

# **SORTING METHODOLOGY TO MANAGE RADIOACTIVE POWDER COMING FROM IRRADIATED FUEL MACHINING**

**(LECA STAR FACILITY CEA CADARACHE CENTRE)**

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## **ABSTRACT**

To meet the R&D needs in terms of microscopy and microanalysis , irradiated fuel samples are cut and polished in dedicated hot cells of the LECA STAR nuclear facility. Samples are encapsulated into an aluminum ring which is filled with an epoxy resin or Wood's alloy before being cut and polished. This process leads to the production of polishing residues containing fuel, aluminum, zircaloy (fuel cladding), resin and/or Wood's alloy. These residues are collected in specific boxes called "caviar boxes" and stored in hot cells.

In the framework of the safety reassessment of the LECA facility, this type of radioactive waste represents a releasable source term which has to be reduced in order to limit the impact of dissemination and internal exposure in case of safety accidental scenarios. The evacuation of these "caviar boxes" is therefore a major issue in terms of waste management.

Intermediate level waste (ILW) produced in the LECA facility are packaged in steel drums (50 liters) in hot cells and then transported to the dedicated waste managing facility INB37, to be compacted with a 500 ton press, packed into a larger containers (500 liters) and injected with concrete. Then, these ILW containers are stored in pits in a waste interim storage facility on CEA Cadarache center, and will be transferred to the future geological repository. To take into account the particular composition of material in these "caviar" boxes, a specific process and method has been developed to match the specifications of the waste management.

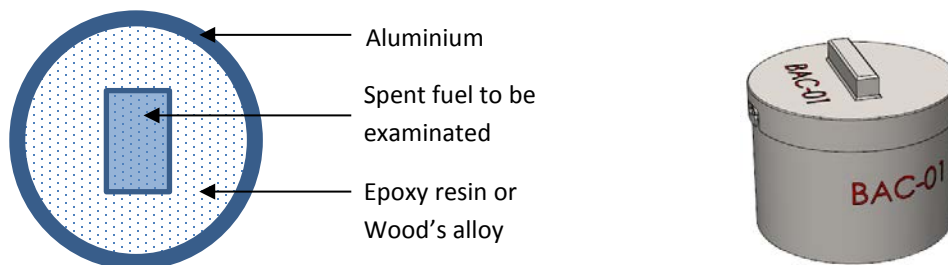
## 1. Introduction

The LECA STAR nuclear facility at the CEA Cadarache centre performs examinations on fuel irradiated in different industrial reactors or research reactors. This facility is dedicated to non-destructive and destructive examinations as to characterise and determine fuel behaviour. For R&D needs in terms of microscopy and microanalysis, irradiated fuel samples are cut and polished in dedicated hot cells at LECA STAR which produced fuel residues. These residues are collected in specific boxes called “caviar container” and stored in hot cells.

In the framework on the LECA safety reassessment, this type of radioactive waste represents a releasable source term which has to be reduced in order to limit the impact of dissemination and internal exposure in case of accident scenarios. Furthermore, as the caviars are stored in the hot cells the experimenters’ workspace is reduced; this lack of free space can impact the R&D programmes and the working conditions of the experimenters. For these reasons, the removal of these residues is a major issue for waste management.

## 2. Main characteristics

For optical examination, samples of spent fuel are embedded in epoxy resin or Wood’s alloy placed in classical aluminium sample holder encapsulated in an aluminum cylinder which is filled with an epoxy resin before being cut and polished. This process leads to producing a mixed powder containing: fuel, aluminum, Zircaloy (from fuel cladding), resin and/or Wood’s alloy. These powders are then collected in stainless steel containers. The volume of each container is about 420 ml.



