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Sipping tests on a failed irradiated MTR fuel element

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This work describes sipping tests performed on Material Testing Reactor (MTR) fuel elements of the IEA-R1 research reactor, in order to find out which one failed in the core during a routine operation. Radioactive iodine isotopes ^{131}I and ^{133}I, employed as failure monitors, were detected in samples corresponding to the failed fuel element. The specific activity of each sample, as well as the average leaking rate, were measured for ^{137}Cs. The nuclear fuels U_3O_8 – Al dispersion and U – Al alloy were compared concerning their measured average leaking rates of ^{137}Cs.

KEYWORDS: ***Fuel elements; Research reactors; Gamma spectroscopy; Fission products***

1. Introduction

Sipping test is a non-destructive technique employed to evaluate the structural integrity of the cladding of irradiated nuclear fuels, which is based on the detection of radioactive fission products leakage to the reactor coolant, usually by means of gamma-ray spectroscopy. Until recently, this technique has been used at IPEN/CNEN-SP only for surveillance of irradiated fuel elements stored for many years inside the spent fuel pool of the IEA-R1 research reactor [1], since no occurrence of fuel element failure in the reactor core was reported at all.

On July 30th 2001, during a routine operation of the reactor at 2 MW, the presence of gaseous radioactive fission products was detected in the hall of the reactor pool. Detailed analysis performed shortly after the incident showed that it was caused by failure in one of the 24 fuel elements of the reactor core. Sipping tests were then carried out not only to identify the failed fuel element, but also to measure the average leaking rate of fission products to the water.

The standard fuel elements used in the IEA-R1 research reactor are plate type, usually designated as Material Testing Reactor (MTR) fuel elements. A typical fuel element has 18 plane parallel fuel plates, mounted mechanically between two lateral aluminum holders with grooves, and its overall dimensions are (7.6 X 8.0) cm by 88.0 cm high. Each fuel plate consists of an aluminum cladding and a meat, where the nuclear fuel is located, containing approximately 10 g of ^{235}U. The fuel plate total thickness is 0.152 cm, and the distance between two successive plates is 0.289 cm [2].

2. Experiment

2.1. Procedure

In order to perform the sipping test, each fuel element was withdrawn from its position in the reactor core 8 hours after shutdown, a rigid plastic pipe was connected to its bottom nozzle and it was placed separately inside an aluminum sipping tube (12 cm diameter, 3 m length, approximately 33 *l* volume). These actions were monitored continuously by the radiological protection staff and always done with the fuel

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element positioned at a depth of at least 1 m inside the pool water, measured from its upper end, to ensure the dose equivalent at the surface of the pool to remain lower than 20 mrem/hour. Before the test, the sipping tube was washed with demineralized water to reduce, as much as possible, any kind of residual contamination of radionuclides.

The sipping tube, with the fuel element inside, was then lifted up and the top edge 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 element in order to wash it. Immediately after the washing, a background sample of the sipping tube water was collected in a small plastic bottle (110 m*l* volume) and submitted latter to gamma-ray spectroscopy measurements.

The fuel element was then left at rest inside the sipping tube during a time interval of at least 4 hours. Once the resting time was finished, compressed air was injected through the plastic pipe for 2 minutes, in order to homogenize the solution that might contain fission products released by the leaking fuel element. A sample of this solution was collected in a small plastic bottle (110 m*l* volume) and submitted latter to gamma-ray spectroscopy measurements.

After a new resting time, this turn of at least 8 hours, compressed air was injected through the plastic pipe for 2 minutes and a second sample was collected. All plastic bottles used for sampling were identical. Each collected sample was identified by a label fixed on the corresponding plastic bottle.

Once again, every stage of the work was monitored continuously by the radiological protection staff.

Sipping tests following this procedure were performed on all 24 fuel elements of the reactor core. Five identical aluminum sipping tubes were used simultaneously.

Regarding this procedure, it is important to emphasize the difference between resting time and sipping time. Resting time is the time interval between the collecting of two successive samples of sipping tube water. Sipping time is the time elapsed since the end of fuel element washing [1].

