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# On-line Fission Products measurements during a PWR severe accident: the French DECA-PF project

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*Abstract* – Following the Fukushima accident, a lot of recommendations was drawn by international organizations (IAEA, OECD, NUGENIA network ...) in order to improve the safety in such accidental conditions and mitigate their consequences. One of these recommendations was to improve the robustness of the instrumentation, which was dramatically lacking at Fukushima, as well as to better determine the Source Term involved in nuclear accident.

The DECA-PF project (Diagnosis of a degraded reactor core through Fission Product measurements) was elaborated in this context and selected as one of 21 collaborative R&D projects in the field of nuclear safety and radioprotection, funded in May 2013 by the French National Research Agency.

Over the months following the Fukushima accident, a CEA crisis team was held in order to analyze on-line the situation taking into account the data delivered by TEPCO and other organizations. Despite the difficulties encountered concerning the reliability of these data, the work performed showed the high capacity of Fission Products (FP) measurements to get a diagnosis relative to the status of the reactors and the spent fuel pools (SFP). Based on these FP measurements, it was possible to conclude that the main origin of the releases was coming from the cores and not from the SFP, in particular for SFP-4 which was of high concern, and that the degradation level of the reactors was very large, including probably an extensive core melting. To improve the reliability of this kind of diagnosis, the necessity to get such measurements as soon as possible after the accident and as near as possible from the reactor was stressed.

In this way the present DECA-PF project intends to develop a new and innovative instrumentation taking into account the design of the French nuclear power plants on which sand bed filters have been implemented for severe accident management. Three complementary techniques, devoted to measure the FP release on-line, are being studied:

- Gamma spectrometry, with an industrial objective to build a prototype aimed at improving the capacity of the present radiation monitoring system,
- Gas chromatography, for the quantification of the fission gases (Xe, Kr) as well as potential carbon oxides produced in case of Molten Corium Concrete Interaction,
- Optical absorption spectroscopy, the objective of this most innovative technique being to quantify the tetra-oxide of

ruthenium, which could be produced in case of lower head failure, and the gaseous forms of iodine (molecular and organic) released in the environment.

A global description and the present status of this project is presented, focusing on the Source Term establishment at the outlet stack of the sand bed filters and on the perspectives of implementation of the on-line gamma spectrometry equipment.

## I. INTRODUCTION

Following the Fukushima accident, a lot of recommendations was drawn by international organizations (IAEA, OECD, NUGENIA network ...) in order to improve the safety in such accidental conditions and mitigate their consequences. One of these recommendations was to improve the instrumentation, which was dramatically lacking at Fukushima, as well as to better determine the Source Term involved in nuclear accident. The DECA-PF project (Diagnosis of a degraded reactor core through Fission Product measurements) was elaborated in this context, based on the experience gained by the CEA crisis team over the months following this accident [1] and taking into account the present equipment and guidelines implemented on the French PWRs [2]. Indeed, during the Fukushima accident, the analysis of the radiation measurements delivered by TEPCO and other international organizations have shown that, despite the difficulties encountered concerning the consistency of these data, Fission Products (FP) measurements were able to get a reliable diagnosis relative to the status of the reactors and the spent fuel pools (SFP). For instance, based on these FP measurements, it was possible to conclude that the main origin of the releases was coming from the cores and not from the SFP, in particular for SFP-4 which was of high concern, and that the degradation level of the reactors was very large, including probably an extensive core melting. To improve the reliability of this kind of diagnosis, the necessity to get such measurements as soon as possible after the accident and as near as possible from the reactor was stressed [1]. In this way the present DECA-PF project intends to develop a new and innovative instrumentation relative to on-line FP

measurements, which would be consistent with the design of the French nuclear power plants on which sand bed filters have been implemented for severe accident management (SAM) [2].

