

The OSCAR-FP V1.4 code: Simulation of fission product and alpha emitter contamination in PWR circuits

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Abstract

Understanding the PWR primary circuit contamination is a crucial issue for reactor operation and design, as well as in the perspective of decommissioning. For this purpose, the CEA has developed, in cooperation with EDF and Framatome, the OSCAR code package, a unique tool to simulate PWR contamination by corrosion products, fission products (FP), and actinides.

This paper presents a general overview of OSCAR-FP V1.4, the latest version of OSCAR computation scheme dealing with fission product and actinide contamination. The source term calculation for these nuclides and the behavior considered in the primary circuit are described in detail, as well as the validation process against primary activity measurements.

Fission products are released in the primary circuit in case of a fuel cladding defect, depending on the nuclear fuel characteristics: composition, temperature, and burn-up. Therefore, the CEA fuel code ALCYONE is included in OSCAR-FP and allows calculating the source term for these nuclides. However, fission product release into rod free volumes is increased due to fuel oxidation in case of a clad defect. The fuel code ALCYONE has then been adapted to defective fuel situation through the adjustment of different parameters: enhanced diffusion in the fuel grain, enhanced grain boundary fracturing, and reduced fuel thermal conductivity.

In addition, in the event of significant cladding damage, fuel oxide particles may disseminate in the primary circuit. These particles solubilize very slowly and are still present in the primary circuit several cycles after the dissemination. They include long life fission products as well as heavy nuclides among which alpha emitters and fissile nuclides. They are mainly deposited on the primary surfaces and are subject to further erosion and deposition mechanisms. As their behavior is linked to the one of corrosion product particles, a specific OSCAR computation scheme is used, allowing coupled simulation between corrosion products and actinides.

Calculated primary activities are compared to measured fission product and alpha activities, obtained from primary gamma activity monitoring as well as from sampling and analysis campaigns of primary coolant or surface deposits. Code qualification is obtained for different types of nuclides: i) gaseous and volatile gamma emitters Xe, K, I, Cs whatever the isotope half-life, ii) alpha emitters, which are mainly insoluble, iii) long life beta emitter ^{90}Sr , which undergoes adsorption/desorption mechanism and is sensitive to pH.

The paper shows that the OSCAR-FP V1.4 code is a powerful tool to study fission product and alpha contamination in PWR primary circuits, and is helpful for fuel and waste management as well as for the alpha risk assessment. Moreover, new results obtained with the V1.4 version are highlighted: mechanistic erosion and deposition models have been calibrated for corrosion products and applied to actinides. This provides a good simulation of actinides with very few adjusted parameters, and confirms the coupled behavior of corrosion products and actinides in the primary circuit.

1. Introduction

Different radioactive nuclides are responsible for the radioactive contamination of the PWR primary circuit. They mainly come from bulk corrosion of primary materials (stainless steel, nickel alloy, stellites...), which induces the release of several kilograms per year of corrosion products in the primary circuit. These corrosion products become radioactive when exposed to neutron flux. They are the main contributor to the dose rate around the primary circuit. However, in case of defective fuel cladding, radioactive fission products also contaminate the primary circuit. In some rare cases, a few grams of fissile material including alpha emitter actinides may also be released in the primary circuit.

While operating or designing a nuclear power plant, it is necessary to address important challenges related to the circuit contamination: optimizing the personnel exposure to radiation, limiting the activity of the wastes and effluents produced during the reactor lifetime and preparing decommissioning. Therefore, the assessment of the radio contamination of PWR circuits is a crucial issue for power plant operators and suppliers.

To address this issue, the OSCAR code package is targeted at simulating PWR primary and auxiliary circuit contamination in nominal and cold shutdown conditions. It is qualified for corrosion product, fission product and actinide contamination.

This paper focuses on the simulation of fission products and actinides with the latest OSCAR version (V1.4). However, as corrosion products represent most of the contamination mass deposited on the primary surfaces, they definitely determine the evolution of other deposited radionuclides, in particular actinides. Thus, the coupled behavior between corrosion products and actinides is considered.

After a general overview of the code basics in section 2, the paper presents the main principles of modeling as well as the validation process for fission products and actinides in case of defective fuel cladding (section 3) and in case of fissile material dissemination (section 4).

2. General overview

The OSCAR code package has been developed within the framework of R&D cooperation between the French Atomic and Alternative Energy Commission (CEA), Electricité de France (EDF) and Framatome. The code uses a fully object-oriented design and a modular structure. Circuits are discretized into as many regions as necessary, according to the geometric, thermal-hydraulic, neutronic, material and operating characteristics. A user-friendly interface for input data and visualisation of results is included.

Even though the origin is different for corrosion products, fission products, and actinides, they are subject to the same severe conditions (300 °C, 150 bar, neutron flux, water velocity up to 15 m.s⁻¹) and follow the same transport mechanisms in the primary circuit. The main processes involved are erosion/deposition, dissolution/precipitation, adsorption/desorption, convection, purification, neutron activation, radioactive decrease [1]. The different mechanisms can be activated separately. Taking into account these mechanisms, mass balances are calculated for each region and for each relevant nuclide (radioactive and stable), either at the metal surface or in the primary coolant (particles and soluble forms). Regarding fission products, 78 nuclides within 27 filiation chains are handled in the OSCAR code. For actinides, a 22 nuclide filiation chain is considered.

Three different computation schemes are now proposed to the OSCAR user. The two original schemes (OSCAR-CP and OSCAR-FP) are dedicated to corrosion products and to fission products and actinides respectively. They can be run independently. However, considering the coupled behavior of corrosion products and actinides, a third scheme (OSCAR-CPFP) has been developed. It allows calculating all types of radio contaminant with the same data set. In return, the data set is more complex as it gathers the data needed for the simulation of the different elements. Compared to the original schemes, the third scheme has no impact on corrosion product simulation as actinides and fission products do not affect the corrosion product behavior. It is useful in case of fissile material dissemination (see § 4).

3. Fission product release out of defective rods

In the event of a rod cladding defect, gaseous (Xe, Kr) and volatile (I, Cs) fission products are released into the primary circuit. The process involves several stages: release out of the rod pellets into the free volumes, migration along the fuel-cladding gap and release through the defect into the primary coolant [1].

Once in the primary circuit, the behavior of these species is simply modeled:

- They are considered as fully soluble.
- They do not interact with the primary walls in the nominal operating conditions.
- The Henry's law coefficient is used to evaluate the partition between liquid and gaseous phases (purification tank, pressurizer).
- Volatile fission products are trapped on the ion exchange resins, whereas gaseous species may be removed through gas flush.

The concentration of the gaseous and volatile fission products in the primary coolant is then easily calculated if the release rate is known. As far as these species are concerned, the assessment of the release rate from a defective rod is the main challenge for the OSCAR code. The other species are supposed not to be released because their diffusion in the fuel matrix is not fast enough.

