FISSION GAS RELEASE UNDER NORMAL AND OFF-NORMAL CONDITIONS : NEW ANALYTICAL DEVICE IMPLEMENTED AT THE CEA-CADARACHE

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ABSTRACT

Fission gas release is generally considered to be a key phenomenon that must be assessed for fuel rod design and licensing under normal and off-normal operating conditions. Under normal conditions, FGR is a potential limiting design factor due to its impact on the fuel rod internal pressure, with the associated risk of fuel thermal degradation. Under off-normal conditions, FGR is an important input data as regard of the radioactive "source term" relatively to the consequences of a nuclear accident on the environment, and predicting correctly its release in these conditions (i.e. RIA, power transients, LOCA, …) remains an important R&D's goal.

As a consequence, the CEA has been involved in a number of research programs devoted to fuel behaviour. From a general point of view, whatever the exact nature of the programs, the efficiency of the experiments needs to be improved in order to, more specifically, allow a better understanding of the basic mechanisms involved in global fuel behaviour. This can be achieved by developing new experimental techniques. That is why the Fuel Studies Department (DEC), at the Cadarache CEA centre, operates a specific annealing facility called "MERARG" in a shielded cell of the LECA-STAR hot laboratory. This induction furnace is operating with irradiated fuel (a few hundred mg to a few g) under inert (or oxidizing) atmosphere at temperature levels ranging from room temperature up to about 2800°C. Qualification tests demonstrated the possibility of performing tests with temperature ramps between 0.1 and 200°C/s. The specimen is swept by a regular gas flow which is analysed by gamma spectrometry.

The aim of this paper is to review the main aspects of these specific experimental activities.

KEYWORDS Fission gas release, gamma spectrometry, annealing facility

1. INTRODUCTION

Fission gas release (FGR) is an important input data of the nuclear fuel licensing process, either in normal operation or as the radioactive "source term", relatively to the consequences of a nuclear incident on the surrounding populations as well as on the environment. For this purpose, several R&D programs have been recently initiated in France through joint actions between the Commissariat à l'Energie Atomique (CEA) and Electricité de France (EDF), including occasionally a support from Framatome-ANP or the Nuclear Radioprotection and Safety Institute (IRSN).

Within this framework, a specific emphasis has been put recently on mechanisms which promote the FGR under normal and off-normal operating conditions. One of the most useful ways for understanding the FGR mechanisms is to achieve reliable tests to measure the absolute level and the time dependence of the released gases and the corresponding fuel micro-structural changes during representative thermal transients. This database is used to: (i) define the fission product (FP) "source term" out of the damaged rods during a given type of accidental sequence, (ii) verify if existing safety criteria are well adapted to new fuels, (iii) enhance models predicting behaviour of various fuels during an accidental sequence, thanks to the understanding and quantifying of the basis mechanisms. That is why the Fuel Studies Department (DEC), at the Cadarache CEA centre, within the framework of grouping activities into « poles » of the Directorate of Nuclear Energy (DEN) built the so-called MERARG facility.

The main goal of this paper is to review the general aspects of this specific experimental device implemented at the LECA-STAR laboratory at the Cadarache CEA centre and devoted to the studies of FGR out of irradiated fuels. After a general description of the facility, the global qualification and calibration phases are presented.

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2. GENERAL DESCRIPTION

As explained above, in order to perform experimental programs on FGR at the CEA/CADACHE, the Pre-MESANGE program consisted in implementing in the LECA-STAR laboratory a high frequency induction furnace and its associated equipment (Figure 1) whose aim is to bring an irradiated fuel sample up to temperature levels allowing the extraction of all or part of the gaseous inventory it contains. The released gases are retrieved in a tight circuit swept by a neutral (helium or argon) or oxidizing (dry air) gas with low flow rates (roughly 60 cm^3/min) and then transported to the experimental circuits at the back zone where active gases (gamma emitters) are online analysed by means of a gamma spectrometry station located in the back zone on the outlet line of the furnace. Finally, all the gases are accumulated in two capacities located in a gloves box in the back zone. A sampling circuit allows to take samples of gas which will be analysed later (gamma spectrometry and a Gas-Chromatography station coupled to a Mass Spectrometer – GC-MS).