In this paper, after having recalled the design and operating conditions of the sand bed filters used in France as Filtered Containment Venting System (FCVS), the general content of the DECA-PF project is described in part 3. Part 4 is devoted to the updating of the Source Term at the outlet of the sand bed filter, where this innovative instrumentation could be implemented. Finally, the preliminary feasibility study performed on gamma spectrometry measurements is presented on part 5. Two other papers of this conference complete the description of the work performed within the DECA-PF project: they concern feasibility studies relative to optical absorption spectroscopy, a more innovative instrumentation devoted to quantify FP gaseous species, such as tetra-oxide of ruthenium, which could be produced in case of lower head failure [3] as well as the gaseous forms of iodine, molecular and organic [4].

## II. RECALL OF THE FRENCH EQUIPMENT AND GUIDELINES IN CASE OF SEVERE ACCIDENT

Studies and R&D programmes in the field of Severe Accidents have been initiated in France following the publication, in October 1975, of the WASH-1400 (Reactor Safety Study), so-called “Rasmussen Report”, the first PRA relative to nuclear power plants. The TMI-2 accident, in March 1979, enhanced the interest of this work. New guidelines were then elaborated by the French nuclear safety authority and Electricité de France (EDF) in order to cope with the consequences of severe accident including large core melting. In such a case, the priority is given to the containment integrity and the limitation of radioactive material releases into the environment. Specific procedures (called “U” for ultimate) were implemented for each situation which could lead to the failure of the containment and a high radioactivity release. In particular the containment could be endangered by a slow pressure increase resulting from the production of steam when water is injected on a hot core, and/or by the production of incondensable gases, such as carbon oxides, resulting from the interaction of the melted core, called corium, with the concrete (named MCCI for Molten Core Concrete Interaction). In such a situation, the U5 procedure is a SAM measure devoted to depressurize the containment through a filtered venting system [2], [5]. Figure 1 gives a general overview of this system, which has been installed on all the French PWRs in operation.

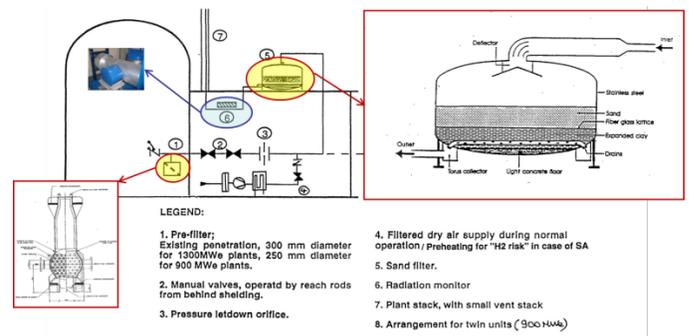


Fig. 1. General diagram of the FCVS in French PWRs

The main components are the following:

- A metallic pre-filter located inside the containment, which aims at trapping at least 90% of the aerosol particles, in order to reduce the Source Term trapped in the sand bed filter. This feature avoids the generation of heat inside the sand bed filter which could lead to potential FP re-volatilisation, and allows the circulation of the personnel on the site even for the most pessimistic situations [6],
- An orifice plate located just downstream from the manual isolation valves, ensuring the vented gases expansion to the atmospheric pressure,
- The sand bed filter, patented by EDF, is set on the roof of the nuclear auxiliary building. It is thermally insulated in order to avoid steam condensation and, as a consequence, the risk of hydrogen explosion during steady state conditions. A pre-heating system has been installed in order to avoid combustion risk due to steam condensation on cold structures at the opening of the system. Its main characteristics are: inner diameter 7.3 m, height 4 m, operational weight 92 tons. The sand bed is 0.8 m thick, supported by layers of light weight concrete and expanded clay.
- A radioactivity measuring device downstream the sand bed filter. It uses a NaI gamma spectrometer with automatic energy calibration, and is designed to measure separately the fission gases, iodine and caesium released activities, when the depressurization is operating.

The filtering efficiency was characterized by the French experimental programs PITEAS (small scale experiments) and FUCHIA (full-scale experiments) [2], [7]. The minimal decontamination factors issued from these programs are set to 1000 for aerosol particles, 10 for molecular iodine, 1 for organic iodine. These values still need R&D program, in particular for the gaseous forms of iodine and ruthenium, presently performed through the European PASSAM project [8] and the French MIRE program [9].

The FCVS is put into operation by opening manually the containment isolation valves, at least 24 hours after the beginning of the accident, when the pressure increases slowly and reaches the design value of 5 bars.

