Detection and Analysis of Fuel Cladding Damages Using Gamma Ray Spectroscopy

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Abstract. The cleanliness of the primary circuit is a safety requirement for nuclear power plants particularly in the case of Gen-IV Sodium cooled Fast Reactors. During operation, the concentration in fission products circulating into the coolant needs to be monitored by dedicated radiation monitoring systems. Such systems could be based on neutron counting or gamma spectroscopy. Gamma spectroscopy can reveal itself to be a powerful technique when examining the status and stage of a clad failure. Using the isotopic information provided by the system, a figure of merit is calculated to anticipate fuel failures and mitigate the risk.

This paper deals with the implementation of a gamma ray spectroscopy system at the ISABELLE-1 test loop in the OSIRIS experimental reactor. A complete fuel failure has been monitored, and the recorded data have been processed using an advanced analysis method. The obtained results are presented and discussed.

Key Words: Fuel, Failure, Gamma, Spectroscopy.

1. Introduction

According to nuclear safety requirements, fission products need to be strictly contained into fuel pins. The clad then forms the first containment barrier and its integrity must be paid special attention. Nuclear measurement systems are used to prevent or mitigate the risk of clad failure. As some fission products are delayed neutron emitters, Delayed Neutron Detection (DND) systems are notably implemented. These systems are remotely set thanks to a continuous coolant sampling, in order to avoid interferences with core neutrons. The neutron count rate (typically obtained from ^{3}He or boron-coated proportional counters) is compared to a preset threshold [1]. The simplicity of the signal processing makes the system robust. However, only some volatile fission products are detected (^{87}Br, ^{137}I, ^{88}Br).

Gaseous fission products (Xe and Kr isotopes) are the only ones to be released prior to volatile products during the depressurization of the pin plenum and before coolant contact with fuel. Now, gamma spectroscopy enables gaseous fission product measurement and isotopic separation. This technique implements more complex signal processing but will guarantee earlier detection and provide information about the failure evolution. Three stages can be distinguished a clad failure signal:

1. A burst of noble gases released until the achievement of pressure equality between pin inner pressure and coolant pressure;

2. A release of fission products by diffusion of atoms in the fuel according to thermal gradients [2];
3. A release of fission products released by recoil and knockout when the failure becomes large enough to allow a contact between fuel and coolant.

In the scenario of a small failure, fission products released into the coolant will be a function of their live time. Indeed, higher the relative quantity of long-lived fission products, while short lives radionuclides will decay during the diffusion process. On the contrary, releases by recoil and knockout are independent from time constants.

High resolution gamma spectroscopy is investigated as a technique to continuously estimate the release rate as a function of the time constant of the fission product $f_{\lambda,t}$ in order to detect failure at early stage and to prevent failure aggravation. This paper presents some tests performed at the ISABELLE-1 experimental loop of OSIRIS reactor.

2. The ISABELLE-1 experiment

OSIRIS is a Material Testing Reactor (MTR) installed at the CEA Saclay and operated from 1966 to 2015. The main task of the reactor was to produce a high neutron flux ($3x10^{18}$ $n.m^{-2}.s^{-1}$ thermal; $4.5x10^{18}$ $n.m^{-2}.s^{-1}$ fast) to qualify materials used by electronuclear industry. ISABELLE-1 is a loop allowing the testing of fuel pins. The flux magnitude is set by moving the fuel carrier close or far from the core [3]. The power released by the pin is continuously estimated by thermal balance and SPND measurement, and pin distortion is measured by LVDT [4]. It can be seen in *FIG. 2.* that DND and gamma spectrometry are carried out in the “out-of-pile” part of the system. The high resolution gamma spectroscopy system has been installed in front of this tube to monitor continuously the number of isotopes passing through the sampled coolant.