2.2. Gamma-ray spectroscopy measurements

Gamma-ray spectroscopy analysis were carried out with a shielded HPGe detector of volume 130 cm^3 with 1.71 keV resolution and 26.1 % relative efficiency for the 1332.5 keV gamma-ray of ^{60}Co [3]. The gamma-ray energy range taken for analysis was from 50 keV to 2800 keV. Data acquisition was performed with a 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 energy calibration of the HPGe detector was obtained using a total of 92 full-energy peaks from 15 calibration sources [4]. Gamma-ray energies were fitted as a second-degree polynomial function of the corresponding full-energy peak channels in the calibration spectra. The resulting calibration curve enabled the identification of all full-energy peaks present in every gamma-ray spectrum obtained from each collected sample. In these spectra, the presence of full-energy peaks corresponding to gamma-rays emitted by fission products indicates occurrence of cladding failure in the fuel element.

Concerning the identification of the failed fuel element, this qualitative analysis is enough. However, after the failed fuel element was identified, one decides to measure also the average leaking rate of the fission product ^{137}Cs. As a consequence, the samples corresponding to the failed fuel element were carefully stored during 6 months, to enable the decay of short-lived radionuclides, and measured not till then.

The efficiency calibration of the HPGe detector for the energy of 661.6 keV under fixed geometry conditions was performed using a standard solution of (100 ± 3) kBq 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 $\varepsilon = (7.66 \pm 0.23).10^{-3}$.

For each measurement, the plastic bottle containing the collected 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.

The 661.6 keV full-energy peak of each sipping spectrum, if any, was fitted by Gaussian function plus a parabolic curve for the continuous background using the computer code IDEFIX [5]. Using this

procedure, the net number of counts (*Area*) under the 661.6 keV full-energy peak was determined and therefore the specific activity *A* of ^{137}Cs in the solution as

$$A = \frac{Area}{\varepsilon \cdot T \cdot I_\gamma \cdot Vol} \cdot e^{\lambda \cdot t_d} \qquad (1)$$

where ε is the efficiency value mentioned before, T is the live time of measurement, I_γ is the gamma-ray absolute emission intensity, Vol is the volume of the sipping sample, λ is the decay constant of ^{137}Cs and t_d is the time interval elapsed between the date of sampling and the date of measurement.

Finally, the average leaking rate R of ^{137}Cs from the failed fuel element to water was determined by means of the following expression:

$$R = \frac{A_2 - A_1}{t_2 - t_1} \qquad (2)$$

where A_2 and A_1 are respectively the specific activities of ^{137}Cs in sipping water samples collected after sipping times equal to t_2 and t_1.

3. Results

As explained before, a sample of sipping tube water was collected immediately after the washing of each fuel element, to measure the background radiation by means of gamma-ray spectroscopy. A typical gamma-ray spectrum obtained from background measurements is shown in figure 1. The full-energy peaks in the spectrum were identified using the energy calibration. It is important to note the total absence of fission products. The full-energy peaks observed in the spectrum correspond to natural background and to activation products.

