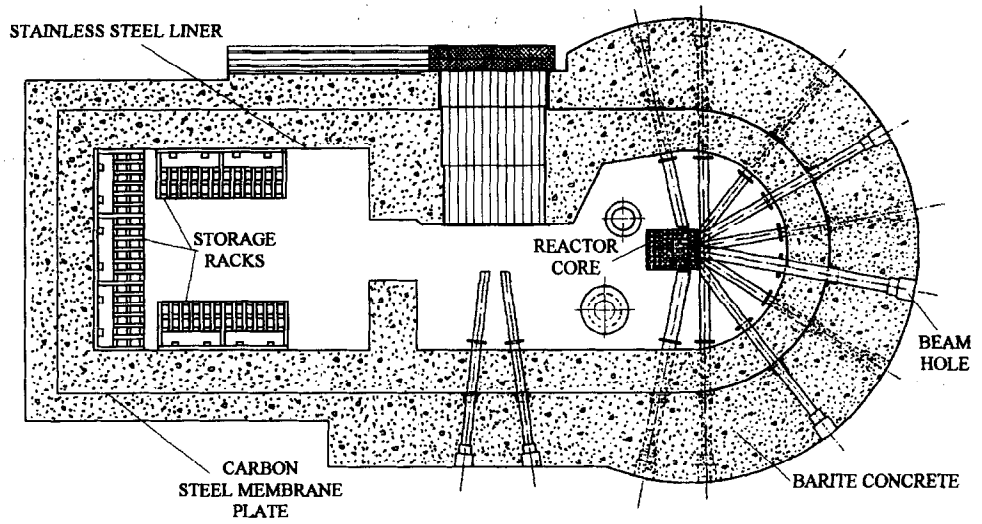

Fig. 1. Horizontal section of the IEA-R1 reactor pool.

The spent fuel assemblies in dry storage are those from the first load (first cycle) that presented corrosion and fission products released at the earlier stages of reactor operation. Their burn ups are almost zero, but some of them have more than $1\,R\,h^{-1}$ of dose rate at the fuel assembly surface. These fuel assemblies are wrapped in plastic bags, and the discussion of whether or not to can 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, with the exception of the first load. Some of these spent fuels have been inside the pool for almost 40 years and inside the spent fuel racks for more than 30 years. Some of these fuels present pitting corrosion nodules, visible to the naked eye, and these corrosion nodules may reach the fuel plate meat, exposing fission products to the pool water.

Pool water radiochemical analysis, along the various years of sampling, have shown a low specific activity of ^{137}Cs (less than $5\,Bq\,l^{-1}$). This means that there are some leaking fuel assemblies. This activity is so low because there is a constant water cleaning system in operation.

The pool water quality is excellent. The pH is kept between 5.5 and 6.5, conductivity is below $2\,\mu S\,cm^{-1}$ and chlorides are less than 0.5 ppm. Despite these excellent water characteristics, the old fuel assemblies show pitting corrosion. This is due to galvanic corrosion because the spent fuel storage racks are made of stainless steel and the fuel cladding is made of aluminum. In the 1970s the ceramic reactor pool wall was changed to a stainless steel liner and the spent fuel storage racks were changed from aluminum alloy to stainless steel. When this was done the possibility of galvanic corrosion between the fuel assemblies cladding and the frame of the storage racks was not taken into account.

Over the next 10 years, the U.S. Government intends to return to the U.S.A. all spent fuels of American origin from around the world. In June 1996, a U.S. Department of Energy (US-DOE) member of staff came to IPEN/CNEN-SP to discuss the shipment of these fuels. One of the points of discussion was the necessity of canning leaking spent fuels. The proposal of the DOE staff was to perform sipping tests inside the shipping cask,

with all fuels loaded. The IPEN/CNEN-SP staff pointed out that this would not be a good solution because if any cesium release was detected, all fuel assemblies would be sent back to the pool storage and an investigation would have to be carried out in order to identify the leaking fuel assembly. IPEN/CNEN-SP proposed to perform an individual sipping test in each fuel assembly before transference to the shipping cask. Once the schedule for shipment was very tight, sipping tests had to be done quickly. IPEN/CNEN-SP performed visual inspection and sipping tests in 60 spent fuel assemblies in 2 weeks, and selected the fuel assemblies that could be shipped without canning.

This work was verified, in late July 1996, by another group from DOE-Savannah River Site (DOE-SRS) that came to IPEN/CNEN-SP. This group agreed with the IPEN/CNEN-SP methodology and results for the sipping tests. They asked for additional sipping tests to confirm the results, carried out some visual inspection of the fuel assemblies using an underwater camera, and also visual inspection of some fuels in dry storage.

This paper describes the work done by IPEN/CNEN-SP in order to identify the fuel assemblies that could be sent back to U.S.A. without canning (Perrotta *et al.*, 1996).

2. VISUAL INSPECTION

The visual inspection was done by the naked eye with the fuel assembly inside the pool with 2 m of depth. As IPEN/CNEN-SP did not have an underwater camera, this visual inspection was just done to verify if there were any visual corrosion pits at the outer surface of the outer fuel plates and the pattern of such occurrences. The inspected fuel assemblies were the LEU fuels of the second core, first cycle (ID number IEA-41 to IEA-80) and the HEU fuels of the second cycle (ID number 81 to 118).

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

1. there are corrosion pits along the interface of external fuel plate with the side plate. The pits seem to be located at the fuel plate side. These regions are normally out of the fuel plate active width, although there is no way to confirm whether the corrosion pit is deep enough to pass through to the fuel plate meat;
2. there are some fuel assemblies with corrosion pits along the fuel plates in regions of the plate where there is contact between the fuel plate surface and the surface of the spent rack frame;
3. 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 is close contact between the fuel assembly and the rack frame;
4. 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 very strong evidence that the pitting corrosion in the fuel assemblies is due to galvanic phenomena;
5. the lateral support plates show few corrosion pits in some fuel assemblies and most of them in the region where there is contact with the rack frame;
6. as the visual inspection did not use any equipment, it was not possible to see any corrosion 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 for the pitting corrosion is the galvanic pair formed between the fuel assembly and the support rack. The material of fuel plate cladding is Al 1060, the side plate is made of Al 6061 T6, and the support rack is made of SS AISI 304. Also there is some galvanic pairing between the fuel plate cladding and the side plate, but this effect would not be so strong if the stainless steel racks were not in contact with the side plate or the fuel plate.

Table 2 shows a resume of the visual inspection.

IPEN/CNEN-SP already had 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 the 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 a lower intensity than that 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 the internal fuel plates do not have (or at least have few) corrosion pits.