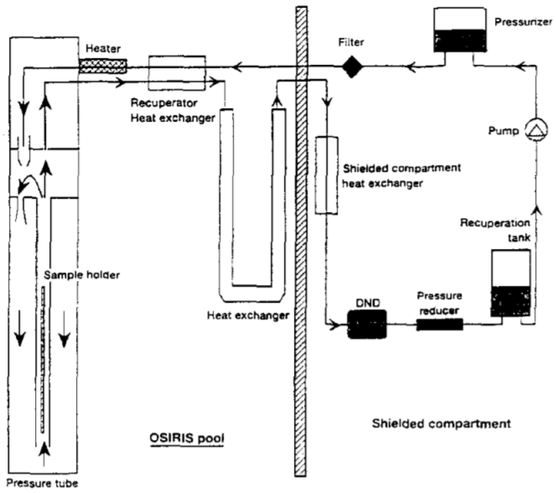
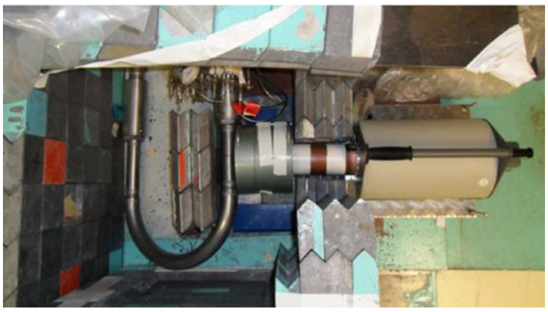


FIG. 2. Schematic of the ISABELLE 1 loop [3].

3. The gamma spectroscopy system

The detector is a Hyper Pure Germanium (HPGe) diode ensuring excellent energy resolution. The Digital Signal Processing unit is the ADONIS system, which is dedicated to process-varying activities [5, 6]. The HPGe diode has been set just behind the pool wall, which corresponds to a transit time from pin to the measurement point equal to 65 seconds. *FIG. 3*. shows the tube and the system composed of the cryostat, the diode and a collimator.



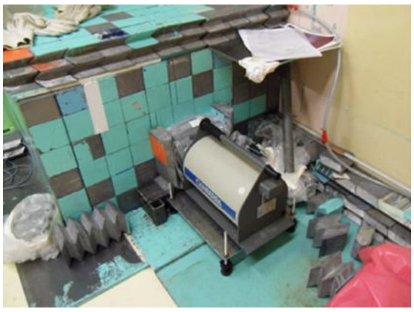
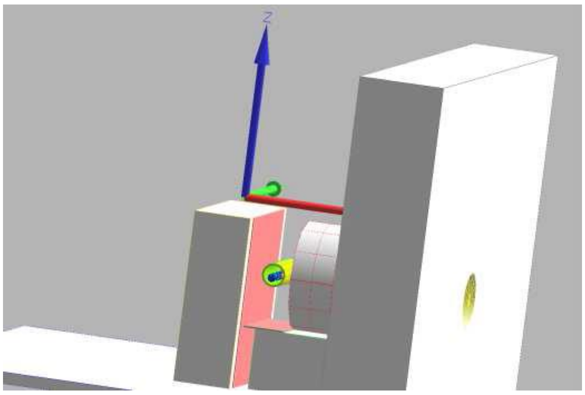


FIG. 3. Photography of the gamma spectroscopy system (left: top view; right: side view).

Firstly, the diode has been characterized using calibrated sources (^{137}Cs, ^{60}Co and ^{133}Ba) in order to build a validated MCNPX model. Then the diode model has been incorporated into the global model of the measurement set-up as illustrated in *FIG. 4*.



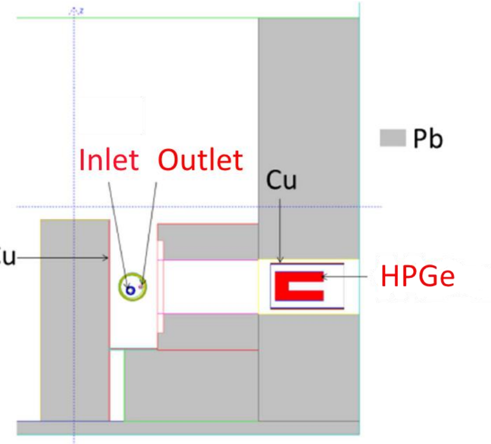


FIG. 4. Geometrical model of the measurement system (left: 3D view; right: cross-section view).