3.1 Release out of rod pellets

Because of the presence of water or vapor inside the defective rod, oxidation of the fuel matrix occurs, which strongly enhances the release of fission products out of the rod pellet compared to a tight fuel rod. Modeling of this phenomenon should rely on defective fuel characterization but given the lack of such experiments, the strategy has been to adapt the MARGARET module developed and validated for tight fuel.

MARGARET [2] is an advanced mechanistic model of fission gas behavior in PWR fuel. It provides the detailed location of the gases – inter- or intragranular, in bubbles or dissolved –, in addition to gas release and swelling. It is developed by the CEA within the framework of the PLEIADES platform [3] and its fuel behavior application ALCYONE, which supplies the thermomechanical data used by MARGARET.

The MARGARET code, which initially dealt only with stable gases, has been adapted to the radioactive fission products. At each time step, a first calculation is performed for stable gases and provides the fuel microstructure: gas bubble distribution as well as grain boundary fracturing or fuel restructuring into HBS (High Burnup Structure). Then a second calculation is performed for radioactive fission products, using the previously calculated fuel microstructure and taking into account the radioactive decay constant and the filiation chain.

Like stable gases, radioactive fission products are released from the grain by diffusion, either towards grain boundaries or towards free surfaces, that means grain surfaces connected with free volumes (fracture or open porosity or fuel-cladding gap).

When released at free surfaces, fission products are considered to migrate instantaneously to the free volumes. When released at the grain boundary, they may migrate towards the free volumes by percolation, when the bubbles at the grain boundaries interconnect at high burnup. The free surface fraction is simulated by the MARGARET code, and is around 1 or 2 % in a tight fuel configuration.

Moreover, to fit the experimental release of short life radionuclides at low temperature, the knockout mechanism [4] at the grain free surface has been added. Knockout means ejection of an atom struck by a fission fragment or another atom of the lattice, due to elastic collisions. The important parameter for this mechanism is the thickness of the layer where knockout can lead to the ejection of fission products out of the grain. This thickness is assessed about 15 nm. The knockout flux can be expressed as an additional sink term in the knockout layer to the diffusion term. This flux is proportional to the fission rate and to the fission product concentration in the layer. Knockout does not significantly affect released fractions by diffusion for ^{133}Xe (5 day period) and ^{131}I (8 day period), which are used for defective fuel calibration (see § 3.3). However, it strongly increases the release of shorter half-life fission products, particularly at low fuel temperatures, as they do not have time to diffuse out of the grain [1].

Thus, mechanisms involved in fission product release into the free volumes are mainly: diffusion within the fuel grain, knockout at the free surface of the grain, and percolation through the grain boundary at high burnup. The behavior is considered to be the same for the different species (Xe, Kr, I, Cs). In particular, the intragranular diffusion coefficient has the same value for each of these species. Only the radioactive decay period and the filiation affect the released fraction. In future, a chemical module will be included in MARGARET, taking into account the solubility of elements in the fuel matrix.

In the event of a cladding defect, most of the primary fluid activity is due to xenon, krypton and iodine isotopes with radioactive half-life shorter than 8 days (see Figure 1). The activity of these radioactive nuclides is continuously monitored during reactor operation.

If the burnup is not too high (no percolation) and the irradiation power not too low (knockout is negligible), the release of these isotopes of interest is due only to diffusion out of the grain. In first approximation, the diffusion coefficient is an apparent diffusion coefficient, taking into account not only the intrinsic intragranular diffusion coefficient but also the trapping and re-resolution coefficients of pores and bubbles. This approximation is always valid for long life fission products and for shorter life nuclides as long as precipitation bubbles do not appear in the grain (burnup lower than 30 GWd/t).

The released fraction can then be expressed as:

$$\frac{R_{pellet}}{B} = F_{free} \cdot \frac{3}{a} \cdot \sqrt{\frac{D_{app}}{\lambda}} \quad (1)$$

where R_{pellet} is the release rate from the pellets into the free volumes, B is the fission product birth rate in the pellets, F_{free} is the fraction of grain surfaces connected with free volumes, a is the grain radius, D_{app} is the apparent diffusion coefficient of the fission product in the grain, and λ the radioactive decay constant of the fission product.

In the logarithmic diagram of the released fractions out of pellets versus the radioactive decay constant, the maximal absolute value of the slope is $\frac{1}{2}$ if the burnup is lower than 30 GWd/t. The slope is lower at low power because knockout becomes faster than diffusion for short life fission products.

3.2 Release out of the rod

Once released from the pellet, as long as the fuel-cladding gap is not closed, noble gases are supposed to migrate to the defect rapidly, where they are released into the primary coolant according to the conductance of the defect. However, at high burnup, the gap is partially or totally closed and the migration is far slower.

Considering iodine and cesium isotopes, the release mechanisms are not well understood. During nominal reactor operation, trapping occurs in free volumes, which makes the release rate out of the rod very difficult to assess. However, during power transients at low level, water enters the free volumes and leads to the release of almost the total amount of iodine and cesium present in free volumes.

This behavior is taken into account in the modeling through two parameters:

- The conductance of the defect: ν (s^{-1})
- The trapping coefficient in the free volumes (ratio of trapped to non-trapped activity): α

According to the modeling, the rod release is the product of defect conductance and non-trapped activity in free volumes. Then, the activity balance in free volumes is the following [A (Bq): total activity of the fission product in the free volumes, R_{pellet} (Bq/s): release rate from the rod pellets into the free volumes, R_{rod} (Bq/s): release rate from the rod into the primary coolant, λ (s^{-1}): radioactive decay constant]:

$$\frac{dA}{dt} = R_{pellet} - \lambda \cdot A - R_{rod}, \quad \text{with } R_{rod} = \frac{\nu}{1 + \alpha} \cdot A$$

In steady state, the relationship between the instantaneous released fractions from rod and pellets is:

$$\frac{R_{rod}}{B} = \frac{R_{pellet}}{B} \cdot \frac{\frac{\nu}{1 + \alpha}}{\lambda + \frac{\nu}{1 + \alpha}}$$

This model is very simple and cannot account for high burnup defective rod, where gases are mechanically retained. Moreover, the parameters ν and α are difficult to assess, though ν can be related to the defect size. Nevertheless, the model is useful for interpreting the instantaneous released fractions in nominal operation (R_{rod}/B), which are obtained from the monitored primary activities and the calculation of the birth rate B .

For noble gases, $\alpha = 0$ if the fuel-cladding gap is not closed. Then $\frac{R_{rod}}{B} = \frac{R_{pellet}}{B} \cdot \frac{\nu}{\lambda + \nu}$

Moreover, if $\lambda \ll \nu$ (this is generally the case for the monitored radionuclides if the size defect is not too small), we obtain $\frac{R_{rod}}{B} = \frac{R_{pellet}}{B}$: the instantaneous released fractions from the pellets and from the rod are equivalent.