3. SIPPING TEST

3.1. Procedure

In order to perform the sipping test, the irradiated fuel assemblies were withdrawn from the spent fuel storage rack, a rigid plastic pipe was connected to their bottom nozzle, and they were placed separately inside an aluminum sipping tube (120 mm dia., 3 m length, 33 l volume), as shown in Fig. 2. This first part of the procedure was always done with the fuel assembly positioned at a depth of approx. 2 m inside the pool water, and was 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. The most important of these residual

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

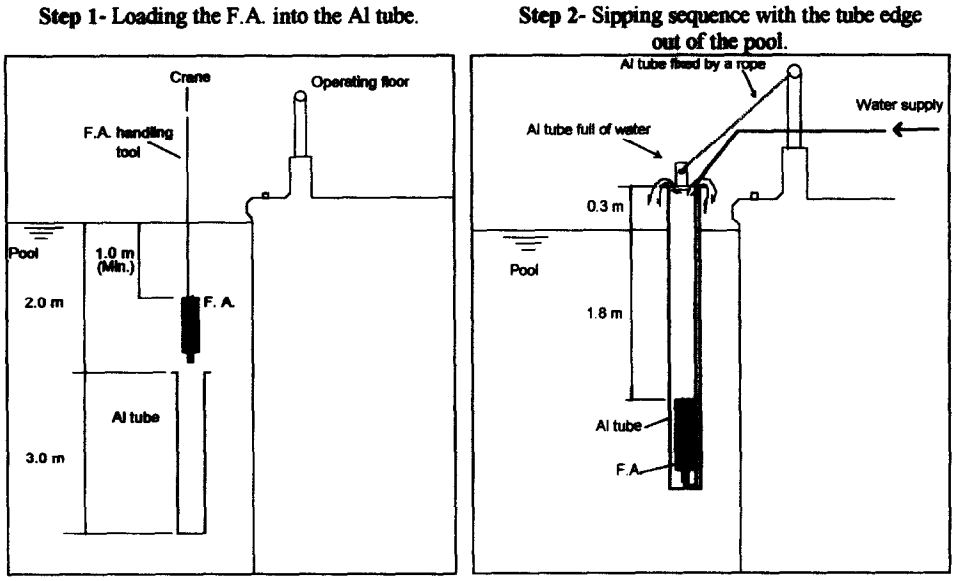

Fig. 2. Sipping scheme.

radionuclides is ^{24}Na, produced by means of the nuclear reaction $^{27}Al(n,\alpha)^{24}Na$. This reaction occurs in the fuel plate cladding of the reactor core assemblies during irradiation.

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 l of demineralized water was then injected through the plastic pipe and flushed through the fuel assembly in order to wash it, as shown in Fig. 2. After that, a background sample of the tube water was collected in a small plastic bottle (100 ml of volume) 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 4 h. Once the resting time was finished, compressed air was injected through the plastic pipe and flushed through the fuel assembly for 2 min 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 volume) 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 2 weeks.

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

3.2. Gamma-ray spectrometry

The gamma-ray spectrometry analysis was carried out with a shielded ORTEC HP Ge detector of volume 130 cm^3 with a resolution of 2.0 keV and an efficiency of 25% for the 1332 keV line of ^{60}Co. The gamma-ray energy range taken for the analysis was from 50 to

2800 keV. The data acquisition was performed with an ORTEC multichannel analyzer system coupled to a microcomputer through a control interface. Gamma-ray spectra were taken in runs of 4000 s of live time each.

The calibration of energy was obtained using a total of 40 peaks from 14 calibration sources (Knoll, 1989). 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 $\epsilon=(8.21\pm0.25)\times10^{-3}$.

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 measured a sample of demineralized water in order to determine the natural gamma-ray background. The second measured a sample of the pool water in order to determine the general gamma-ray background.

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 (Gouffon, 1983). 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

$$A=\frac{Area}{\epsilon.T.I_\gamma.Vol} \tag{1}$$

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.

3.3. Results

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

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

1. samples
   - background — Analysis of the gamma-ray spectrum from the water sample taken after water flushing through the fuel assembly, and before resting time;
   - first — Analysis of gamma-ray spectrum from the first water sample taken after resting time (called short sipping, within 4 h of resting time);
   - second — Analysis of gamma-ray spectrum from the second water sample taken after resting time (called long sipping, $\geq$8 h of resting time).

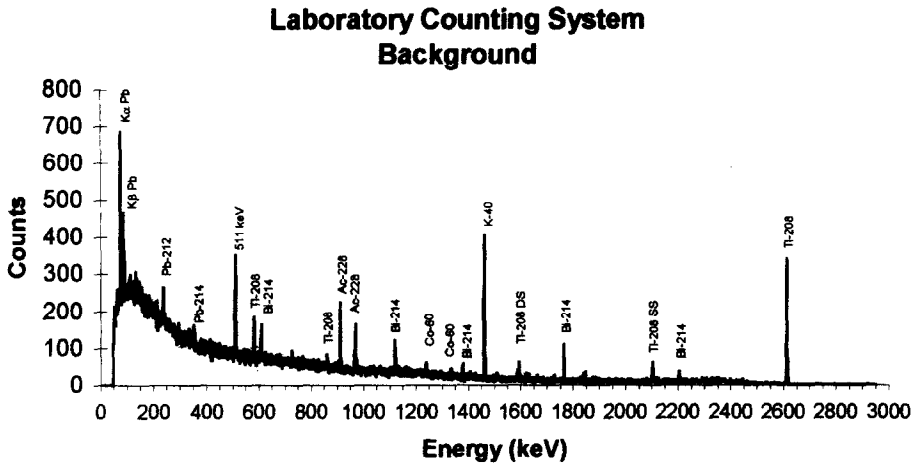

Fig. 3. Gamma-ray background spectrum of the laboratory system.

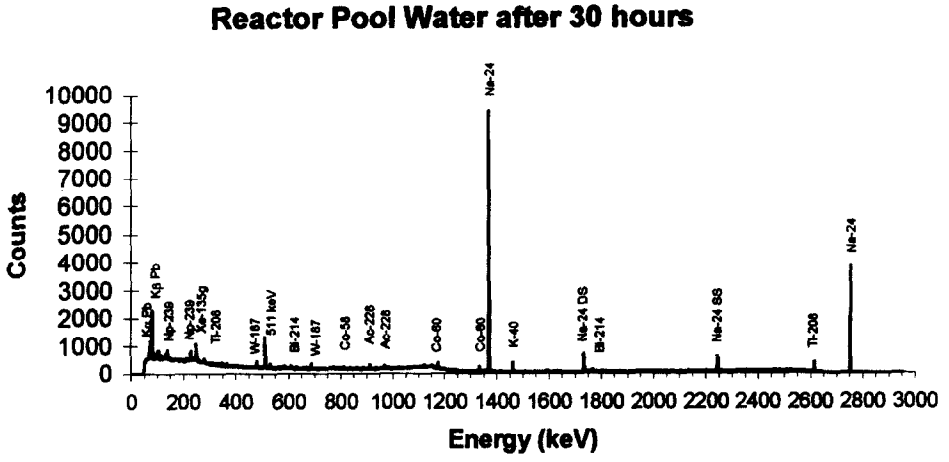

Fig. 4. Gamma-ray spectrum of the pool water sample, recorded 30 h after sampling.