### III. GENERAL CONTENT OF THE DECA-PF PROJECT

Three complementary techniques, devoted to measure on-line the FPs released at the outlet of the sand bed filter, are being studied in the DECA-PF project: gamma spectrometry, gas chromatography and optical absorption spectroscopy. All of them would be robust, able to operate under the severe environment encountered in such accidental situations.

These measurements aimed at improving the assessment of radioactive materials released in the environment: fission gases, volatile FPs, and so far as possible all other FPs transported under aerosol particles. A second objective of these measurements is to help in drawing a diagnosis relative to the core degradation, in a similar approach to what was done during the weeks following the Fukushima accident [1].

Gamma spectrometry is a technique widely used on industrial scale. It can be noticed in particular that the present radiation monitor equipment measuring the radioactivity at the outlet of the sand bed filter is a NaI gamma detector. Gamma spectrometry gives access to the main radioactive gases and FPs, except pure beta emitters. In the DECA-PF project, the objective is to study the feasibility of a new system, which could replace the present NaI detector, and could be able to measure a wide range of FPs released in the environment by using detectors of better resolution. If the feasibility is confirmed, a prototype could be built at the end of the project.

Gas chromatography is a complementary technique which gives access to the amount of fission gases released in the environment (mainly the stable elements Xe and Kr). Since they are not trapped in the sand bed filter, the quantitative measurement of these elements allows to assess the level of the core degradation (in a similar way to the measurement of  $^{133}\text{Xe}$  by gamma spectrometry, see part 5). In addition, the detection of CO and/or CO<sub>2</sub> can be interpreted as a signature of MCCI resulting from the rupture of the reactor pressure vessel.

Optical absorption spectroscopy is especially dedicated to the measurements of FPs which are under gaseous forms inside the containment, other than the fission gases Xe and Kr. The first category is the gaseous forms of iodine, molecular and organic, which can persist inside the containment in the long term for every kind of accidental scenario. The second category is the tetra-oxide of ruthenium RuO<sub>4</sub>, which can be produced after the rupture of the lower head, when the air of the containment enters in the reactor vessel and oxidises the non-volatile ruthenium still present in the reactor core. This innovative technique allows quantifying these gaseous species at trace level. RuO<sub>4</sub> measurements are studied by high resolution FTIR (Fourier Transform Infrared Spectroscopy) [3], molecular iodine by IBB-CEAS (Incoherent Broad-Band Cavity-Enhanced Absorption Spectroscopy) and organic iodine by FTIR and/or CRDS (Cavity Ring-Down Spectroscopy) [4].

### IV. UPDATE OF THE SOURCE TERM AT THE OUTLET OF THE SAND BED FILTER

The first task of the DECA-PF project was to establish the potential Source Term at the outlet of the sand bed filter. This Source Term is used as input data for the other tasks of the project. This work has been done based on the up-to-date knowledge relative to the FP behaviour under severe accident conditions, resulting from the most recent experimental R&D programs. Two Source Terms were established, based on realistic accidental scenario leading to the opening of the sand bed filter. They correspond respectively to an upper boundary scenario and a lower boundary scenario.

The upper boundary scenario is a severe accident leading to a total core melting, rupture of the vessel lower head, entrance of air through the reactor vessel and corium relocation to the reactor pit. This scenario leads to the production and release of gaseous RuO<sub>4</sub> by air ingress and to the production of carbon oxides by MCCI. In this scenario, it is considered that the containment spray fails in order to minimize the FP deposits in the containment (and therefore maximize the aerosol concentration in the atmosphere of the containment). Finally, it is considered that the U5 procedure is activated 24 hours after the beginning of the accident.

The lower boundary scenario is a less severe accident, characterised by a LOCA (Loss Of Coolant Accident) with late recovery of water safety injection. In this scenario, we consider a rupture of all the fuel rods, but no core melting. As for the upper boundary scenario, the containment spray is considered to fail, but in this case, in order to avoid steam condensation and, as a consequence, reach realistic conditions to implement the U5 procedure 24 hours after the beginning of the accident.