For volatile species, the trapping coefficients are assessed to 600 and 10 000 in nominal operation, respectively for iodine and cesium.

Then $\lambda \gg \frac{\nu}{1 + \alpha}$ for the nuclides of interest.

In Figure 1, the logarithmic diagram shows the simulated rod released fractions in nominal operation versus the radioactive decay constant of the fission products. It represents the ideal case where the burnup is low, the size defect is large and the irradiation power is high enough to neglect the knockout mechanism. In this case, the theoretical slope is around $-1/2$ for noble gas isotopes (red line) and $-3/2$ for iodine isotopes (green line). The blue line represents the released fractions in case of fissile material contamination and will be described in section 4.

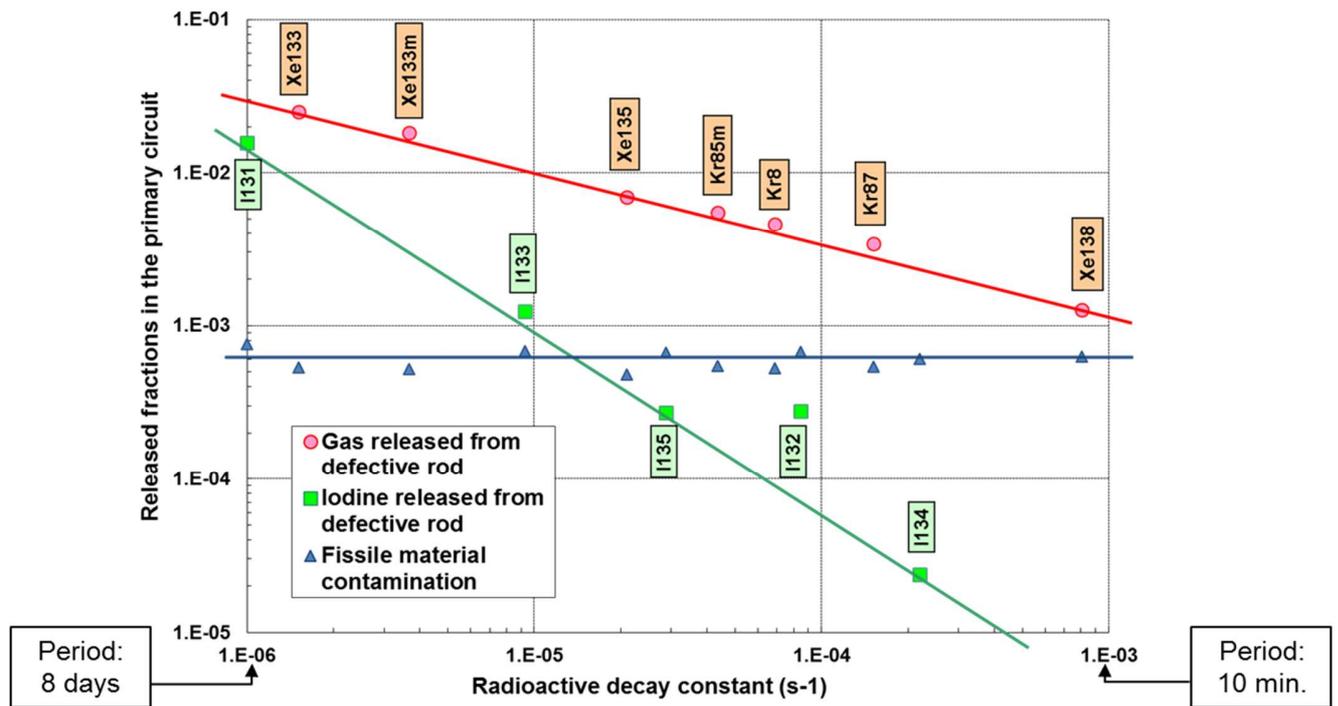


Figure 1 – Fission product released fractions in nominal operation

Lastly, the MARGARET code is used to assess the iodine and cesium activity which is trapped in free volumes. This activity is released during a power transient and leads to an activity peak in primary coolant. Moreover, the code allows setting to zero the trapping coefficient during the transient and then calculating a maximum peak height considering that iodine and cesium are released according to the defect conductance. However, it cannot reproduce the actual kinetic of the iodine or cesium release which is observed during the transient and is often slower than the theoretical one.

3.3 Validation method

The ALCYONE/MARGARET code is validated for tight fuel. The latest version has been adapted to simulate the release from the pellets in case of a cladding defect.

For defective rods, oxidation of the fuel matrix leads to released fractions much higher (by a factor of roughly 30) than for non-defective fuel rods. This impact from oxidation is taken into account in the simulation by the following adjustments:

- Fraction of grain surfaces connected with free volumes F_{free} : it is evaluated to 30%.
- Intragranular diffusion coefficient D : a correction factor is applied to the intrinsic stable gas diffusion coefficient used for tight fuel. It is assessed to 1.1.
- Fuel temperature: besides the fact that for a defective configuration, water vapor replaces helium in the free volumes of the rod and that the internal pressure of the rod is balanced with external pressure (no further clad creeping), a degradation of the thermal conductivity of the fuel is taken into account (factor 0.65).

The calibration of these model parameters (fraction of free surface, intragranular diffusion coefficient and thermal conductivity degradation) has been performed against a unique experimental database including defective PWR rods well identified and characterized, as well as analytical irradiations in the SILOE reactor.

Actual fuel released fractions are obtained as follows from the primary activity monitoring:

- The ^{133}Xe released fraction from the fuel in nominal operation is supposed to be equal to that of the rod, as ^{133}Xe is the longest half-life noble gas which is monitored (see Figure 1). Then it is not affected by the transfer in the fuel-cladding gap or by the size of the defect. However, the absence of peak during power transients must be checked, ensuring that gases are not mechanically retained in the rod in nominal operation.
- The ^{131}I released fraction from the pellets is obtained from activity monitoring during transients, considering that activity trapped in the free volumes is totally released during the transient.

As they are likely not affected by the knockout mechanism, the fuel released fractions obtained for both radionuclides correspond to equation (1). They should be at the same level as their radioactive decay constant is similar. This allows reducing the uncertainty about the measured released fraction.

Figure 2 gives a comparison between measurements and OSCAR calculations after calibration, concerning the released fractions from defective UO_2 fuel rods of the database (including EDITH 02 and EDITH 06 analytical irradiations, respectively for low burnup and 50 GWd/t UO_2 rods). The validation domain is up to 50 GWd/t. We note a relatively good agreement (within a factor of 2) between experimental and calculated released fractions. Such a fair agreement (not shown here) is also obtained for analytical irradiations EDITHMOX 01 (low burnup MOX rod) and EDITHMOX 02 (50 GWd/t MOX rod), which allows widening the validation domain to MOX fuel, for the same burnup range.