The whole facility is today called MERARG, a French acronym for “Means of Study by Annealing and Analysis of Gaseous Releases”. MERARG does not include the device for the post-test analyses of gases. In the following section, the main components of MERARG (induction furnace, online gamma spectrometry device, gloves box and the global control command) are described successively in more details.

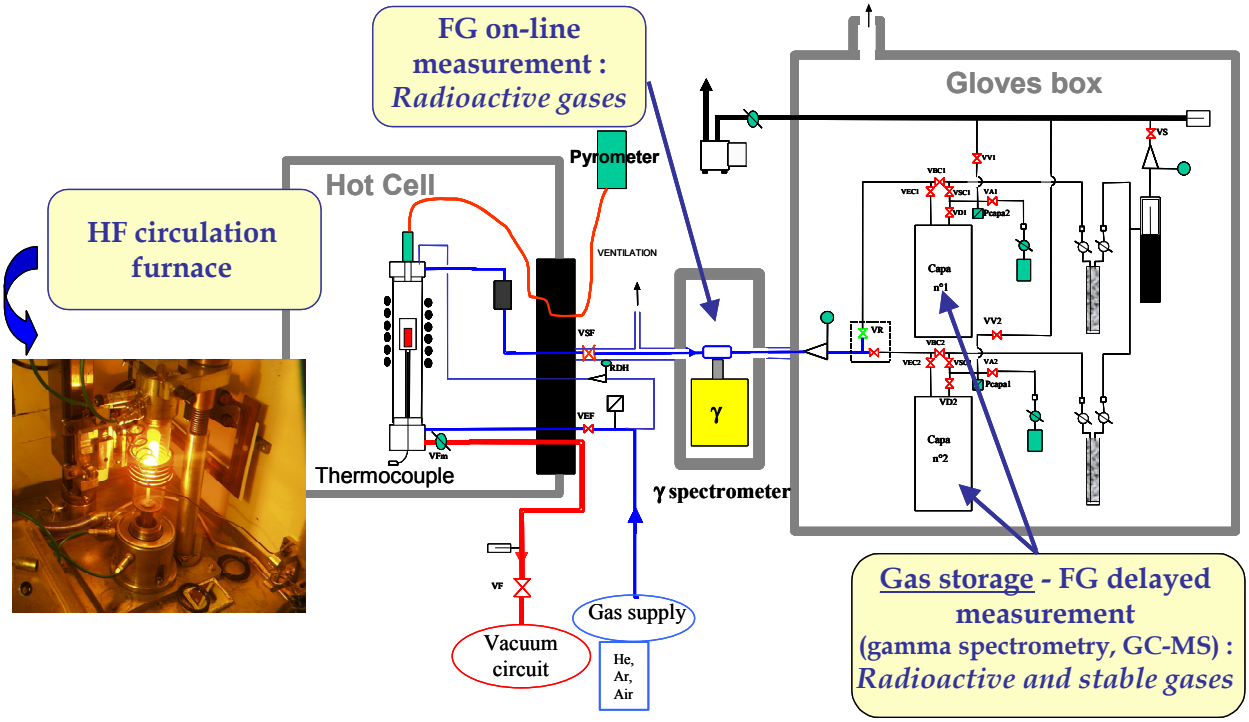


Figure 1. Simplified diagram of MERARG facility

The induction furnace is shown in Figure 2a. It includes a metallic crucible, which acts as the susceptor, heated by induction thanks to 6 coils. Tungsten, molybdenum or platinum crucibles are available depending on the type of thermal treatments needed. The metallic crucible located in the centre of the induction coil consists of two dissociated chambers (Figure 2b): an upper chamber containing the fuel pellet to be characterized (generally one pellet with its clad), covered by a removable lid with a central hole, and a lower chamber in which the instrumentation is found (i.e. thermocouple). The pellet temperature is monitored both by a pyrometer which sights on the top of the pellet through the lid hole and by a thermocouple located in the lower chamber of the crucible. The tight furnace containment is made of a quartz tube in which the crucible is placed. The induction coil, the lower and upper bases of the furnace are cooled by water. The entry of the circulation gases is on the lower base of the furnace, the gas outlet is on the upper base.