A third and intermediate scenario has also been investigated for gamma spectrometry measurements, since it does not affect the other techniques. It is relative to the impact of the time of the U5 filter opening on short half-life FP detectability; it considers the upper boundary scenario with an opening 8 days after the reactor shut down, instead of 24 hours.

The hypotheses relative to FP behaviour have been updated. For the overall behaviour, the results of the integral PHEBUS-FP program have been taken into account [10]. The released fractions into the containment have been completed with the results of the separate-effect tests performed in the VERCORS and VERDON programs [11]-[12]. For the iodine speciation, it has been taken into account the results of the CHIP program [13] relative to its transport in the reactor coolant system, and of the EPICUR program [14] relative to its behaviour in the containment. Finally, the fraction of RuO<sub>4</sub> has been evaluated, according to the most recent knowledge of its behaviour under nuclear severe accident [15], including the first insights from

the OECD STEM program [16], as well as the VERDON-2 test [17].

For the filter decontamination factors (DF), the following values have been applied: 1000 for FPs under aerosol forms, 10 for molecular iodine, 1 for the fission gases Xe and Kr, organic iodine and RuO<sub>4</sub>. However these hypotheses still suffer from large uncertainties, in particular the DF for gaseous iodine and RuO<sub>4</sub>. They would have to be updated later, according to the present R&D programs relative to this topic [8]-[9].

As an example for gas chromatography and optical absorption spectroscopy, the volumetric concentration of the gaseous species flowing in the pipe located at the outlet of the sand bed filter for the upper boundary scenario is given in table I.

TABLE I. AMOUNT OF GASEOUS SPECIES AT THE OUTLET STACK OF THE SAND BED FILTER FOR THE UPPER BOUNDARY SCENARIO

Gaseous species	Maximal concentration (atoms/m <sup>3</sup> )
Xe	5.52x10 <sup>21</sup>
Kr	6.28x10 <sup>20</sup>
RuO <sub>4</sub>	3.23x10 <sup>18</sup>
I <sub>2</sub>	3.52x10 <sup>16</sup>
ICH <sub>3</sub>	3.52x10 <sup>17</sup>

For gamma spectrometry, the volumetric activity has been evaluated for the main FPs and covers more than 100 isotopes (see section 5).

## V. ON-LINE GAMMA SPECTROMETRY STUDIES

### A. Objective of the measurements

The first objective is to quantify the amount of fission gases released into the environment. Since they are not reactive in the containment and not trapped in the sand bed filter, they load the potential highest radioactive release. In addition, their quantitative assessment makes it possible to evaluate the level of the core degradation; it is indeed possible to correlate their released fractions with the amount of core melting. Among them, the main isotopes which would have to be measured are the following:

- <sup>133</sup>Xe; with a half-life of 5.2 days, it is measurable at any time of the sand bed filter opening, from 24 hours up to around 15 days. Its detectability could however be impeded by its low gamma ray energy at 81 keV.
- <sup>135</sup>Xe and <sup>88</sup>Kr could give complementary information with a possibly better detectability, in particular for <sup>88</sup>Kr which has gamma ray at high energy (2196; 2392 keV ...) in an area without any interference. But their very short half-lives, respectively 9.1 hours and 2.8 hours limit their potential measurement in the first hours after the beginning of the accident, so probably only for sand bed opening at 24 hours.

The second objective is to quantify FPs which are partly under gaseous forms in the containment, iodine and ruthenium. Even if their gaseous fraction in the containment is very low compared to their aerosol forms, they load the main radiological impact in the environment, since these gaseous species are little or no trapped in the sand bed filter and are more hazardous for human beings than rare gases. In this category, the more reliable isotopes to measure are the following:

- <sup>131</sup>I for the iodine release. With its half-life of 8 days, it is measurable at any time of the sand bed filter opening, from 24 hours up to around 15 days. Its detectability could however be impeded by peaks overlapping at its main gamma ray of 364.5 keV. Other complementary iodine isotope of interest is <sup>133</sup>I (half-life of 20.8 hours, and main gamma ray at 529.9 keV).
- <sup>103</sup>Ru for ruthenium release. Its half-life of 39.3 days makes possible to measure it at any time of the sand bed filter opening. In addition, its main gamma rays at 497.1 keV is more favourable in terms of detectability than <sup>106</sup>Ru, gamma rays of which are located in energy areas where other rays of volatile iodine are presents (<sup>132</sup>I).