From the simulation of the defective rods of the experimental database, we observe that the burnup activated mechanisms are negligible for ^{133}Xe and ^{131}I : intergranular percolation, trapping in intragranular precipitation bubbles, HBS restructuration. Nevertheless, these mechanisms could become significant at higher irradiation power.

3.4 Validation domain

Validation of released fractions out of the fuel pellets is obtained for ^{133}Xe and ^{131}I . However, the validation domain can be extended to shorter and longer half-life nuclides.

For iodine and cesium isotopes, the fuel released fractions are obtained from activity monitoring during power transient. The integral of the primary activity peak corresponds to the amount trapped in free volumes before the transient. It is related to the instantaneous release (Bq/s) for short life nuclides (^{131}I , ^{133}I) and to the cumulated release (Bq) for long life nuclides (^{134}Cs , ^{137}Cs). Indeed, steady state is not reached for long life nuclides. Uncertainty on cesium release obtained from primary activity measurements is higher than for iodine because of the unknown operation time for cationic resins. Moreover, the uncertainty is higher for ^{133}I than for ^{131}I due to the shorter radioactive period. However, despite the higher uncertainty, the measured released amounts for ^{134}Cs (period: 2

years), ^{137}Cs (period: 30 years) and ^{133}I (period: 21 hours) are consistent with the calculated ones, allowing the extension of the validation domain to each iodine and cesium isotope.

Regarding noble gas nuclides, the shortest half-life nuclides are sensitive to the defect conductance, leading to a steeper slope for the released fractions in the primary circuit versus the radioactive decay constant than for the pellet released fractions. We can verify that measured released fractions in the primary coolant are lower than or equal to the calculated pellet released fraction, and that they can be simulated adjusting the defect conductance if the fuel-cladding gap is not closed.

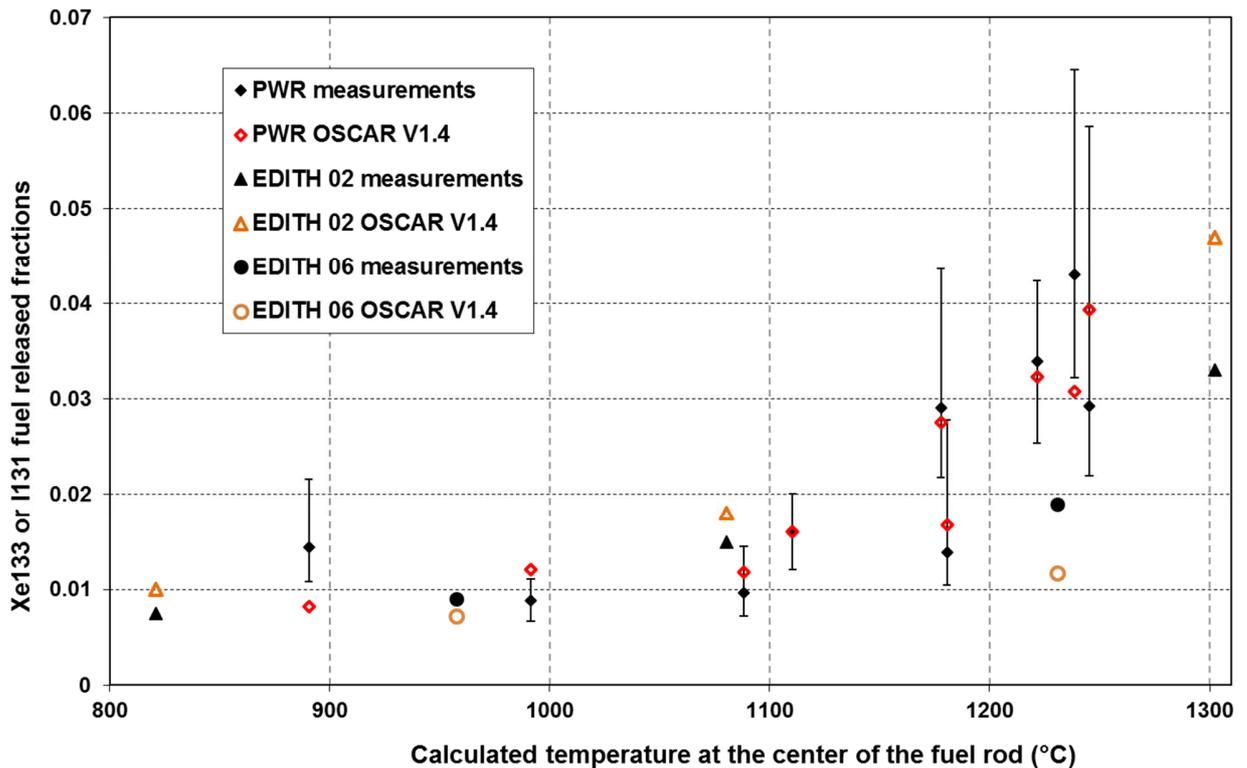


Fig. 2 – Comparison between experimental (black symbols) and calculated (red symbols) fuel released fractions

Finally, the validation domain for gaseous and volatile fission product release in the OSCAR V1.4 code is the following:

- Fuel:
 - UO_2 and MOX,
 - Burnup range up to 50 GWd/t,
 - Irradiation power range up to 220 W/cm.
- Fuel released fraction: validated for every isotope of Xe, Kr, I, Cs. The released fraction is sensitive only to the radioactive period. Difference between calculation and measurement is lower than a factor of 2 in the validation domain (see Figure 2).
- Released fraction into the primary circuit during nominal operation: validated for every gaseous isotope (Xe, Kr) if the conductance of the defect can be assessed and if the gap is not closed. A conservative value can be obtained considering a very high defect conductance.
- Total amount of activity released during a power transient: for every volatile isotope (I, Cs), a conservative value can be obtained considering no release during nominal operation. Then the total released amount corresponds to the calculated activity in the free volumes before the power transient.

3.5 Defective fuel diagnosis methods

Diagnosis methods of the defective fuel characteristics have been developed and gathered in the DIADEME software. These methods are based on the previously described modeling and have already been presented in detail [5]. They are used along with continuous gamma primary activity monitoring and are helpful for the operator to assess in operation the number of failed rods and their characteristics and to prepare the failed fuel replacement for the next core loading.