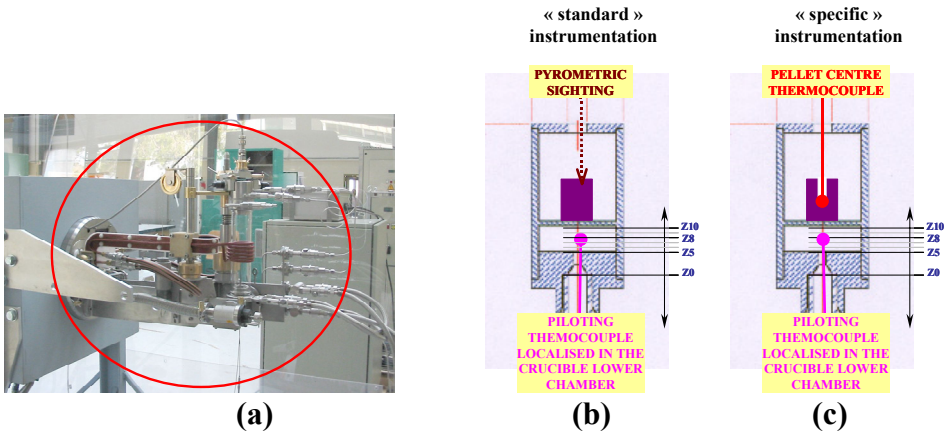


Figure 2. (a) Photography of the induction furnace, (b) scheme of the crucible with its standard instrumentation, (c) scheme of the crucible with its specific instrumentation

During its standard use (i.e. inside the hot cell), the MERARG induction furnace is able to operate in three different configurations in order to perform different types of thermal sequences: (i) The "**Molybdenum**" configuration is characterized by the use of a molybdenum crucible which could perform thermal transients at temperature levels up to 2200°C, a W/W-Re thermocouple or "C type" thermocouple which equips the lower chamber of the crucible, an inert atmosphere (helium or Argon regular flow in the circuits and in the furnace), (ii) The "**Platinum**" configuration is characterized by the use of a platinum crucible which could perform thermal transients at temperature levels up to 1400°C, a Pt/Pt-Rh thermocouple or "B type" thermocouple which equips the lower chamber of the crucible, an oxidant or inert atmosphere (air, helium or Argon regular flow in the circuits and in the furnace), (iii) The "**Tungsten**" configuration is characterized by the use of a tungsten crucible which could perform thermal transients at temperature levels up to 2800°C, a W/W-Re thermocouple or "C type" thermocouple which equips the lower part of the crucible, an inert atmosphere (helium or argon regular flow in the circuits and in the furnace)

The gamma spectrometry station monitors the fission gas release kinetics during the experimental sequence. The measurement device (counting capacity and detector) were designed to be able to quantify less than 1% of the release of the ^{85}Kr contained in a sample of 200 mg of UO_2 fuel at 50GWj/t_m over one hour. The gamma spectrometry device shown in Figure 3 consists of:

- **Within the protection shield** (i.e lead envelope of 10 cm in thickness equipped with a tight opening allowing access to the detector. This shield has a double protection function for the experimenters versus the radiation of radioactive gases circulating and for the detector versus the ambient gamma radiations):
  - ⇨ A stainless steel counting capacity, a simple geometry volume adapted to online gamma measurement,
  - ⇨ The high purity coaxial Germanium detector with 60% of relative efficiency placed as close as possible to the counting capacity.

- **Outside the protection shield**:
  - ⇨ A table used as a support to the entire gamma spectrometry device,
  - ⇨ The cooling system by compression, of type Cryoelectric II.

The counting losses correction is best up to 20000 pulses/s and remains acceptable up to 90000 pulses/s since the losses are less than 2%. The resolution of the gamma spectrometry chain meets the values expected for this type of detector (1.7 keV for the ray from ^{85}Kr).