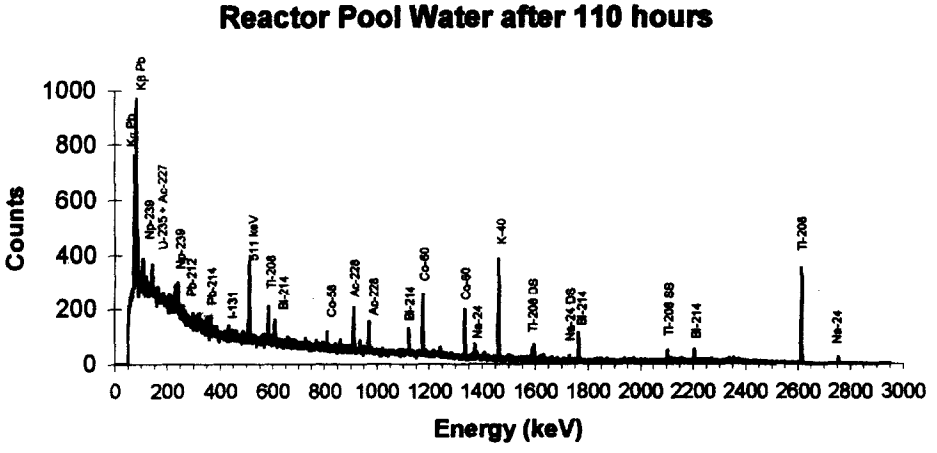

Fig. 5. Gamma-ray spectrum of the pool water sample, recorded 110 h after sampling.

Table 3. Fuel element assemblies sipping for 4000s of counting time (exceptions are indicated)

Number	Sample	Resting time (min)	Sampling date and time		Measurement date and time		Photopeak and counts in 661.6 keV channel		Photopeak area	Specific activity of Cs-137 ($Bq\,l^{-1}$)
IEA-41	BG	—	02/07	09:00	02/07	09:17	No	63	—	—
	First	280	02/07	13:40	02/07	16:20	No	72	—	—
IEA-42	BG	—	02/07	09:40	02/07	22:14	No	66	—	—
	First	255	02/07	13:55	03/07	08:20	Yes	85	52 ± 34	19 ± 12
IEA-43	BG	—	02/07	10:17	03/07	09:40	No	63	—	—
	First	297	02/07	15:14	03/07	11:15	Yes	89	65 ± 39	23 ± 14
IEA-44	First	180	02/07	18:14	03/07	19:31	No	79	—	—
	Second	795	03/07	08:30	04/07	10:19	Yes	90	36 ± 27	13 ± 10
IEA-45	First	262	03/07	13:32	04/07	14:41	No	83	—	—
IEA-46	First	240	03/07	13:52	04/07	18:19	No	77	—	—
IEA-47	BG*	—	01/07	12:16	01/07	19:05	No	135	—	—
	First*	264	01/07	16:40	01/07	17:01	Yes	165	149 ± 52	30 ± 10
IEA-48	First	225	03/07	18:50	05/07	08:09	No	74	—	—
	Second	850	04/07	08:39	05/07	12:36	Yes	95	35 ± 22	13 ± 8
IEA-49	First	185	03/07	18:05	05/07	13:47	No	90	—	—
	Second	820	04/07	08:40	05/07	14:56	Yes	107	62 ± 47	22 ± 17
IEA-50	First	247	04/07	14:02	05/07	16:19	Yes	84	25 ± 17	9 ± 6
IEA-51	First	235	04/07	14:20	05/07	17:27	No	75	—	—
IEA-52	First	790	05/07	08:35	08/07	15:27	No	88	—	—
IEA-53	BG-1	—	04/07	17:55	09/07	12:22	No	75	—	—
	First	765	05/07	08:40	08/07	16:43	Yes	178	261 ± 55	93 ± 20
	First**	765	05/07	08:40	12/07	20:02	Yes	3672	5984 ± 171	99 ± 4
	BG-2	—	10/07	16:13	16/07	08:45	No	69	—	—
	Second	1041	11/07	09:34	16/07	10:38	Yes	194	422 ± 32	151 ± 12
	Second	1041	11/07	09:34	23/07	15:30	Yes	261	382 ± 30	137 ± 11
IEA-54	First	740	05/07	08:45	08/07	18:05	Yes	112	115 ± 67	41 ± 24
IEA-55	First	345	05/07	15:33	09/07	07:02	No	87	—	—
IEA-56	First	320	05/07	15:39	09/07	08:18	No	97	—	—
IEA-57	First	310	05/07	15:43	09/07	09:27	Yes	108	83 ± 25	30 ± 9
IEA-58	BG	—	17/07	11:45	18/07	10.05	No	73	—	—
	First	304	17/07	16:49	18/07	12:42	Yes	94	30 ± 27	11 ± 10
	Second	1080	18/07	10:52	18/07	14:42	Yes	155	193 ± 32	69 ± 12