The guidelines of the methods are the following:

- Mean defective rod power: related to the slope of fuel released fractions versus radioactive decay constant. The absolute value of the slope increases with the irradiation power because diffusion becomes faster than knockout.
- Number of defective rods: related to the ratio of measured ^{133}Xe fuel released fraction to the calculated released fraction at the mean defective rod power.
- Defect size: related to the ratio of ^{131}I to ^{133}Xe released fractions into the primary coolant in operation. The ratio increases with the defect size.
- Defective rod burnup: related to the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio measured in the primary coolant during power transients. The ratio increases with the defective fuel burnup.
- Fuel nature (UO_2 or MOX) of the defective rod: related to the $^{85\text{m}}\text{Kr}/^{135}\text{Xe}$ activity ratio measured in the primary coolant. The ratio is higher for MOX fuel.

4. Fuel oxide dissemination

In addition to fission product release, fuel oxide may disseminate into the PWR primary circuit in the event of significant cladding damage. The disseminated fuel oxide is also called “tramp uranium”: though mainly deposited on the primary surfaces, it is continuously transferred between the different parts of the circuit through the erosion and deposition mechanisms.

The disseminated fuel oxide consists of actinides and fission products, which contaminate the primary circuit. Actinides include alpha emitters, which induce alpha contamination, and fissile nuclides (mainly ^{235}U and ^{239}Pu), which generate fission products directly in the primary circuit when under neutron flux. Fuel oxide dissemination causes a long-term contamination. Indeed, trapping on purification filters is not efficient as the actinide fraction transported in the primary coolant is very low.

4.1 Actinide contamination behavior

Fissile material disseminates in particulate form and is mainly insoluble in the primary circuit. The ALCYONE code is used to give the particle composition, which corresponds to the one of the disseminating fuel at the pellet surface.

Fuel oxide particles are mainly deposited on the primary surfaces. However, they undergo erosion and deposition, which induce transfer between out-of-flux surfaces and under flux surfaces. Consequently, fuel assemblies reloaded after the dissemination become rapidly contaminated with actinides.

When some fissile material is deposited under neutron flux, fission products are emitted directly by recoil in the primary coolant. As the recoil mechanism is immediate, the fission product released fractions are independent of the radioactive period. The blue line in Figure 1 illustrates this behavior for a few grams of disseminated material. Moreover, due to its short radioactive period (54 minutes), ^{134}I is the tracer of the fissile material under neutron flux. Indeed, as shown in Figure 1, its release from a defective rod is negligible: it can be measured in the primary coolant only in case of fissile material contamination.

Lastly, uranium oxide may solubilize. According to OSCAR modeling, it induces the separation between uranium and transuranium elements in the primary circuit. Then each species evolves separately, but transuranium elements remain in particulate form in nominal operation [6]. The rate of uranium oxide dissolution is still not well known. It seems to be very low in nominal operation but significant in oxidizing conditions. In any case, actinides in the primary circuit likely remain in the form of the disseminated fuel oxide during many cycles.

4.2 Modeling improvements for actinide simulation in OSCAR latest version

In the OSCAR previous version, empirical deposition and erosion rates were specified in the data set for actinides, except during cycles following the dissemination where corrosion product deposition rates were used [6]. With the last version V1.4, according to the hypothesis that actinides are transported by corrosion product particles, the goal is to use for actinides the erosion and deposition models which have been validated for corrosion products.

The deposition and erosion rates for corrosion products are obtained from mechanistic models [7,8]. The particle deposition rate takes into account 1) the turbulent diffusion and the effects of inertia, 2) the sedimentation for horizontal ducts and 3) the thermophoresis for temperature gradients between the coolant and the wall. Regarding the erosion rate, it results from the coolant friction forces, and involves the deposit thickness. These mechanisms have been calibrated for corrosion product simulation [8].

Before actinide calculation, a preliminary corrosion product calculation must be performed to evaluate the corrosion product deposited mass, which is needed for the erosion mechanism. Therefore, the OSCAR-CPFP computation scheme is used as it allows calculating corrosion products as well as actinides and fission products with the same data set. Consequently, it is possible to use the same deposition and erosion mechanisms for every insoluble species.

However, compared to corrosion products, the erosion mechanism is adapted for actinides as follows:

- The erosion flux for each isotope is proportional to the occupied surface fraction $\frac{S_{spec} \cdot m_k}{S_w}$ for actinides, whereas it is proportional to the mass fraction $\frac{m_k}{m_{tot}}$ for corrosion products. This is to account for the fact that actinides are deposited at the surface of a corrosion product layer and not intimately blended in the corrosion layer. In the above expressions, S_{spec} is the specific surface of deposited actinides (m^2/g), S_w is the wet surface of the region (m^2), m_{tot} is the total deposited mass of corrosion products in the region (g) and m_k is the deposited mass of the radiocontaminant k in the region (g).
- The erosion rate decreases with time on out-of-core surfaces, because actinides are supposed to be continuously covered by corrosion products.

4.3 Validation of alpha simulation

The actinide simulation with OSCAR V1.4 has been adjusted to fit CATTENOM 3 PWR measurements [6]. Fissile material dissemination occurred at cycle 8 during 231 days, with disseminating rods belonging to third cycle assemblies. The core was managed in three batches. At the end of the disseminating cycle, two batches were discharged and only the first cycle fuel batch was maintained in the core for two more cycles (cycle 9 and 10).

4.3.1 During nominal operation

Two calculations are presented hereafter with and without uranium dissolution in reducing conditions (during nominal operation and during end-of-cycle shutdown before primary circuit oxygenation). However, uranium dissolution in oxidizing conditions is considered for both calculations [6].

- n°1. "w/o U diss.": No uranium dissolution is considered in reducing conditions.
- n°2. "w/ U diss.": Uranium dissolution is considered in reducing conditions.

Calculation results are compared to the following measurements:

- ^{134}I primary activity, as tracer of fissile material under neutron flux, during the disseminating cycle and the seven subsequent cycles. It comes mainly from ^{235}U and ^{239}Pu fissions in the deposited fissile material.
- Alpha activity measured in the primary coolant, during the disseminating cycle and the seven subsequent cycles.
- Scraping on steam generator channel head surfaces, performed at the end of the disseminating cycle. Alpha analysis of the sample provides alpha surface activity on these surfaces. However, uncertainty on this measurement is high because the scraping is realized only on a few square centimeters.

From the ^{134}I primary activity comparison, the following adjustments are done:

- The dissemination rate and duration are adjusted during cycle 8 to match with the increase in ^{134}I activity as there is no phenomenological modeling of the dissemination. Compared to calculations with the previous OSCAR version [6], the dissemination rate has been increased by a factor 2.5. Indeed, the deposited fraction on the core surfaces is around 20% according to the corrosion product deposition rate, whereas it was assessed to 50% for actinides with the previous version.
- At the end of cycle 8, two fuel batches over three are discharged. Consequently the core contamination is automatically reduced by two thirds at the beginning of cycle 9. Nevertheless an additional reducing factor is applied at beginning of cycle 9 to take into account the fact that fissile material deposition is actually not homogeneous during dissemination and occurs preferentially in the vicinity of dissemination rods. This reducing factor is determined in order to match calculated with measured ^{134}I activity at the beginning of cycle 9.
- The specific surface for actinides in the deposit layer is adjusted to $700\text{ cm}^2/\text{g}$. This is the only adjusted parameter for deposition and erosion mechanisms applied to actinides, where four parameters were independently adjusted for actinides in the previous OSCAR version [6]: deposition and erosion rates, on core surfaces and on out-of-core surfaces.
- The erosion rate decay constant on out-of-core surfaces is adjusted to 8.10^{-4} per day and 5.10^{-4} per day for calculations n°1 and n°2, respectively.