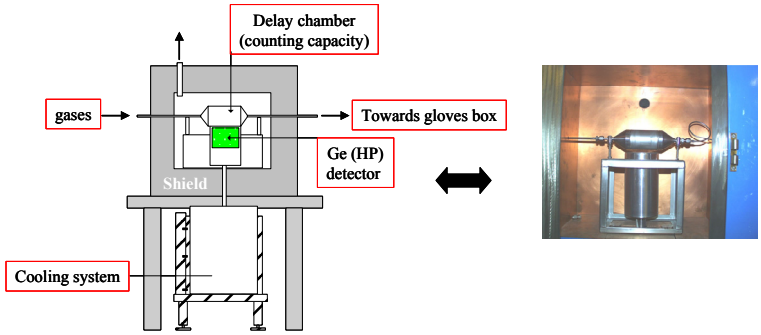
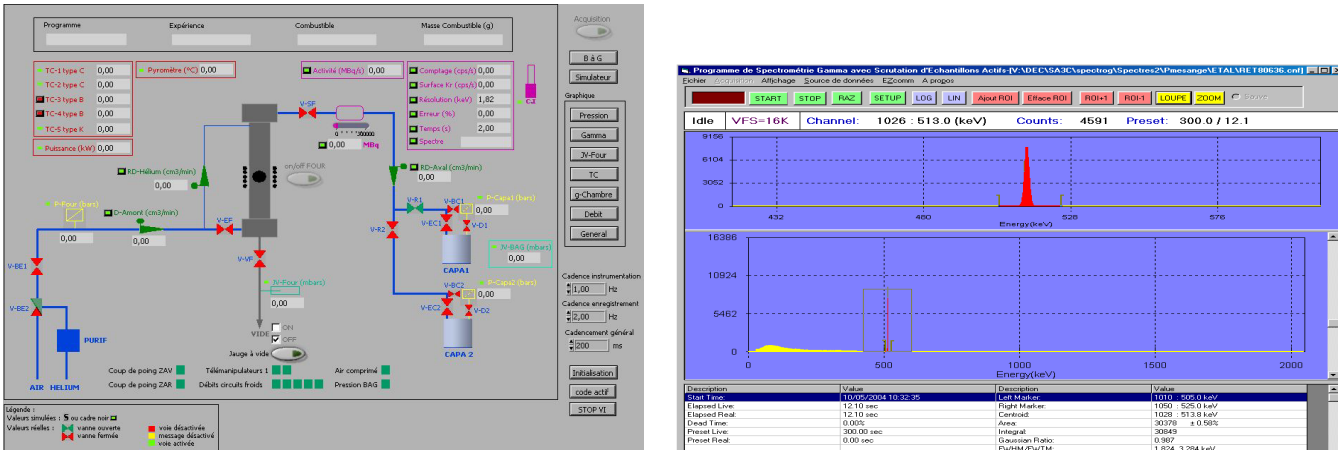


Figure 3. Online gamma spectrometry device, (a) schematic diagram and (b) photography

The gloves box (Figure 1) mainly includes: (i) a flow rate regulator at the glove box inlet (located between the gamma spectrometry capacity and those of the gas sampling) ensuring the required gas circulation, (ii) two storage capacities (chosen among 25, 12 and 6 liters depending on the test requirements) in which the gases accumulate throughout the experiment, (iii) two sampling sites located on the two connected capacities. At the end of the experiment, gas samplings are made in the glove box for post-test analyses, (iv) two gas cold traps and (v) an ionisation chamber used to control online the removal of Krypton 85.

Two systems ensure the global **control command of MERARG**: CAMARG which manages the acquisition of most of the sensors located in the facility and PEGASE2 which manages the data acquisition of online gamma spectrometry.
The CAMARG measurement and command station was developed so as to pilot experiments from an industrial PC. It drives all the control command functions and acquires the data from the facility except for the flow meters, which were not connected, to the application. A DIGISOFT program delivered by QUALIFLOW (supplier of flow rate meters) commands and acquires the data from these apparatus. The main synoptic is shown in Figure 4a. CAMARG consists of a software part developed under LabVIEW (graphic program language) and a hardware part including a PXI (upgraded industrial PC) equipped with a RS 485 communication card and FieldPoint modules (inlet/outlet of digital and analogical signals) connected to the detectors and to the regulation elements.