**Fig. 1: Sample to be assessed and a “caviar container”**

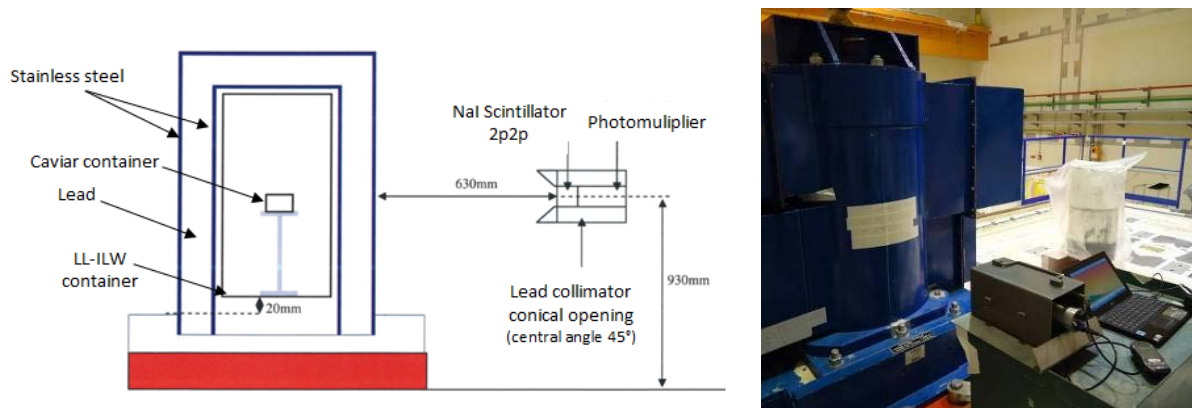
The nuclear material is tracked at every step of the process. The main radioactive characteristics of spent fuel are: U235 + Pu with an isotopic composition of U235 up to 0.71%. Each container holds different amounts of fuel rod residues which make each “caviar” unique.

A series of gamma spectrometry measurements were carried out to assess the amounts and characteristics of gamma-emitting radionuclides contained in these cylinder’s, focussing on contribution of Co-60 to the measured dose rate.

Two types of parameters were identified for the dose rate and transfer function calculation:

- The known parameters, such as the radioisotopes of interest, ILW container dimension, shield lead thickness.
- The unknown parameters such as the composition of the matrix and the location of the source for which assumptions were needed :
  - o The caviar container is at 100% filled,
  - o The matrix is mainly composed of carbon (due to the epoxy resin) with a density of  $1 \text{ g/cm}^3$ ,
  - o The source is uniformly distributed in the matrix.

The materials used are :



**Fig. 2: Gamma spectrometry measurement**

$^{60}\text{Co}$  has never been detected, only  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{154}\text{Eu}$  radionuclides were measured.

### 3. Zircaloy and aluminum issues for waste acceptance

Characteristics of caviar correspond to long-lived intermediate level radioactive waste (LL/ILW). The management of standard LL/ILW produced in the LECA-STAR facility involves a conditioning into double steel containers to meet the acceptance criteria of the receiver INB37 facility. Steel drums received are compacted with a 500 ton press to reduce their size and then packed inside into a 500 liters large containers and finally injected with concrete. Then, these ILW containers are stored in pits on a waste CEDRA interim storage facility located on the CEA Cadarache centre, and will be transferred to the future geological repository CIGEO.

INB37 has defined a quality-assurance reference system supported by specification of waste and packaging (waste acceptance criteria). As radioactive-waste producer is responsible for conditioning waste in the form of packages, he is also responsible for the preparation and submission of application files to be review by the receiver facility.

After a first examination, it appears that the presence of aluminium powders and Zircaloy powders into the composition of these “caviar” could be a problem for this waste acceptance. To fulfill requirements for ILW radioactive packages, it was necessary to justify the absence of ignition’s risk, explosion or exothermic reaction due to the interaction of Zircaloy powders from the “caviar” with the concrete from the packaging process of INB37. Studies realized about the corrosion of steel and

zirconium in aerobic concrete [1] agrees that interaction of Zircaloy with a hydraulic cementation is close to zero: "Speed generalized corrosion varying between 0.02 and 0.1 $\mu$ m per year at pH between 12 and 13.8. Passivation Zircaloy occurred almost instantly after scratching the zirconia layer". Thus, the polishing process guarantees the creation of this Zircaloy oxide very quickly:

- This process involves water, which is then evaporated in a retention tank to collect only dry residues.
- The atmosphere of LECA hot cells is in air; they are not filled with inert gas.

This method makes the oxidation of Zircaloy instantly, so there is no risk of ignition, explosion or exothermic reaction.

Aluminium can react with water cementitious materials in the basic range (pH 13) and lead to a dihydrogen release. This gas release can be a problem while the ILW container is filled with concrete. Difficulties encountered could be:

- " Bubbling " of the concrete which can cause contamination of the package cover and may request a specific exemption for interim storage facility acceptance,
- Explosion due to the dihydrogen release,
- Deterioration of concrete mechanical strength characteristics.

Aluminium surfaces' limits in the package are prescribed by the receiving facility to prevent these risks. The surface area of aluminium must not exceed 0.1m<sup>2</sup> per ILW container to be agreed and derogation is assessed between 0.1 m<sup>2</sup> and 1.5m<sup>2</sup>.