We observe that both calculations are close to measurements for ^{134}I primary activity (Figure 3). It must be noted that no particular parameter adjustment is done from cycle 9 to cycle 15.

The ^{134}I activity decrease from one cycle to the following is due to the partial reloading of the core. After the dissemination, the ^{134}I activity increase along a cycle is due to:

- Evolution of the fissile material under neutron flux,
- Slow uranium dissolution (calculation n°2),
- Transfer from the out-of-core surfaces due to erosion/deposition mechanisms. In case of uranium dissolution, the erosion rate decay constant is lower and a higher transfer from out-of-core surfaces counterbalances the uranium dissolution on the core. Indeed, dissolved uranium is trapped on the ion exchange resins and disappears from the primary circuit.

Alpha activity in the primary coolant is due to disseminated particles during cycle 8 and to eroded particles during the following cycles. Figure 4 shows that the calculated alpha activity in the primary coolant during nominal operation is consistent with the measurement. This indicates that the erosion and deposition rates applied to actinides are at the right level with very few adjustments to the corrosion product rates.

Nevertheless, alpha activity seems to become too low after a few cycles. This could be improved by increasing the size of the particle. The deposition rate would be lower, and the particle activity higher.

Lastly, the calculated alpha surface activity on steam generator's channel head is also consistent with the measured value (within 20%). It must be noted that the calculated alpha surface activity is 40% higher on steam generator tubes, due to the smaller hydraulic diameter.

All these results show the consistency between corrosion products and actinides in particulate form in nominal operation. Regarding the uranium dissolution rate, it is difficult to assess as both calculations allow good simulation of actinides. Nevertheless, a higher dissolution rate than in calculation n° 2 could not lead to correct ^{134}I activity simulation without separate adjustment of the erosion rate for core surfaces and out-of-core surfaces. Therefore the uranium dissolution rate used in calculation n° 2 is likely a maximal value. It leads to the distribution illustrated in Figure 5 between ^{238}Pu included in uranium oxide particles (linked form) and ^{238}Pu evolving separately after uranium oxide dissolution (free form). The distribution is the same for other alpha emitters. This shows that 7 cycles after dissemination, around 33% of the disseminated particles has dissolved.

According to the simulation, the alpha activity in the primary circuit is mainly in the form of uranium oxide particles, and deposited on the surfaces. The ratio of in water to deposited alpha activity is lower than 10^{-4} during dissemination, and lower than 10^{-6} after.