We can see that the signal will be composed of two parts: the signal from the inlet circuit and the signal from the outlet circuit. The total detection efficiency ε is calculated based on the independent efficiencies ε_{in} and ε_{out}, weighted by their associated volumes V_{in} and V_{out}. Detection efficiencies ε_{in} and ε_{out} are estimated using the pulse height tally (f8) of the MCNPX2.7 code [7].

$$\varepsilon = \frac{V_{in}}{V_{in} + V_{out}} \varepsilon_{in} + \frac{V_{out}}{V_{in} + V_{out}} \varepsilon_{out} \quad (1)$$

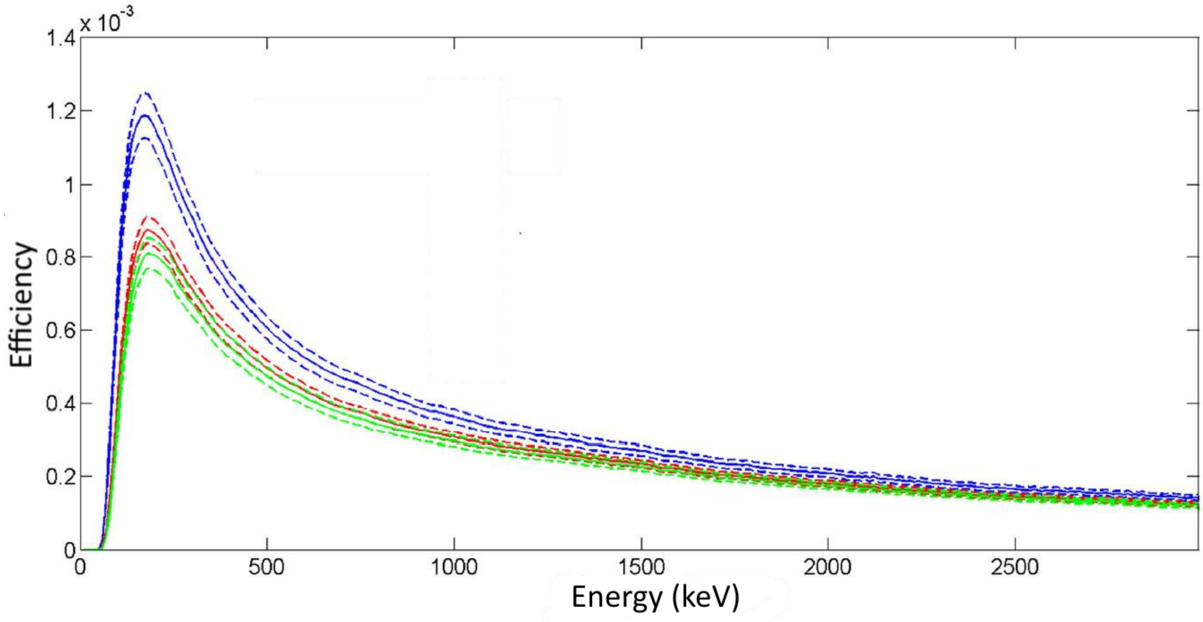


FIG. 5. Detection efficiencies for the inlet circuit (green) and for the outlet circuit (blue). Dash lines are 1σ uncertainties.

Values of the matrix $f_{\lambda,t}$ are estimated by gamma spectrometry using:

- the count rate S_{X,E_γ} obtained by peak deconvolution of a radionuclide X emitting gamma rays at the discrete energy E_γ,
- The detection efficiency ε_{E_γ} for photons emitted at the energy E_γ (from the distributions in *FIG. 5.)*,
- The branching ratio β_{X,E_γ} corresponding to the probability for photons at the energy E_γ to be emitted by a disintegration of the radionuclide X,
- The cumulative fission yields Y_X corresponding to the probability for a fission product X to be produced by a given fissile nucleus (typically ^{235}U),
- The radiative decay constant λ_X of the radionuclide X,
- The transit time τ from pin to the measurement point.