**Fig. 3: Consequences of corrosion by cementation on aluminum massive bloc**

Thus, surface area of aluminium in "caviar" was evaluated:

- 1) Every operation that conducts to produce "caviar" is registered in a dedicated report. Among information's available, we can differentiate cutting tasks from polishing tasks. As samples of spent fuel are cut before being encapsulated in an aluminium cylinder, cutting tasks are not involved in caviar aluminium residues. Only polishing operations can cause aluminium residues in caviar container.
- 2) For R&D needs, different types of polishing setting are processed. Volume of removed material from each type of polishing setting is quantified according to the dimensions of aluminium cylinder, sample heights polished and the grain size from the abrasive used.

Surface area of aluminium is settled for each sample polished and these surface areas are summed to assess the amount of aluminium in each caviar containers. This evaluation shows that 82% of caviar containers meet the aluminium specification (aluminium surface is less than 0.1m<sup>2</sup>), 13% could be accepted by derogation and 5 % cannot be accepted without a specific treatment (not yet determine).

To provide the most guarantee to the receiving facility, a specific packaging process has been carried out involving seven layers between the “caviar” and the outside of the ILW container. Besides the container is fulfilled with some “free aluminium” ILW waste produced in the LECA facility.

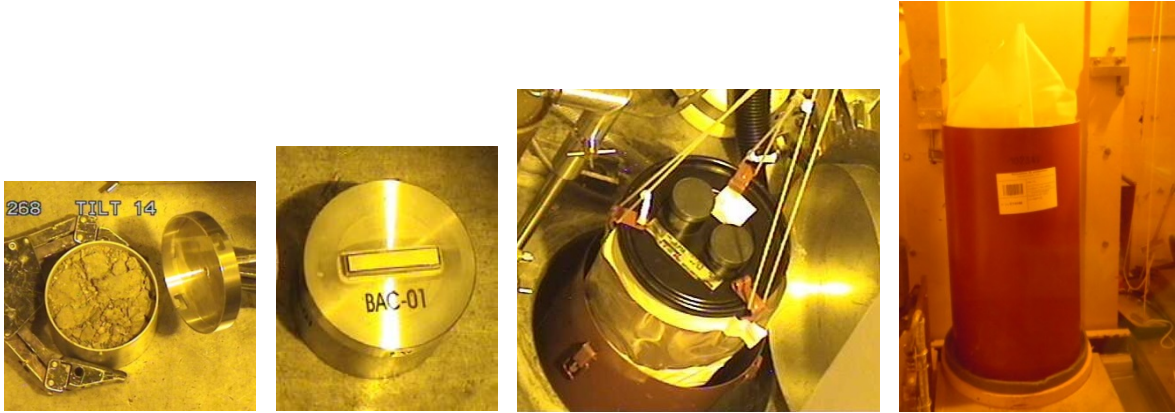


Fig. 4: Specific “caviar” packaging involving several metallic containers, a settling tank canister, plastic covers and ILW container

4. Activity and nuclear mass assessment

Due to, the mix of “caviar” containers and standard waste, the determination of specific activities contained in the package is complex. The method to quantify this activity is based on both measurement of the final container by gamma spectrometry and calculation from the known masses of plutonium and uranium contain in “caviar” with their specific activity. As no “caviar” is identical because each container has a certain amount of various fuel rods residues, nuclear masses materials and activity has been assessed for every “caviar” containers.