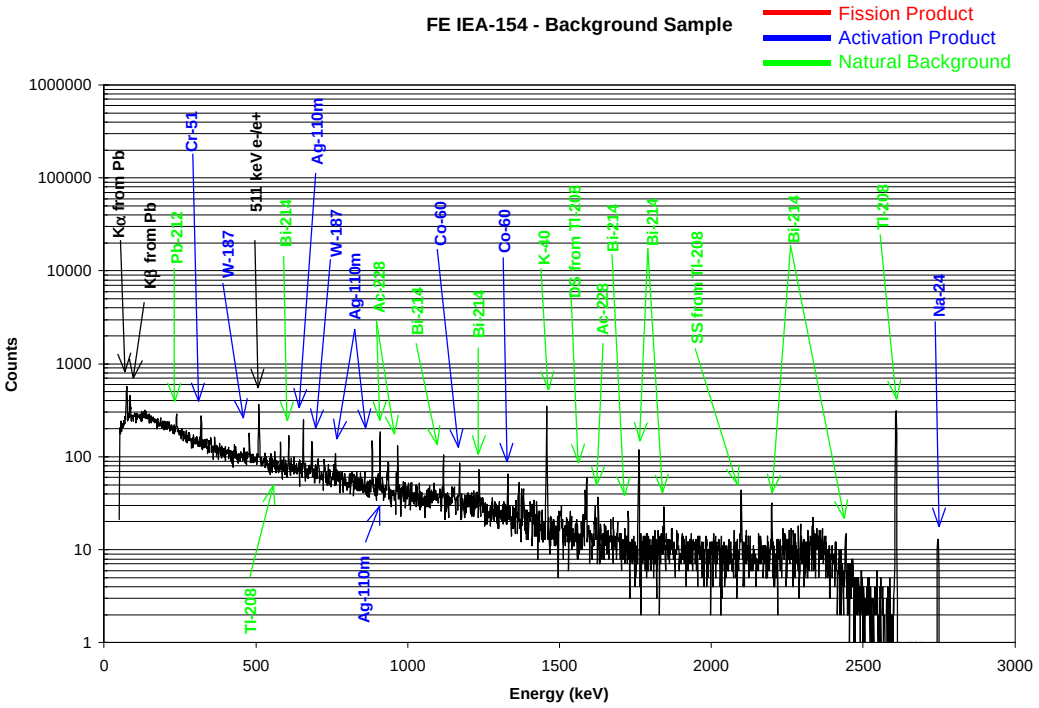

Fig. 1. Gamma-ray spectrum obtained from measurement of 4000 s of live time performed on a background sample of the sipping tube water, collected immediately after the washing of the fuel element IEA-154. The origin of each peak is indicated.

Concerning fuel elements without failure, the total absence of fission products was also the most important characteristic of every gamma-ray spectrum obtained from measurements of sipping water samples collected even after the longest sipping times. A typical gamma-ray spectrum obtained from these

measurements is shown in figure 2. Compared with gamma-ray spectra obtained from background measurements, the presence of activation products is clearly enhanced, specially of ^{24}Na, ^{51}Cr, ^{60}Co, ^{110m}Ag and ^{187}W.

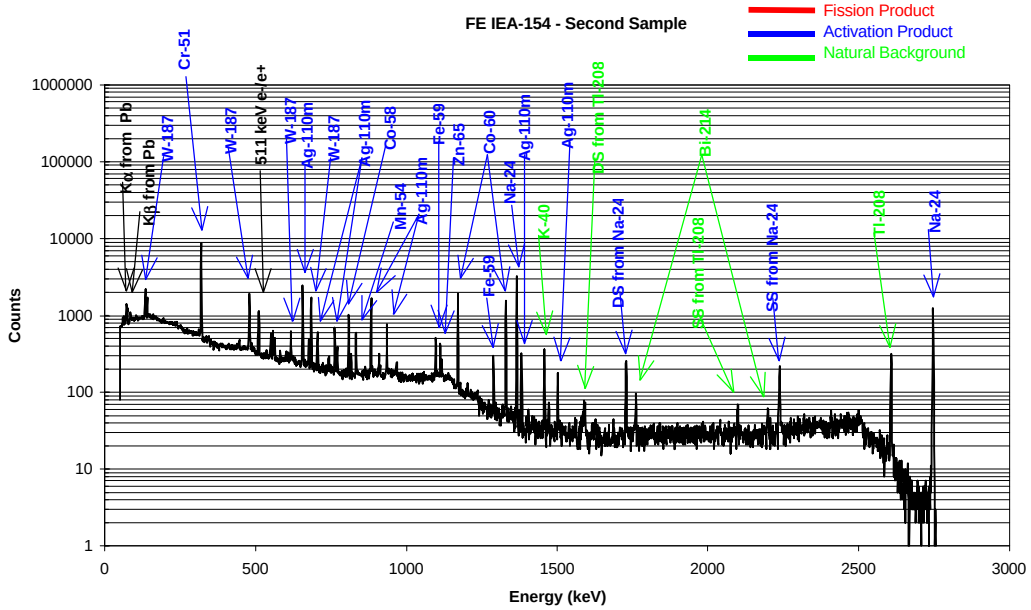

Fig. 2. Gamma-ray spectrum obtained from measurement of 4000 s of live time performed on a sample of the sipping tube water corresponding to the fuel element IEA-154, collected after a sipping time equal to 1106 min. The origin of each peak is indicated.