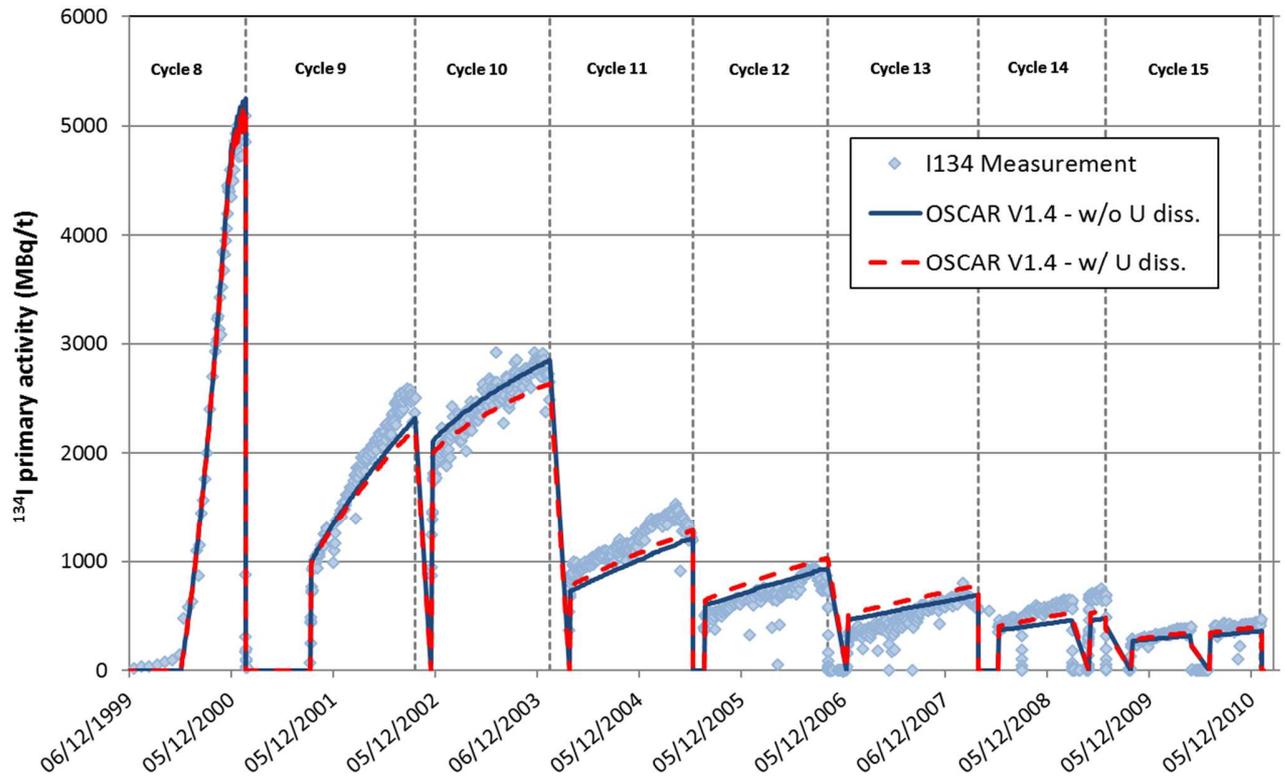


Figure 3 – Calculation/measurement comparison for ^{134}I primary activity

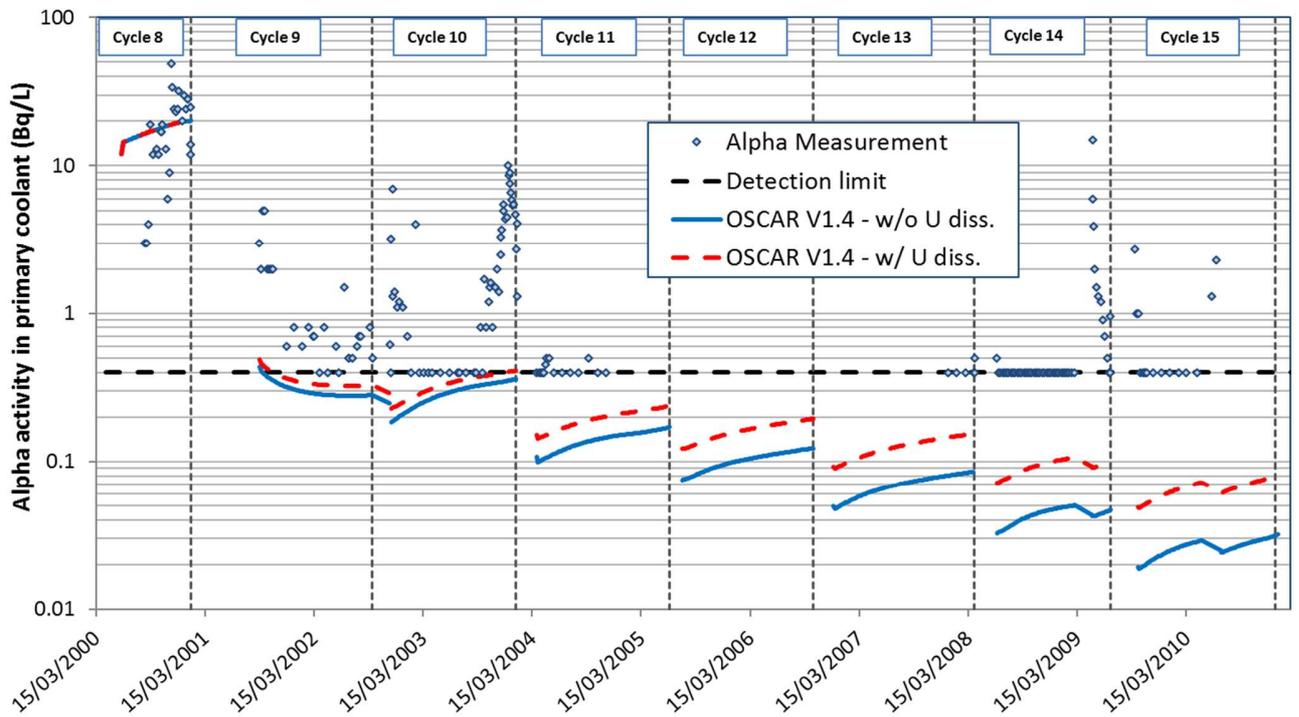


Figure 4 – Calculation/measurement comparison for alpha activity in the coolant

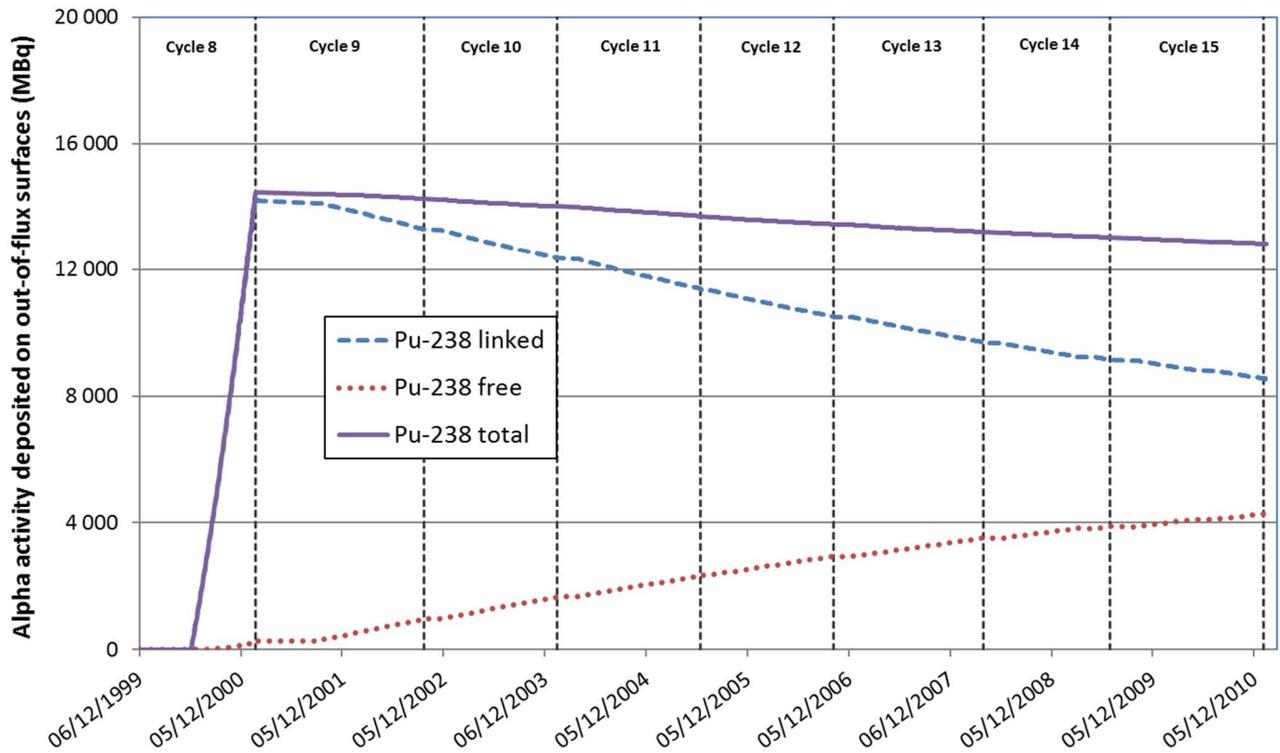


Figure 5 – Distribution between linked and free forms of ^{238}Pu alpha emitter calculated with OSCAR V1.4 (calculation n° 2)

4.3.2 During end-of-cycle shutdown

Filtering campaigns carried out during end-of-cycle shutdown [6] show that alpha activity in the coolant increases by a factor 10 to 100 after the power shutdown and remains mainly in particulate form. Moreover, a correlation between the alpha activity and the corrosion product ^{60}Co in particulate form is observed along the shutdown process.

These observations suggest that dissolution of the corrosion layer during shutdown induces the release of actinides into the coolant. However, the dissolution rate of corrosion products is maximal at the time of primary circuit oxygenation whereas it is not the case for alpha activity in the coolant. Therefore, the alpha release is not directly linked in the OSCAR modeling to the corrosion product dissolution rate but to an increased erosion rate during the shutdown process, accounting for deposit embrittlement due to corrosion layer partial dissolution.

In calculations n°1 and n°2, the erosion rate is increased during shutdown by a factor of 10 for corrosion products, and a factor of 50 for actinides. Results are presented in Figure 6 at the end of cycle 11. Activity levels for alpha emitters and ^{60}Co in particulate form are correctly simulated, as well as the correlation between alpha and ^{60}Co particulate activity along the shutdown process.