Table 3—*contd*

IEA-59	BG	—	02/07	09:10	02/07	15:09	No	71	—	—
	First	275	02/07	13:45	02/07	17:58	No	80	—	—
IEA-60	BG	—	02/07	10:00	03/07	16:12	No	88	—	—
	First	310	02/07	15:10	03/07	17:23	No	97	—	—
IEA-61	First	200	02/07	18:10	04/07	11:44	Yes	93	46 ± 37	16 ± 13
	Second	815	03/07	08:25	04/07	13:11	Yes	132	96 ± 31	34 ± 11
IEA-62	BG	—	02/07	15:30	02/07	19:40	No	89	—	—
	BG	—	02/07	15:30	17/07	17:33	No	67	—	—
	First	167	02/07	18:17	03/07	14:52	Yes	90	56 ± 27	20 ± 10
	First	167	02/07	18:17	17/07	18:45	Yes	92	79 ± 46	28 ± 16
	Second	780	03/07	08:31	03/07	13:08	Yes	104	78 ± 26	28 ± 9
	Second	780	03/07	08:31	17/07	19:56	Yes	121	113 ± 32	40 ± 12
IEA-63	First	240	03/07	13:37	04/07	19:31	No	96	—	—
IEA-64	First	253	03/07	14:20	05/07	18:38	Yes	102	68 ± 35	24 ± 13
IEA-65	First	243	03/07	14:25	05/07	19:49	Yes	92	42 ± 24	15 ± 9
IEA-66	BG	—	03/07	14:42	17/07	10:07	No	65	—	—
	First	200	03/07	18:02	08/07	07:58	No	91	—	—
	First	200	03/07	18:02	17/07	14:11	No	81	—	—
	Second	834	04/07	08:36	08/07	09:24	Yes	128	111 ± 82	40 ± 29
	Second	834	04/07	08:36	17/07	15:27	Yes	150	125 ± 25	45 ± 9
IEA-67	First	266	04/07	14:00	08/07	10:36	No	83	—	—
IEA-68	First	237	04/07	14:07	08/07	12:06	Yes	90	62 ± 45	22 ± 16
IEA-69	First	225	04/07	14:25	08/07	14:02	Yes	95	55 ± 32	20 ± 11
IEA-70	BG	—	04/07	17:40	12/07	17:18	No	80	—	—
	First	780	05/07	08:37	09/07	10:40	Yes	123	123 ± 50	44 ± 18
IEA-71	First	752	05/07	08:42	09/07	13:30	Yes	121	147 ± 87	53 ± 31
IEA-72	First	390	05/07	15:30	09/07	14:54	Yes	88	95 ± 30	34 ± 11
IEA-73	First	332	05/07	15:35	09/07	16:23	Yes	108	75 ± 26	27 ± 9
IEA-78	BG	—	17/07	12:00	18/07	15:53	No	73	—	—
	First	300	17/07	17:00	18/07	17:38	Yes	104	56 ± 24	20 ± 9
	Second	1072	18/07	10:56	18/07	19:44	Yes	213	326 ± 38	117 ± 14
IEA-79	BG	—	17/07	12:16	18/07	21:13	No	62	—	—
	First	295	17/07	17:11	18/07	22:35	No	72	—	—
	Second	1065	18/07	11:00	19/07	00:01	Yes	86	36 ± 25	13 ± 9

(continued)

Number	Sample	Resting time (min)	Sampling date and time		Measurement date and time		Photopeak and counts in 661.6 keV channel		Photopeak area	Specific activity of Cs-137 ($Bq\,l^{-1}$)
80	First	293	08/07	14:13	09/07	17:31	Yes	85	30 ± 22	11 ± 8
81	First	1048	09/07	08:45	09/07	18:45	No	72	—	—
83	First	274	08/07	14:16	10/07	08:39	No	78	—	—
84	First	307	09/07	15:17	10/07	09:58	No	96	—	—
88	First	260	08/07	14:20	10/07	11:22	No	71	—	—
91	First	269	09/07	15:25	11/07	19:03	Yes	94	25 ± 20	9 ± 7
92	First	221	08/07	14:28	10/07	14:39	Yes	75	23 ± 16	8 ± 6
93	First	1029	09/07	08:53	10/07	17:49	No	81	—	—
95	First	254	10/07	15:24	15/07	16:00	No	74	—	—
96	BG	—	08/07	15:53	17/07	08:12	No	65	—	—
	First	1023	09/07	08:56	10/07	19:25	No	83	—	—
97	First	1013	10/07	09:00	12/07	09:11	Yes	96	58 ± 38	21 ± 14
98	First	328	10/07	15:13	12/07	14:07	No	85	—	—
99	BG	—	09/07	9:53	17/07	12:07	No	66	—	—
	First	320	09/07	15:13	11/07	15:34	Yes	110	63 ± 36	23 ± 13
100	First	997	10/07	09:05	12/07	10:26	No	78	—	—
101	First	1057	09/07	08:40	10/07	15:54	Yes	104	65 ± 31	23 ± 11
102	First	264	10/07	15:16	15/07	11:14	No	73	—	—
103	First	993	10/07	09:15	12/07	11:40	No	91	—	—
104	First	1017	10/07	08:55	11/07	23:45	Yes	99	28 ± 24	10 ± 9
105	First	252	10/07	15:27	15/07	17:08	No	75	—	—
106	First	983	10/07	09:20	12/07	12:59	Yes	92	47 ± 29	17 ± 10
107	First	282	09/07	15:21	11/07	16:46	No	83	—	—
108	First	244	08/07	14:24	10/07	13:10	No	63	—	—
109	First	260	10/07	15:20	15/07	14:23	Yes	85	55 ± 25	20 ± 9
111	First	1007	09/07	09:02	11/07	09:15	No	82	—	—
112	First	335	09/07	15:10	11/07	10:45	No	70	—	—

BG, Background.
*, Counting time of 7200 s.
**, Counting time of 86 400 s.

Table 4. Background measurements

Number	Measurement date	Starting time of measure	Counting time (s)	Photopeak of 661.6 keV	Counts in 661.6 keV channel	Specific activity of Cs-137 ($Bq\,l^{-1}$)
1	02/07	09:17	7200	No	135	—
2	03/07	19:31	4000	No	63	—

Table 5. IEA-R1 reactor pool water measurements

Sampling date and time		Measurement date	Starting time of measure	Counting time (s)	Photopeak of 661.6 keV	Counts in 661.6 keV channel	Specific activity of Cs-137 ($Bq\,l^{-1}$)
10/07	10:20	11/07	19:03	4000	No	199	—
		12/07	18:29	4000	No	120	—
		15/07	08:45	4000	No	62	—
		16/07	11:49	4000	No	66	—

2. photopeak and counts in 661.6 keV channel
   - no — There is no defined photopeak in the channel related to the energy of 661.6 keV;
   - counts — Number of counts (for the established counting time of 4000 s) in the channel related to the energy of 661.6 keV;
   - photopeak area — Net number of counts (*Area*) under the 661.6 keV photopeak.
   - specific activity — Specific ^{137}Cs activity of the sample, calculated by means of equation (1).