However, in case of a failed fuel element, the gamma-ray spectra obtained from measurements of sipping water samples show mainly fission products with high solubility in water, as shown in figure 3. As a result, the total activity of the sample is considerably higher. Full-energy peaks corresponding to gamma-rays emitted by radioactive isotopes of iodine are the most prominent in the spectrum. The presence of ^{131}I and ^{133}I is remarkable. This kind of gamma-ray spectrum was obtained only from measurements of sipping water samples corresponding to the fuel element IEA-156, identifying it as failed.

In order to determine the average leaking rate of ^{137}Cs from the failed fuel element to water, the samples corresponding to the fuel element IEA-156 were measured after 6 months, giving gamma-ray spectra like the one shown in figure 4. The only fission products that remain are ^{134}Cs and specially ^{137}Cs.

Once the decay constant of ^{137}Cs is $\lambda = 7.2925.10^{-10}$ s^{-1} and the gamma-ray absolute emission intensity of the 661.6 keV gamma-ray is $I_\gamma = 0.851$ [6], the specific activity A of ^{137}Cs in each measured sample was determined using expression (1), with the results given in Table 1.

Table 1. Net number of counts under the 661.6 keV full-energy peak, specific activity A of ^{137}Cs and sipping time t for each sample corresponding to the failed fuel element IEA-156

Sample	*Area* [counts]	A [Bq/l]	t [min]
First	4353 ± 89	1534 ± 56	251
Second	8543 ± 123	3011 ± 101	1144

Finally, the average leaking rate of ^{137}Cs from the fuel element IEA-156 to water was determined using expression (2) and the data of Table 1, with the result R = (1.65 ± 0.13) Bq/l.min.

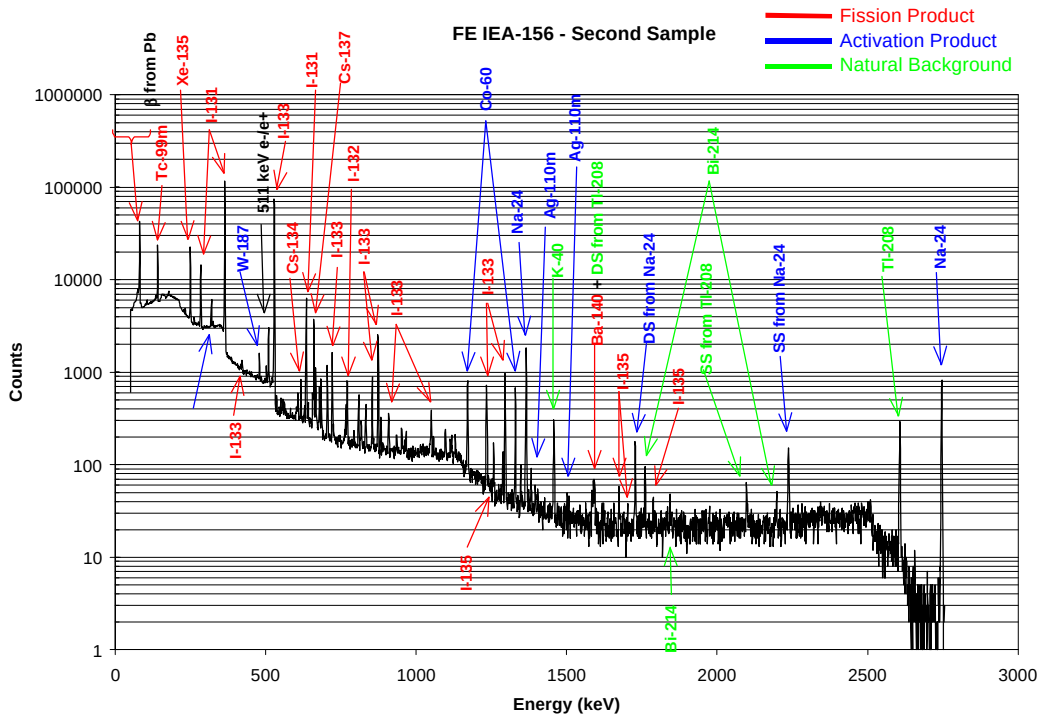

Fig. 3. Gamma-ray spectrum obtained from measurement of 4000 s of live time performed on a sample of the sipping tube water corresponding to the failed fuel element IEA-156, collected after a sipping time equal to 1144 min. The origin of each peak is indicated.