(a) (b)

Figure 4. (a) main synoptic of CAMARG, (b) main synoptic of MERARG

PEGASE 2 pilots the acquisition of the gamma spectrometry station. This program developed in Visual Basic 6.0 Professional Edition of Microsoft requires the object libraries of ACTIVE X S560 of the program Genie2000 from Canberra Eurisys Mesures as well as acquisition modules S500 and S502. The visualization of a ^{85}Kr spectrum on PEGASE2 is shown in Figure 4b.

3. MERARG FACILITY QUALIFICATION

The qualification of the MERARG facility was performed in two main phases :

- The “cold qualification phase” which took place from June to November 2003 with most of the main components of the facility gathered in a specific test hall, called “cold test” hall. This phase consisted principally in characterising the thermal behaviour and the annealing capabilities of each or the three possible configurations of the MERARG induction furnace.
- The “hot qualification phase” which took place in May 2004 with the MERARG facility fully operational in the LECA STAR hot laboratory. This phase was principally devoted to check and operate the facility, to qualify the on-line gamma spectrometry and to perform thermal treatments that had already been performed in 2002 in a similar facility located in the LAMA hot laboratory.

During the “**cold qualification phase**”, the MERARG facility characterization was principally devoted to understand the thermal behaviour of the crucible and especially the fuel sample according to the piloting process of the HF generator (regulation device programming). In order to do that, one used an alumina pellet whose dimensions were those of a standard PWR fuel rod pellet and two ways of instrumentation for the furnace crucible. The first one is the “standard instrumentation”, presented in the first paragraph : this instrumentation is the one used when operating the furnace in the LECA-STAR shielded cell. The second one is a “specific instrumentation” used only during the cold qualification phase. This “specific instrumentation” consists in equipping the alumina pellet centre with a thermocouple which then replaces the pyrometric sighting (Figure 2c). About 140 tests, were performed during the cold qualification phase: 70 identical tests in both cases of instrumentation and for the three MERARG configurations. In each test, one especially followed: (i) the surface pellet temperature T_{SP} (standard instrumentation case), (ii) centre pellet temperature T_{CP} (specific instrumentation case) and (iii) crucible lower chamber temperature T_{CC} (both standard and specific instrumentation cases). The analysis of the results of those tests consisted in combining the T_{CC}, T_{CP} and T_{SP} measurements for each thermal sequence programmed on the HF generator regulation device.
First, the axial temperature profile was determined for each type of crucible (Mo, Pt and W) and enabled the experimental team to fix for the three crucibles the best axial localisation for the piloting thermocouple in order to minimise the uncertainties on its measurement and to avoid risks of failure (2200°C for C type thermocouples and 1500°C for B type thermocouples) whatever the thermal sequence is and up to the maximal temperature which could be reached by the crucible (2200°C for Mo, 1400°C for Pt and 2800°C for W). Those axial levels were Z8 (8 mm from the top of the crucible support tube corresponding to the centre of the lower chamber) for Mo and Pt crucibles and Z0.5 (0.5 mm from the top of the crucible support tube) for the W crucible.
Then, for this specific position of the piloting thermocouple, the analysis of the 140 tests provided the efficiency of each configuration of MERARG annealing furnace in : (i) the steady-state operating conditions, (ii) the controlled transient operating conditions in which thermal sequences, with ramps between 0.1 to 50°C/s followed by controlled temperature plateaus, are performed by operating the HF generator power regulation device and (iii) the free transient operating conditions. These conditions are obtained without operating the HF generator power regulation device (the HF generator works at a specific programmed power during an appropriated time). it lead to ramps between 50 to almost 200°C/s for the maximal HF generator power ; those ramps can’t be followed by any controlled temperature plateau.