The third objective is to quantify all other FPs transported under aerosol particles. This is the most challenging task, since their concentrations in the flow at the outlet of the sand bed filter are very low, as a result of the filter high efficiency. Nevertheless, their measurements would be a useful complementary way to get a diagnosis of the core status by analyzing the presence of FPs of decreasing volatility, representative of a more and more degraded core. It can be noticed that, instead of fission gases, this diagnosis does not need a quantitative measurement, since it is based on ratio of FP of decreasing volatility with a volatile FP, such as cesium.

- Volatile FPs are released as soon as the rupture of the fuel cladding occurs. In this category, the most reliable isotopes are <sup>131</sup>I, <sup>133</sup>I, <sup>132</sup>Te, <sup>134</sup>Cs, <sup>136</sup>Cs and <sup>137</sup>Cs.
- Semi-volatile FPs are characterized by a significant release from the irradiated fuels, sometimes equivalent to those of the volatile FPs, but requiring a higher temperature in the reactor core, in the range of 1800-2200°C, thus below fuel melting. Their presence in the atmosphere of the containment is nevertheless limited by a high retention in the reactor vessel and the reactor cooling system. Among them, the most reliable isotopes are <sup>99</sup>Mo and <sup>140</sup>Ba.
- Low volatile FPs are characterized by a low release from the irradiated fuels, occurring at very high temperature when the core begins to melt. Their presence in the atmosphere of the containment can be clearly considered as a signature of core melting. Among them, the main confident isotopes are <sup>95</sup>Nb, <sup>103</sup>Ru, <sup>141</sup>Ce, <sup>144</sup>Ce and <sup>239</sup>Np.

### B. Status of feasibility studies

Based on the updated Source Terms previously established, a first assessment of gamma spectra which could be recorded in sight of the pipe located at the outlet of the sand bed filter has

been done, including more than 100 isotopes and more than 4000 gamma rays.

Figure 2 shows the “emission spectrum” (product of the activity of each FP by the gamma ray intensity) for the upper boundary scenario, above which a gamma spectrum corresponding to a LaBr<sub>3</sub> detector has been modelled by the MCNP code [18].

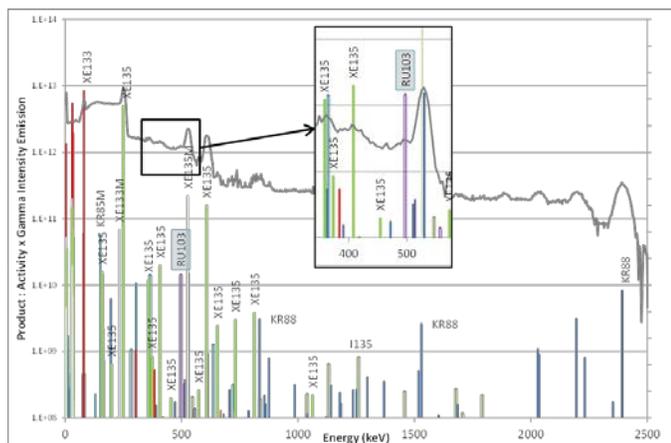


Fig. 2. Gamma spectrum modelled with a LaBr<sub>3</sub> detector for the upper boundary scenario at the outlet of the sand bed filter

This preliminary analysis, applied on the three boundary scenarios, shows that:

- Fission gases would be easily measurable, in particular <sup>133</sup>Xe for each boundary scenario,
- Gaseous iodine (<sup>131</sup>I) and ruthenium (<sup>103</sup>Ru) measurements would need to develop a specific gamma spectra processing and/or use a gamma detector of high resolution, such as Ge(HP), for scenario corresponding to the opening of the sand bed filter at 24 hours, in order to be able to separate their respective gamma ray at 364.5 keV and 497.1 keV to potential interferences with <sup>135</sup>Xe (secondary ray at 358.4 keV) and <sup>135m</sup>Xe (main gamma ray of high intensity at 526.6 keV).
- Other FPs would be very difficult to measure by this on-line measurement, due to their very low activity resulting from their high retention in the sand bed filter. Their potential measurement would require implementing a second system, in derivation to the main pipe, where these FPs could be accumulated on a trapping media without the presence of the fission gases, in order to increase their detectability.