Such as:

$$f_{\lambda,t} = ln\left(\frac{S_{X,\gamma,t}exp(\lambda_X\tau)}{\lambda_X\beta_{X,E_\gamma}Y_X\varepsilon_{E_\gamma}}\right) \quad (2)$$

The figure of merit α_t is defined as the slope of lines given by the functions f_λ (which are affine curves) such as:

$$\alpha_t = \frac{\partial f_{\lambda,t}}{\partial\lambda} \quad (3)$$

An aggravation of the failure will reduce the time between fission products production and release. Thereafter, the concentration of short period radionuclides at the measurement point will increase in comparison with longer period ones, inducing an increase in α_t values.

The count rates $S_{X,\gamma,t}$ are estimated by the processing of ADONIS recorded data (list mode) using the SINBAD code. SINBAD implements a nonparametric Bayesian approach for peak deconvolution [8]. This code is particularly suited for the treatment of highly convolved spectra, as the ones acquired during a clad failure [9]. A typical example of such a spectrum is given in *FIG.6*. Moreover, a temporal smoothing is added using a nonlinear filtering method [10, 11].

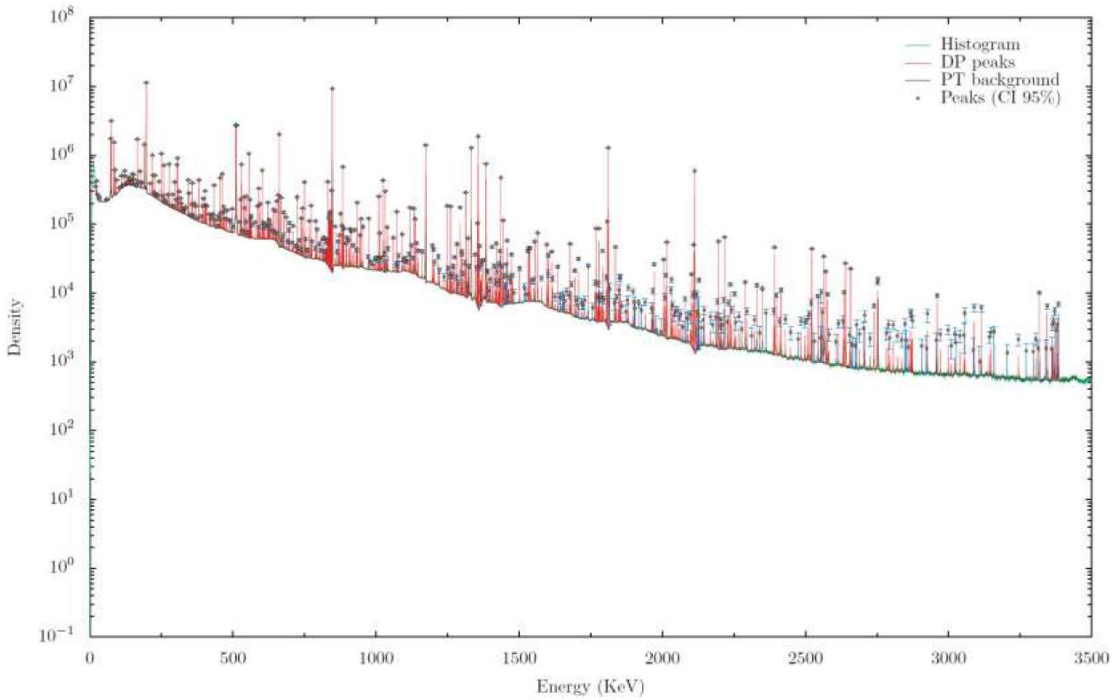


FIG. 6. Spectrum obtained during water coolant measurement after a clad failure.

4. Results

The analysis of a real clad failure is presented and discussed in this section.

a. Total gamma count rate

The total gamma count rate is first analyzed. It is important to notice that ADONIS provides an accurate estimation of the dead time and therefore unbiased total count rate estimation whatever its magnitude is (up to 5 Mcps).