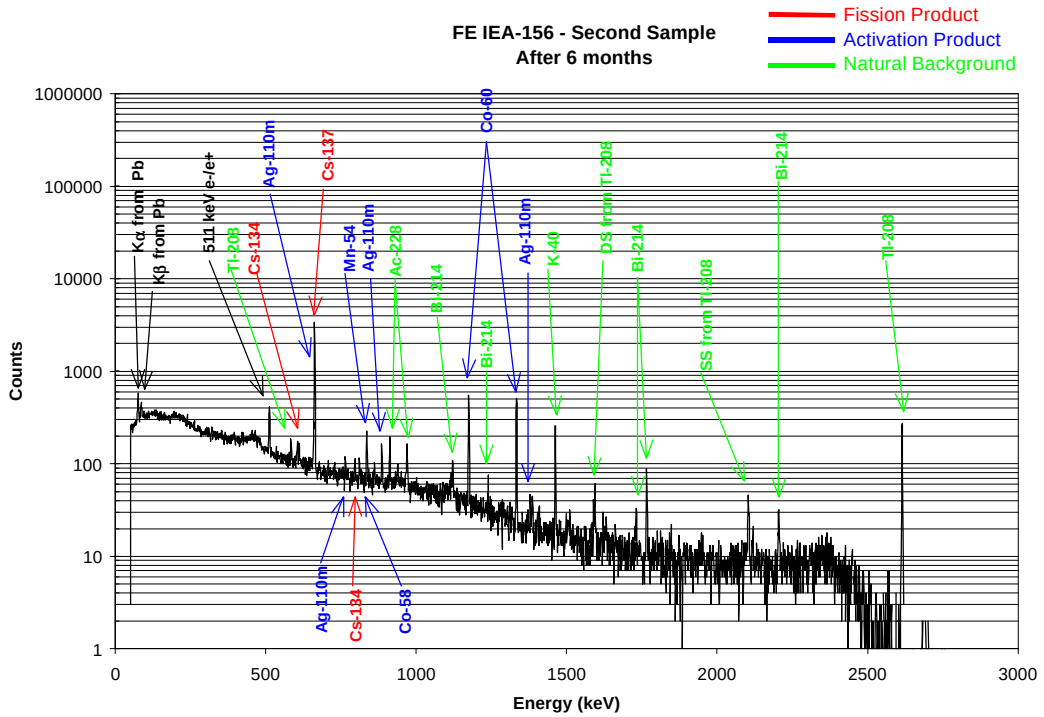

Fig. 4. Gamma-ray spectrum obtained from measurement of 4000 s of live time performed on a sample of the sipping tube water corresponding to the failed fuel element IEA-156, collected after a sipping time equal to 1144 min. This measurement was carried out precisely 6 months after sampling. The origin of each peak is indicated.

4. Discussion

4.1. Choice of failure monitor

For sipping tests on irradiated fuel elements stored for many years inside spent fuel pools, the most suitable fission product for use as failure monitor is ^{137}Cs, due to its long half-life (30.14 years), great fission yields and high solubility in water [1].

However, for sipping tests on newly irradiated fuel elements, as in the case of this work, the choice of a failure monitor is not so obvious. Of course, great fission yields and high solubility in water remain indispensable characteristics of the radionuclide to be used. Nevertheless, in this case the radionuclide half-life must be much shorter, typically about some days, to provide a high specific activity in the sample and an easy identification in the gamma-ray spectrum. The gamma-ray spectra obtained from measurements on samples corresponding to the failed fuel element show clearly that ^{131}I and ^{133}I, radioactive isotopes of iodine with half-lives respectively equal to 8.02 days and 20.8 hours [6], are the suitable failure monitors in these conditions. Although present in these gamma-ray spectra, the full-energy peak of 661.6 keV, emitted in the decay of ^{137}Cs, is not prominent.

An additional evidence for the choice of ^{131}I and ^{133}I as failure monitors regarding sipping tests on newly irradiated fuel elements is that, shortly after unwanted releases of fission products, these two radioiodine isotopes are the most easily detectable radionuclides by means of gamma-ray spectroscopy [7,8].

On the other hand, precise values for specific activity and average leaking rate of ^{131}I and ^{133}I are difficult to obtain, because their most prominent full-energy peaks, corresponding to gamma-rays with energies of 364.5 keV for ^{131}I and 529.9 keV for ^{133}I, are partially overshadowed by Compton continua from many other gamma-rays with higher energy.