The use of high resolution Ge(HP) detector has been rejected for robustness purpose. As a consequence, the feasibility study has been focused in a second step on the aerosol FP detectability through an accumulating media as described on figure 3.

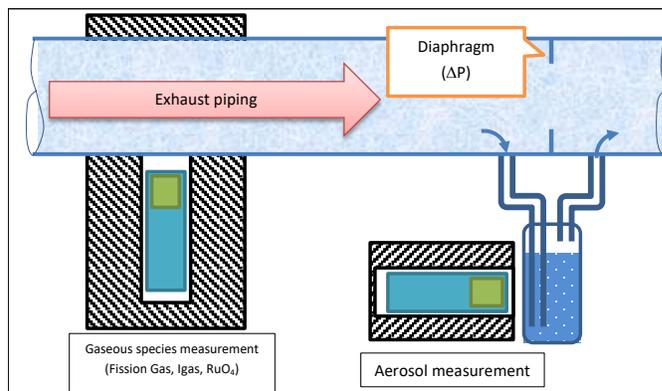


Fig. 3. Aerosol measurements on a trapping media in derivation to the main pipe

Gamma spectra corresponding to a LaBr<sub>3</sub> detector were modelled for each boundary case. LaBr<sub>3</sub> detector has been selected since it represents the best “compromise” between the “robustness goal” and the “resolution”, this latter being better than for a NaI detector by around a factor 2. Figure 4 shows this spectrum for the upper boundary scenario.

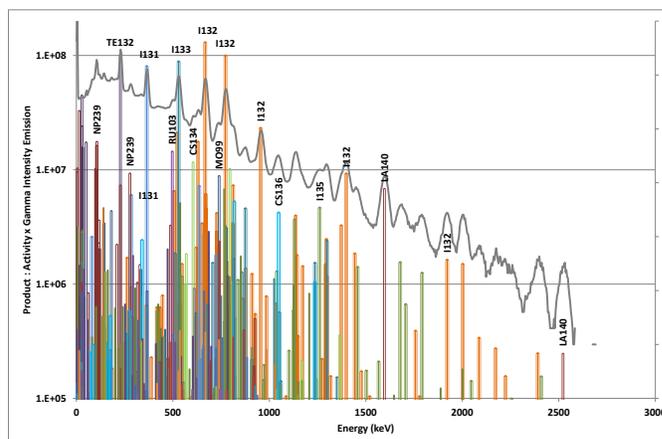


Fig. 4. Gamma spectrum modelled with a LaBr<sub>3</sub> detector in sight of the proposed aerosol device for the upper boundary scenario

Even if the spectrum magnitude is still very high, covering more than 5 decades, this first analysis appears promising, confirming the ability to measure volatile FPs such as <sup>132</sup>Te, <sup>131</sup>I, <sup>134</sup>Cs, <sup>136</sup>Cs, and possibly also FPs of lower volatility, such as <sup>95</sup>Nb, <sup>99</sup>Mo, <sup>103</sup>Ru, <sup>239</sup>Np, using a dedicated gamma spectra processing.

## VI. CONCLUSION

A general overview of the DECA-PF project, devoted to improve the instrumentation at the outlet of the sand bed filters of the French nuclear power plants, in case of severe accident, has been presented. The feasibility of implementing on-line gamma spectrometry equipment is under study, based on an updated Source Term. In order to be able to measure on-line by this technique all the fission products, which could be potentially released into the environment, it seems that at least two separate systems are necessary, in order to be able to

discriminate the fission gases (Xe, Kr), the fission products which are partially under gaseous forms in the containment (iodine in all scenarios, ruthenium in case of lower head rupture), and the other fission products which are under the form of aerosol particles in the containment.

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