During the “**hot qualification phase**”, the MERARG facility characterization was, after having checked all its components, principally devoted to : (i) qualify the on-line gamma spectrometry and (ii) reproduce thermal treatments that had been already performed in 2002 in a similar facility in the LAMA hot laboratory on a well known and characterized fuel.

As far as the online gamma spectrometry qualification is concerned, a calibration of the facility with certified standard sources is performed in order to establish the relation between the number of pulses counted by the measurement system and the corresponding number of radionuclide present in the counting capacity during the acquisition and that at a given energy. The studies conducted in MERARG only being focused on non-re-irradiated fuel, the efficiency of the facility was determined for the emission ray of ^{85}Kr at 514 keV. The calibration is done based on standard sample made of ^{85}Kr glass seal weld. The sample is placed in a tight sheath located on the gas line upstream from the thermal treatment furnace. A circulation under pure helium in the device is then performed. The flow rate of the loop is set at 60 cm^3/min via the flow rate regulator. The loop is in equilibrium when the furnace pressure and the upstream flow rate meter no longer fluctuate. At that time, the standard sample is “broken”. The active gas is then swept away by the vector gas in the furnace circuit up to the retrieval glove box. The online activity of ^{85}Kr5 is monitored by using gamma spectrometry on the counting capacity. When the activity recorded becomes nil, the calibration is finished and the gas circulation stopped. Three calibrations

were made. Table 1 summarizes the efficiencies obtained after the calibrations. It also emphasizes the very good reproducibility of the experiments.

Table 1. Results of the online gamma spectrometry calibration

Test	Start TT	Number of spectra	Average flow rate (cc/s)	Average P (bar)	Theoretical Kr85 (at/cc)	Yield (shots/gamma)
1	27/04/2004 09:46	120	1.000	1.312	1.85E16	2.052E-02
2	04/05/2004 10:25	97	1.021	1.208	1.85E16	2.079E-02
3	10/05/2004 09:46	113	1.000	1.210	1.85E16	2.067E-02
					η (514)	**2.0661E-02**

Then, the so-called adjustment test was performed. Its objective is mainly to compare the results in terms of fission gas fraction released, obtained on MERARG and on the LAMA thermal treatment furnace, the "ancestor" of MERARG, on a reference test so as to qualify the entire experimental process, the LAMA furnace having largely been validated in the past. The choice of the thermal sequence was made in order to provide a good parametric approach of the phenomena involved. The choice of the sample was made based both on the fuel characterizations available and the availability of samples identical to those of the LAMA. All these considerations resulted in tests on 6 cycles PWR UO_2 fuels. The thermal hydraulic conditions of the tests made as well as the corresponding fractions released are provided in Table 2. This results emphasise the very good reproducibility of the two experiments and as a consequence qualify the facility.

Table 2. Thermalhydraulic conditions of adjustment tests and corresponding results.

Laboratory	LECA	LAMA
Pressure (average over the experiment, bar)	1.186	1.230
Flow rate (average over the experiment, cc/s)	1.001	1.020
Oxide mass (g)	6.522	6.640
History of applied temperature (T° pellet core). Thermalization : Ramp : Threshold : Length of threshold :	 300°C, 30 min 0.19°C/s 1200°C 15 min	 300°C, 30 min 0.23°C/s 1213°C 15 min
Fraction released (%)	17.43	17.04

4. CONCLUSION

The MERARG facility allows **accurate fission gas release measurements** thanks to on-line gamma spectrometry with different crucible configurations:

- The Mo configuration (inert atmosphere) allows to perform accurate ramps between 0.1 to 50°C/s followed or not by a temperature plateau and that, up to 2200°C (main part of the MERARG experiments).
- The Pt configuration (oxidant or inert atmosphere) allows to perform ramps between 0.1 to 7°C/s followed or not by a temperature plateau and that, up to 1400°C. This configuration is generally used for air ingress type experiments.
- The W configuration (inert atmosphere) allows to perform less precise ramps between 0.2 and 20°C/s followed by a temperature plateau up to 2800°C or to perform very fast ramp around 200°C/s up to about 1300°C. This configuration is generally used for complete gas extraction thermal sequences which requires high temperature or RIA type treatments.