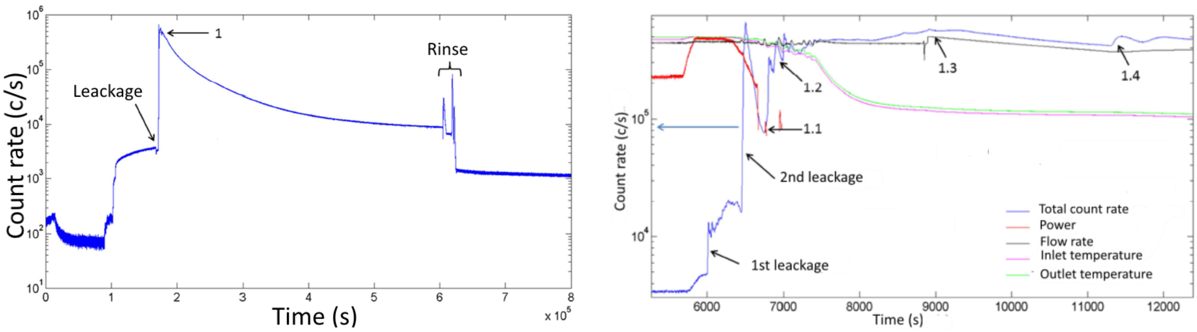


FIG. 6. Total gamma count rate as a function of time during the clad failure. Left: global profile; right: focus on the leakage.

It can be seen in *FIG. 6.*, that the failure has occurred in two steps: a first leakage 160 seconds after the target power step was achieved, with a count rate increasing from 5000 cps to 11000 cps; the second leakage recorded 450 seconds after the first one with a count rate increase up to 657000 cps. The decrease (1.1) is due to the moving back of the loop (i.e. power decrease) and the periodic bonces (1.3) are attributed to the cyclic transit of the coolant.

b. Spectrometry analysis

FIG. 7. shows isotopic signals observed during the clad failure. The first leakage (between 19000 and 22000 seconds) is dominated by noble gas fission products, corresponding to the pin plenum depressurization. The second leakage (between 22000 and 23000 seconds) appears during the recoil of the loop and the power decrease. The isotopic mixture is composed by noble gas and also volatile products, due to a probable increase of the fissure size. This type of release is the sign of water penetration into the pin and can be explained by a temperature decrease, inducing an enlargement of clad-pellet intervals.

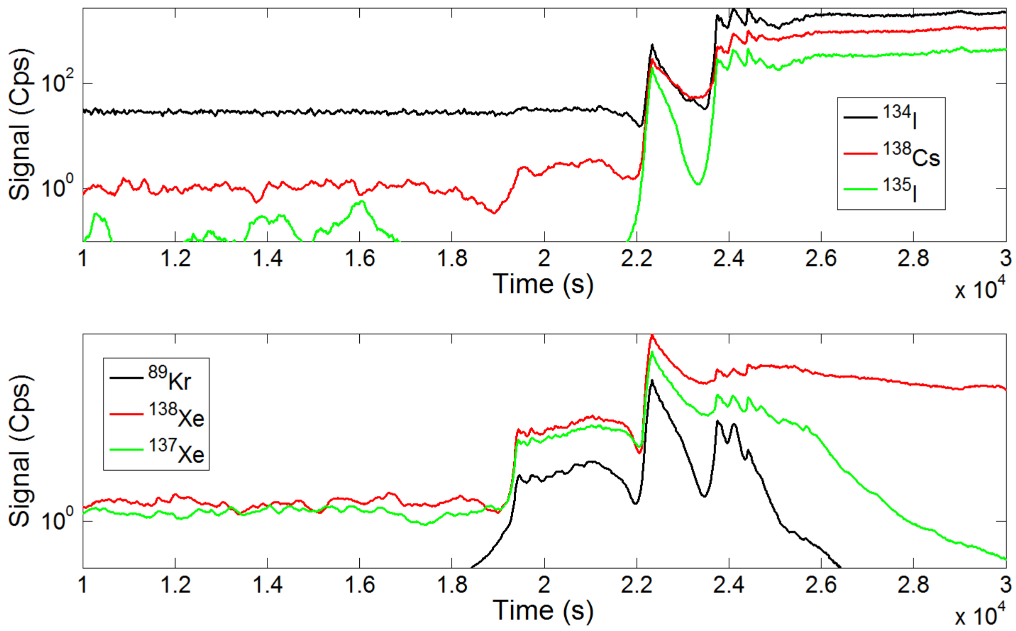


FIG. 7. Count rates by fission product as a function of time.