The following observations can be taken from Table 3:

1. Some fuel assemblies show the photopeak of 661.6 keV.
2. For those fuel assemblies where two samples were taken, it is noticed that the specific activity increases with sipping resting time, indicating that the fuel assembly is really leaking. It is possible to determine a leaking rate for ^{137}Cs from such a fuel assembly. For example, for the fuel assembly number IEA-53 this rate is $\sim 0.2\,Bq\,l^{-1}\,min^{-1}$ or $\sim 14\,Bq\,min^{-1}$.
3. For some fuel assemblies the specific activity is very low and the associated error in the analysis is large. For longer counting times the specific activity is the same but the associated error decreases.
4. The analysis date influences the measured specific activity. This is due to the activation of the pool water that gives a high background level in the gamma-ray spectrum. The sipping tests were done with the reactor in operation and the activity of ^{24}Na (half-life of 15.02 h) in the water was high enough to increase background due to Compton continuum. Table 5 shows the pool water background and it is observed that the number of counts at the channel related to the energy of 661.6 keV decreases with the time of the analysis after sampling. Figs 4 and 5 show the gamma-ray spectra for the pool water, where one can see the influence of ^{24}Na in the background due to Compton continuum.
5. The water volume of the sipping tube is ~ 33 l. The volume of the fuel assembly is 2 l, and the volume lost during the compressed air injection is of 3 l. So the activity for the fuel assembly sipping is obtained multiplying the specific activity by 28 l.

Fig. 6 shows the counts of the 661.6 keV 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 count level of between 60 and 85 (for 4000 s). For those fuel assemblies that indicate the 661.6 keV photopeak the counts are higher than 90.

Fig. 7 shows the specific activity calculated for each fuel assembly sipping test. It can be seen that some fuel assemblies show a leaking pattern, and that most of the fuel assemblies have specific activities lower than 30 Bq l^{-1} (for sipping resting time of 4 h).

Figs 8–11 show the gamma-ray spectra for one non-leaking fuel assembly and one leaking fuel assembly.

The procedure for sampling and performing gamma-ray spectra analysis used a 100 ml water sample and 4000 s 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 specific activities, water samples of 850 ml were taken for some fuel assemblies. These samples were analyzed in a gamma-ray spectrometry system of a higher efficiency than the one used for the sipping tests. Also the counting time was increased to 50 000 s, which gives minor counting errors. Specific activities obtained from both measurements were compared. Fig. 12 shows the results of this comparison. One can see from this figure that the system used in the sipping tests always overestimated the results, and gave good results for specific activities higher than 20 Bq l^{-1}. For lower specific activities the associated deviation is bigger. As the level of specific activity is so small, the results of this comparison show that the system used in the sipping tests is very suitable for determining leaking fuel assemblies.

4. 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 (< 30 Bq l^{-1})	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^{-1} and < 60 Bq l^{-1})	61, 70	54, 71, 72	
High indication of Cs-137 (> 60 Bq l^{-1})	53, 58, 78		

5. DETERMINATION OF ^{137}Cs LEAKING RATE

The DOE-SRS group came to IPEN/CNEN-SP for the assessment of the fuel assemblies for shipment. This group agreed with the methodology used by IPEN/CNEN-SP in performing sipping tests and also with the results obtained. Some additional work concerning fuel assessment was done. One task was to remove some of the external fuel plate pitting corrosion nodules from 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, this was the one that showed the highest sipping activity among all fuel assemblies.

Table 7 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 see from Table 7 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 see from Table 7 the results for fuel assembly IEA-53. Fig. 13 shows the specific activity along sipping resting time. The results show a straight line with a constant leaking rate of $\sim 0.2\,Bq\,l^{-1}\,min^{-1}$ or $\sim 14\,Bq\,min^{-1}$. Fig. 14 compares the rate before and after cleaning and one can see that there is almost no difference.

As fuel assembly IEA-53 was the one with the highest leakage rate, this value of 14 Bq min^{-1} can be compared to the criteria established by DOE-SRS (staff communication, July 1996) for canning leaking fuel assemblies.

The DOE-SRS interim criteria presented to IPEN/CNEN-SP was $13.57\,\mu Ci\,h^{-1}$ per cask shipment, which is equivalent to $35.9\,pCi\,ml^{-1}\,h^{-1}$ for one fuel assembly (assuming 3.6 gallons of water per fuel assembly).

The value obtained at IPEN/CNEN-SP fuel assembly sipping, correcting to the volume of DOE-SRS criteria, is $0.64\,pCi\,ml^{-1}\,h^{-1}$ ($0.4\,Bq\,l^{-1}\,min^{-1}$), which is far below the criteria limit.

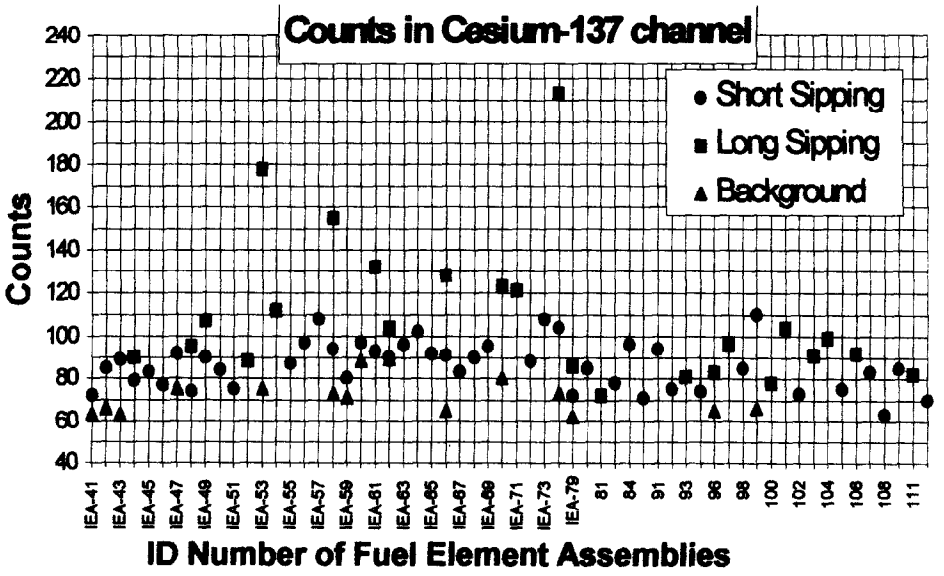

Fig. 6. Counting results for sipping water samples of fuel assemblies.