Under these circumstances, one decides to wait 6 months to enable the decay of short-lived fission products and activation products in the samples, in order to measure the average leaking rate of ^{137}Cs from the failed fuel element to water.

4.2. Comparison of measured average leaking rates of ^{137}Cs

The nuclear fuel used in the failed fuel element, IEA-156, is U_3O_8 – Al dispersion fuel, with 2.3 gU/cm^3 and 19.88 % enrichment. This fuel element, fabricated at IPEN/CNEN-SP, was positioned in the reactor core on September 8th 1997 and reached a burnup of 21.95 % [9]. It must be emphasized that 5 other fuel elements with identical fabrication characteristics were placed inside the reactor core at the same date and are still operating with no problems at all.

Although it is not possible to evaluate the fuel performance, concerning the release of fission products under cladding failure, only based on the measured value R of the average leaking rate of ^{137}Cs, it can be used for comparison with other measurements of the same kind.

Visual inspections and sipping tests performed on 60 irradiated fuel elements stored for many years inside the spent fuel pool of the IEA-R1 research reactor showed that the fuel element IEA-53 presented many corrosion pits along the height of each external fuel plate and the highest average leaking rate of ^{137}Cs. The nuclear fuel used in the fuel element IEA-53, fabricated by Babcock & Wilcox Co. in the fifties, is U – Al alloy fuel with 1.8 gU/cm^3 and 20 % enrichment. In the date the average leaking rate was measured, the fuel element IEA-53 had a total ^{137}Cs activity equal to $1.406.10^{12}$ Bq. The measured average leaking rate of ^{137}Cs from this fuel element to water resulted r = (0.208 ± 0.017) Bq/*l*.min [1].

Been the total ^{137}Cs activity of the failed fuel element IEA-156, in the date the sipping test begun, equal to $5.350.10^{12}$ Bq, the measured average leaking rate of ^{137}Cs is, proportionally to the total activity, approximately 2.1 times greater than the one obtained for the fuel element IEA-53.

Before any other factor, it is important to remember that the cladding oxidation conditions of these two fuel elements are totally different, once the fuel element IEA-53 remained under wet storage during almost 28 years.

Apart cladding oxidation conditions, it would be interesting to elucidate how two other factors contributed to this difference: a) type of nuclear fuel used (U_3O_8 – Al dispersion in IEA-156 x U – Al alloy in IEA-53); b) characteristics of the failure present in each one of the fuel elements (cause, type, size, depth, location).

However, with the non-destructive techniques available at IPEN/CNEN-SP, it was not possible to get any information about the characteristics of the failure in the fuel element IEA-156. For this kind of research, examinations in hot cells where this fuel element could be dismantled are necessary.

5. Conclusions

Sipping tests were employed successfully at IPEN/CNEN-SP in order to identify cladding failure in fuel elements under use in the core of the IEA-R1 research reactor. These tests not only identified, among 24 fuel elements, which one failed during a routine operation at 2 MW, but also enabled the measurement of the average leaking rate of ^{137}Cs from the failed fuel element to water.

The results of these sipping tests showed that the radioactive isotopes of iodine ^{131}I and ^{133}I are the most suitable failure monitors for newly irradiated fuel elements. The fuel element IEA-156 was readily identified as failed due to the remarkable presence of these fission products in gamma-ray spectra obtained from measurements on its sipping water samples.

In order to measure the average leaking rate of ^{137}Cs from the fuel element IEA-156 to water, its sipping water samples were carefully stored during 6 months and submitted to gamma-ray spectroscopy not till then. The obtained value is, proportionally to the total ^{137}Cs activity, approximately 2.1 times greater than the one obtained for the fuel element IEA-53, which remained under wet storage during almost 28 years and presented many corrosion pits along its external fuel plates.

A more conclusive comparison between the performance of the two types of nuclear fuel (U_3O_8 – Al dispersion in IEA-156 x U – Al alloy in IEA-53), concerning the release of fission products in the condition of cladding failure, requires an investigation of the characteristics of the failure present in each one of the fuel elements. For this kind of research, examinations in hot cells where the fuel element IEA-156 could be dismantled are necessary.

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