The evolution of the figure of merit $\alpha(t)$ as a function of time is presented in *FIG. 8.*, for noble gas and volatile fission products. It can be observed that the parameter α associated with noble gases starts to increase (from -2 to -0.5) one-hour-and-a-half before the first burst of signal observed at 19000 s. This result shows the added-value of monitoring α in order to anticipate clad failure aggravation. Similarly, a brutal increase of the parameter α associated with volatile fission products is observed for the second leakage at 22000 seconds with an increase from -0.7 to -0.1. This flatness of the profile is the sign of a release dominated by recoil and knockout phenomena, induced by a direct contact between fuel and coolant.

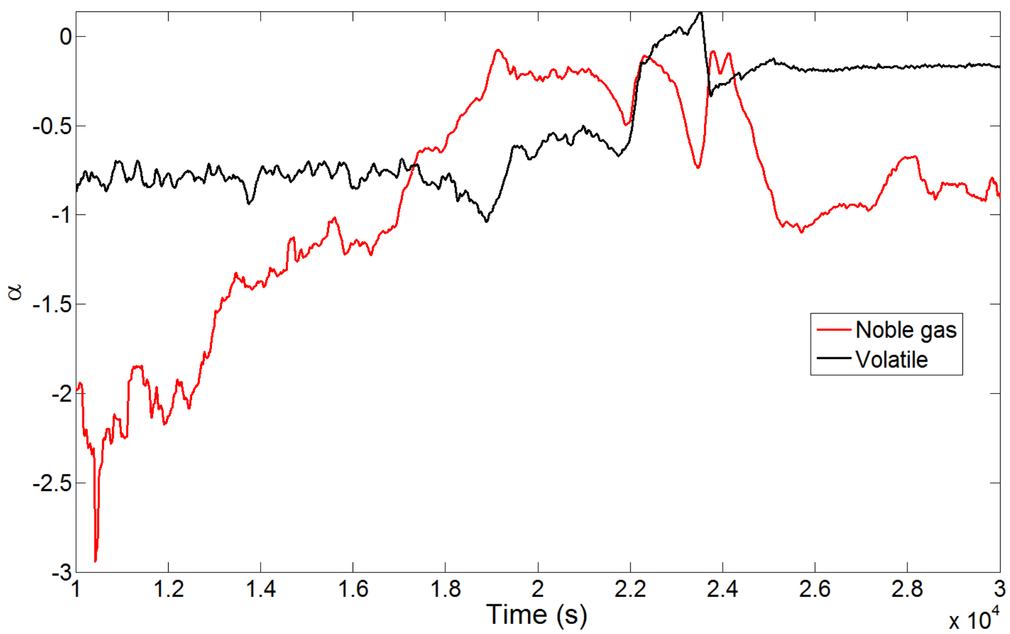


FIG. 8. Count rates from fission products as a function of the time.

5. Conclusion

A high resolution gamma spectrometry system has been installed in the ISABELLE-1 experiment during a fuel pin test. During this test a clad failure occurred and the spectrometric signal was recorded. The signal has been processed along both temporal and energetic dimensions to build a figure of merit dedicated to clad failure anticipation.

It has been proven the relevancy of this figure of merit to detect early failure aggravation has been established. This result opens perspectives in NPP's safety, with the implementation of on-line and *in-situ* gamma spectroscopy system, monitoring the primary coolant. This approach is currently under development in the framework of Gen-IV Sodium-cooled Fast Reactors.

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