Table 7. Fuel element assemblies sipping before and after cleaning

Number	Sample	Before cleaning					After cleaning				
		Resting time (min)	Photopeak of 661.6 keV	Counts in 661.6 keV channel	Photopeak area	Specific activity of Cs-137 ($Bq\,l^{-1}$)	Resting time (min)	Photopeak of 661.6 keV	Counts in 661.6 keV channel	Photopeak area	Specific activity of Cs-137 ($Bq\,l^{-1}$)
IEA-41	First	280	No	72	—	—	240	No	82	—	—
IEA-45	First	262	No	83	—	—	240	No	68	—	—
IEA-53	BG	—	No	75	—	—	—	No	68	—	—
	First	765	Yes	178	261 ± 55	93 ± 20	275	Yes	147	119 ± 29	43 ± 10
	Second	1041	Yes	228	401 ± 22	143 ± 8	1370	Yes	418	740 ± 39	265 ± 16
	Third	—	—	—	—	—	1770	Yes	525	988 ± 43	353 ± 19
	Fourth	—	—	—	—	—	2730	Yes	778	1517 ± 47	543 ± 24
	Fifth	—	—	—	—	—	3127	Yes	773	1737 ± 56	622 ± 28
IEA-73	First	332	Yes	108	75 ± 26	27 ± 9	240	No	77	—	—
92	First	221	Yes	75	23 ± 16	8 ± 6	240	No	77	—	—
93	First	1029	No	81	—	—	315	No	75	—	—
97	First	1013	Yes	96	58 ± 38	21 ± 14	240	No	70	—	—
101	First	1057	Yes	104	65 ± 31	23 ± 11	240	No	75	—	—
103	First	993	No	91	—	—	240	Yes	89	27 ± 19	10 ± 7
104	First	1017	Yes	99	28 ± 24	10 ± 9	240	No	83	—	—
106	First	983	Yes	92	47 ± 29	17 ± 10	255	No	72	—	—

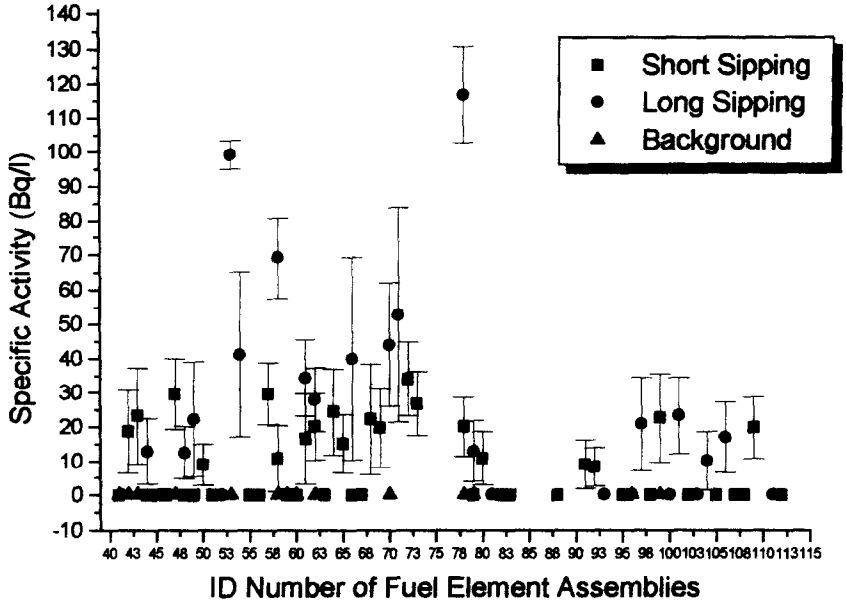

Fig. 7. Specific activity results for sipping water samples of fuel assemblies.

6. GAMMA-RAY SPECTROMETRY OF A PITTING CORROSION NODULE

A sample of a pitting corrosion nodule obtained from a fuel plate of fuel assembly IEA-53 was taken to perform gamma-ray spectrometry analysis. Fig. 15 shows the gamma-ray spectrum obtained. One is able to find a high level of ^{137}Cs activity and also ^{235}U, 155Eu, and ^{154}Eu activities. This indicates that the pitting corrosion nodule penetrates through the cladding up to the plate meat where there is uranium and fission products. It also indicates that besides ^{137}Cs, U and Eu isotopes are migrating from the meat to the

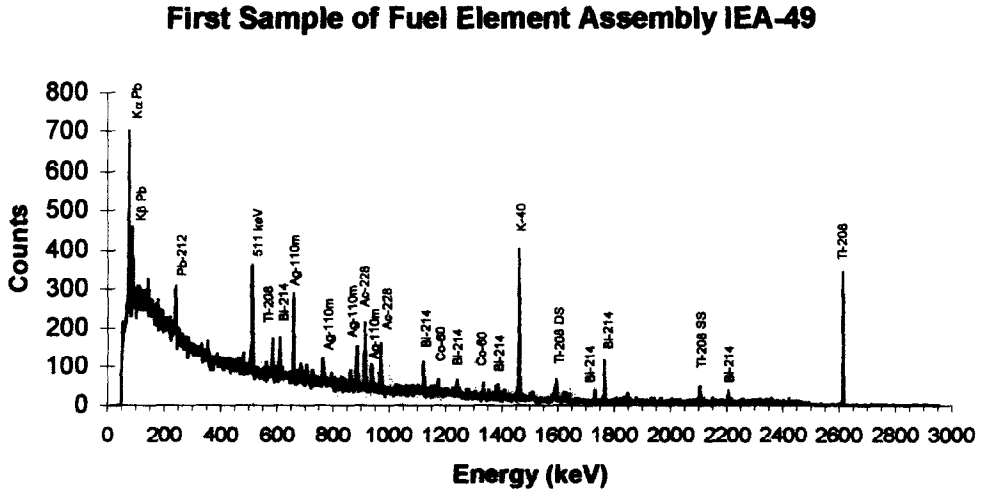

Fig. 8. Gamma-ray spectrum of the first sipping water sample of the fuel element assembly IEA-49.

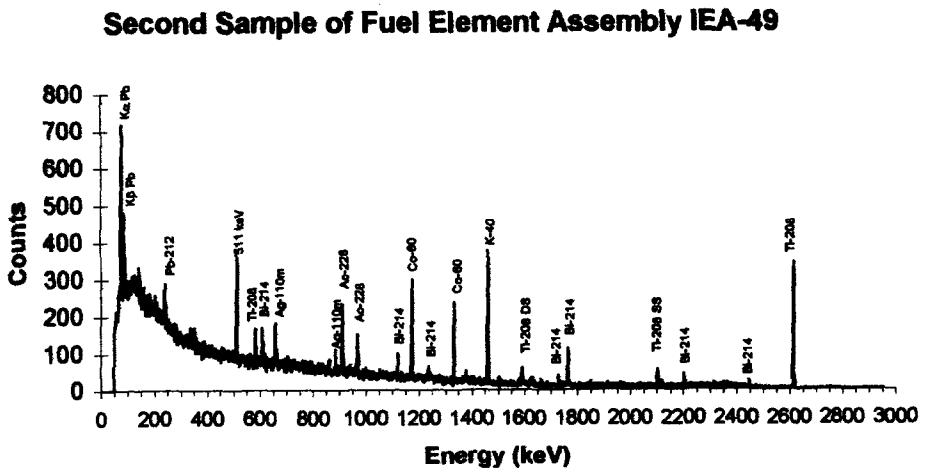

Fig. 9. Gamma-ray spectrum of the second sipping water sample of the fuel element assembly IEA-49.

nodule. U and Eu are not migrating (or migrating in a very small rate) to the water, although these elements were not found in the gamma-ray spectrometry of the sipping water. The mechanism of migration of U and fission products from the plate meat through the corrosion pit to water has to be studied in more depth.