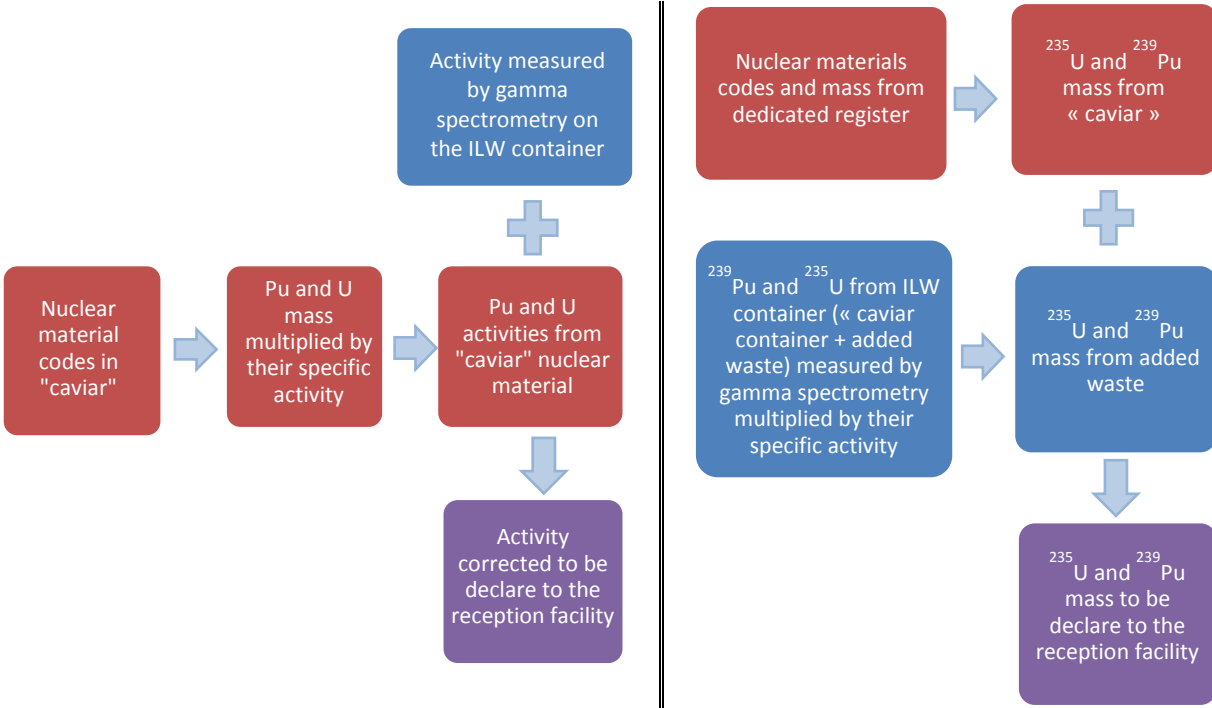


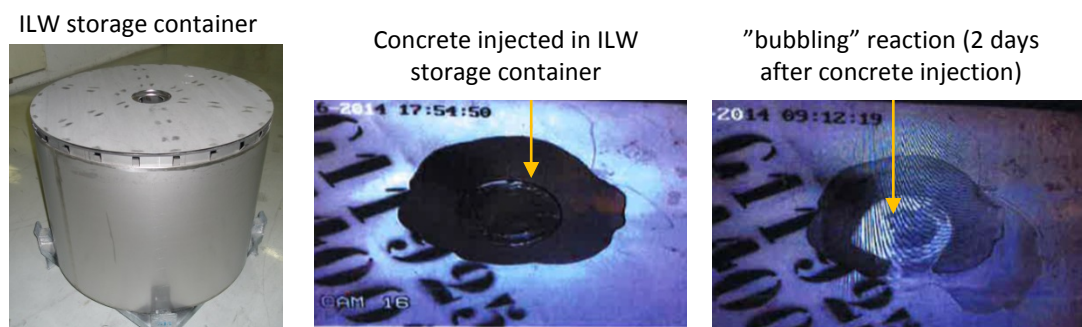
Fig. 5: Activity and fissile mass material evaluation

Gamma spectrometry is carried out on ILW container fulfilled with added wastes. Measured activity of Cs-137 is used to determine the activity of other radionuclides and also total activity of the ILW container by using a known spectrum. The whole measured activity is assumed to come from added waste because “caviar” does not contain enough nuclear material to be registered during gamma spectrometry measurements.

Nuclear material codes are used to describe the content of each object created in “caviar”. Thus, plutonium and uranium isotopes mass are calculated from nuclear materials mass present in “caviar” container. Plutonium and uranium activity is then calculated from the known masses of plutonium and uranium contain in “caviar” with their specific activity.

## 5. Feedback

At the end of the application files review, the waste workshop facility delivered a certificate of acceptance as “caviar” meets the requirements of waste packages and ILW containers. The first caviar removed from LECA-STAR was compacted and injected with some aluminium free ILW container and two days later a “bubbling” reaction was observed. This reaction can come from aluminium residue which was evaluated to 0.931 m<sup>2</sup>.



**Fig. 6: Reaction from the first “caviar” processing**

The storage container was decontaminated and sends to the interim storage facility with a specific exemption. The receiver facility decided to allow only “caviar” container with aluminium surface area under 0.931 m<sup>2</sup>.

Since this decision, six more “caviar” containers were sent to the waste workshop facility and were treating with the same process as the 1<sup>st</sup> “caviar” container without any problem. For those upper from this limit, contents will be divided to meet this specification criteria.

## References

[1] CM HANSSON, The corrosion of steel and zirconium in aerobic concrete, Material Reaserch Society Symposium, Proc. Vol. 50, P 475, 1985