The activity of ^{137}Cs in the pitting corrosion nodule sample was determined to be 2400 Bq or 5600 Bq g^{-1}. 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 (RSIC, 1987), taking into consideration the plate burn up and decay time to the date of the sipping test, is ~2 Ci or 7.4×10^{10} Bq. Considering the weight and volume of the U–Al alloy of the plate meat, the specific activity is 3.4×10^{9} Bq cm^{-3} or 7.5×10^{8} Bq g^{-1} (U–Al).

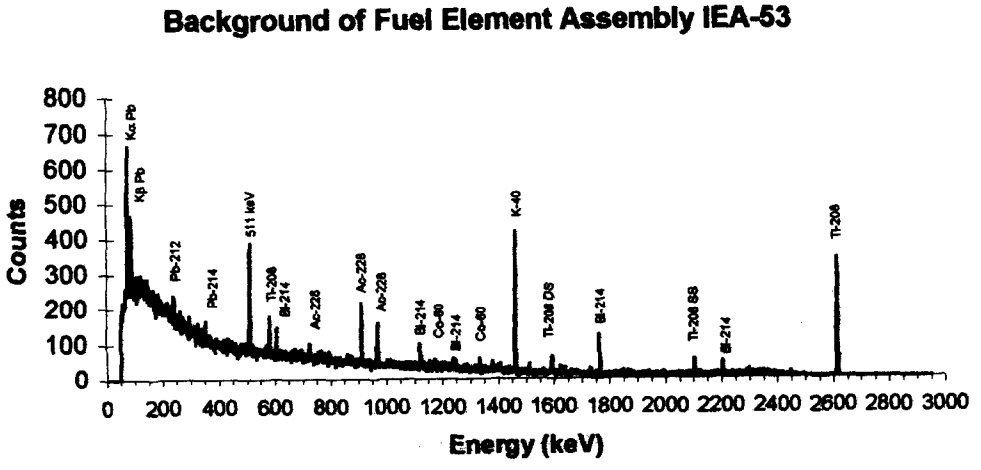

Fig. 10. Gamma-ray background spectrum of the fuel element assembly IEA-53.

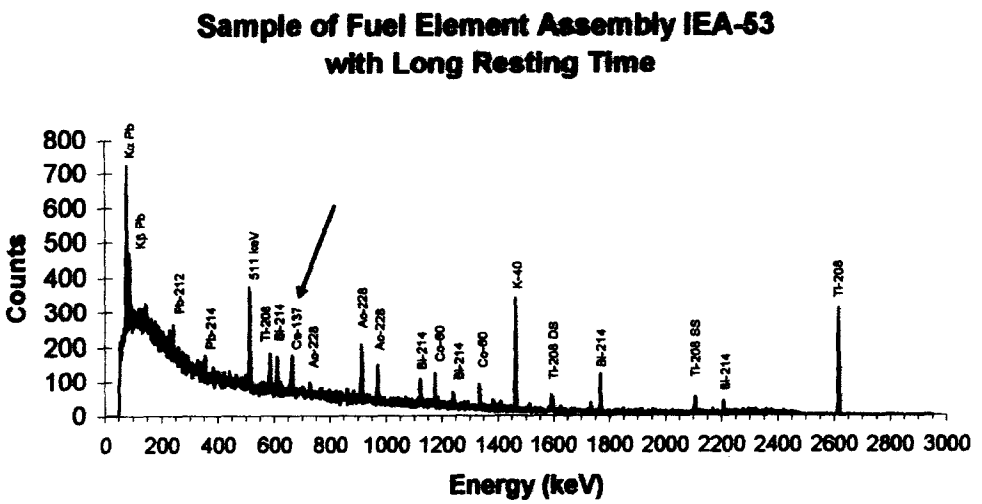

Fig. 11. Gamma-ray spectrum of the sipping water sample, collected after long resting time of the fuel element assembly IEA-53. The arrow indicates the 661.6 keV photopeak.

So, the three values of ^{137}Cs activities related to the fuel assembly IEA-53 are:

1. meat — 7.5×10^{8} Bq g^{-1}; or $\sim7.4\times10^{10}$ Bq/plate; or $\sim1.3\times10^{12}$ Bq/F.A.
2. corrosion pit nodule — 5.6×10^{3} Bq g^{-1}; or $\sim2.4\times10^{3}$ Bq/nodule.
3. sipping water sample — 14 Bq min^{-1}; or 0.2 Bq l^{-1} min^{-1}; or 2×10^{-5} Bq g^{-1} min^{-1}.

There is a migration of ^{137}Cs from the meat to the water, with a retention at the pitting corrosion nodule, which results in a leaking rate that is very far below the meat activity.

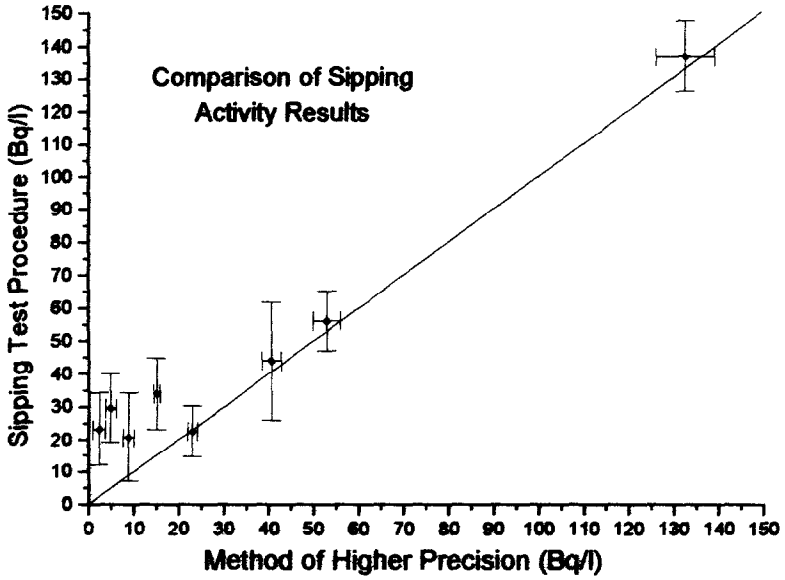

Fig. 12. Comparison of measured specific activities.

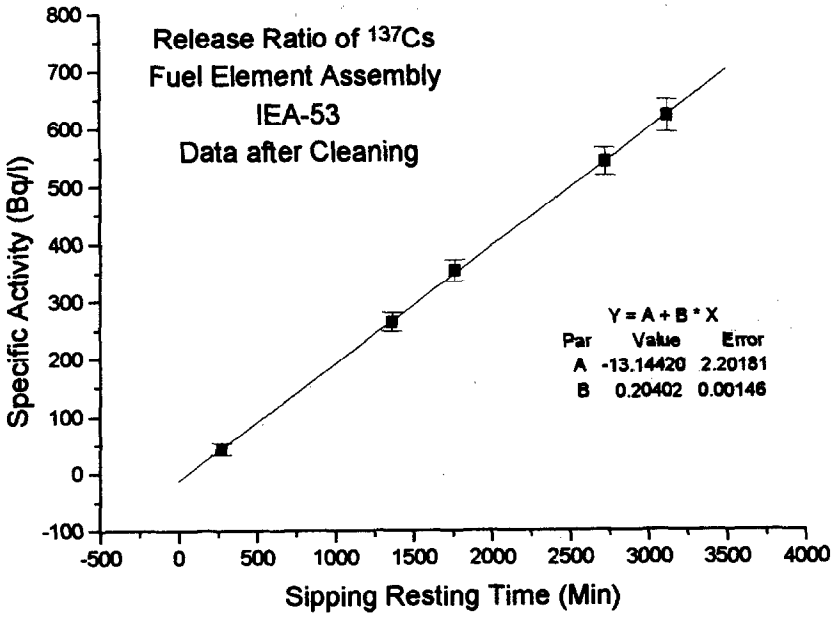

Fig. 13. ^{137}Cs leaking rate determination for the fuel element assembly IEA-53, considering only data obtained after cleaning.

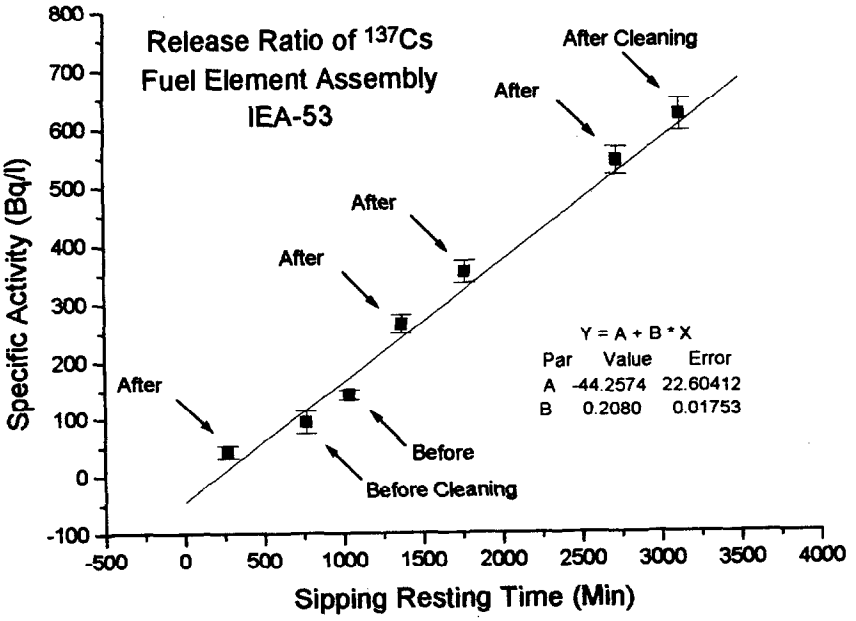

Fig. 14. ^{137}Cs leaking rate determination for the fuel element assembly IEA-53, considering data obtained before and after cleaning.

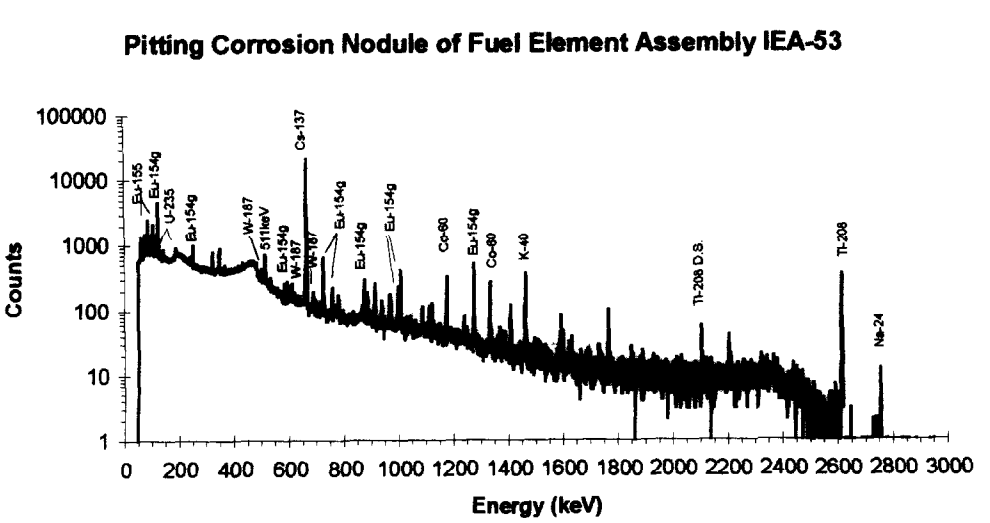

Fig. 15. Gamma-ray spectrum of a pitting corrosion nodule removed from the fuel element assembly IEA-53.

7. CONCLUSION

The visual inspection of the spent fuel assemblies stored at the pool storage racks showed that pitting corrosion is present in the external fuel plates of many fuel assemblies. Those which have been in 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 stainless steel storage racks.

The sipping test methodology and equipment used by IPEN/CNEN-SP was shown to be efficient in determining leakage of fission products from fuel assemblies.

A ^{137}Cs leaking rate of 14 Bq min^{-1} was obtained for the worst leaking fuel assembly. This value is far below the DOE-SRS criteria presented to IPEN/CNEN-SP for canning leaking MTR fuel assemblies.

Gamma-ray spectrometry of a pitting corrosion nodule shows the presence 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.

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