The enhanced erosion factors could have been chosen differently. Nevertheless, we can conclude that enhanced erosion during shutdown can account for increased alpha activity and for correlation between alpha emitters and corrosion products in particulate form in the primary coolant. Moreover, this enhanced erosion does not affect the radiocontaminant distribution on the different surfaces of the primary circuit: surface activity does not significantly vary during the shutdown.

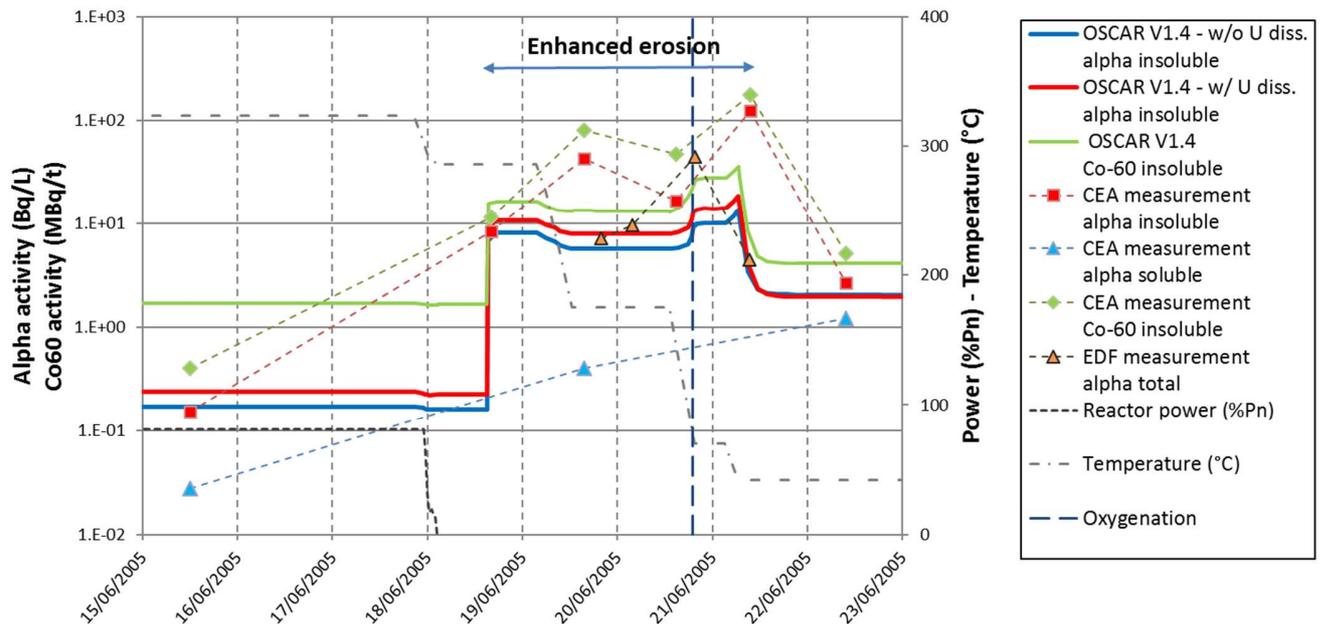


Figure 6 – Calculated and measured alpha activity in the coolant during shutdown at the end of cycle 11

4.4 Fission products released within the disseminated fissile material

In case of fuel oxide dissemination, fission products are enclosed in the disseminated particles. The whole fission product spectrum may be found in the primary circuit, and not only the gaseous and volatile fission products.

From ^{134}Cs (2 year period) and ^{137}Cs (30 year period) primary activity measurements for reactors contaminated with fissile material, we can assess that fission products stay enclosed in fuel oxide particles during several cycles. Indeed, unlike ^{137}Cs , ^{134}Cs is not created from fission but from neutron capture on ^{133}Cs , a stable nuclide coming from fissions. In nuclear fuel, ^{133}Cs and ^{137}Cs accumulate along irradiation. Therefore the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in the fuel is around zero at the beginning of irradiation but gradually increases with fuel burnup. In the primary coolant, we observe that the $^{134}\text{Cs}/^{137}\text{Cs}$ measured ratio corresponds to the burnup of the disseminated fuel during several cycles. This shows that cesium is very slowly released from disseminated fuel oxide particles. If cesium was quickly released from fuel oxide particles, there would be no more ^{133}Cs accumulation in the primary circuit and the measured $^{134}\text{Cs}/^{137}\text{Cs}$ ratio would be very low.

Consequently, it is assumed that disseminated fission products, whatever their related element, follow the same behavior than actinides, undergoing the same erosion and deposition mechanisms. Fission products are likely released from the fuel particles at the rate of the uranium oxide dissolution. This behavior is important mainly for long life fission products, as shorter life fission products quickly disappear from fuel particles. It should be considered even for gaseous nuclides like ^{85}Kr (11 year period).

4.5 Fission Products emitted by recoil from the disseminated fissile material

Fission products are emitted into the primary circuit from fissions in the fissile material contamination when it is deposited under neutron flux. Once again, the whole fission product spectrum can be released. Then each fission product evolves in the primary circuit according to the behavior of the related element. This behavior is well known only for gaseous and volatile fission products, which are soluble in the primary coolant (see § 3).

However, fission products related to other elements induce negligible radioactivity in the primary circuit compared to xenon, krypton, iodine, and cesium isotopes. Moreover, if their radioactive decay period is short enough (lower than one year), they are not a concern for waste and effluent management as they disappear rapidly. On the other hand, long life fission products in the primary circuit are mostly included in fuel oxide particles rather than emitted by fissions in the actinide contamination. Therefore they can be considered as insoluble and following the same mechanisms as actinides. As an example, ^{99}Tc , ^{93}Zr , ^{107}Pd are treated as insoluble in the OSCAR modelling. These difficult-to-measure long life fission products are important for waste and effluent management.

Lastly, a specific adsorption/desorption model [1] has been introduced in OSCAR to describe the strontium behavior. It is used to assess the ^{90}Sr (29 year period) release into the primary coolant during an end-of-cycle shutdown. This behavior is related to the pH decrease, which induces desorption of strontium trapped on the primary circuit surfaces. However, the amount of strontium involved has been directly emitted from fissions in the primary circuit, whereas strontium disseminated within fuel particles stays enclosed in these particles and does not contribute to the strontium release.

5. Conclusion

This paper shows that the OSCAR V1.4 code is a powerful tool for simulating fission product and actinide contamination in PWR primary circuits, and is helpful for fuel and waste management as well as for the alpha risk assessment. The main conclusions are as follows:

- The fuel code ALCYONE and its fission gas module MARGARET have been adapted to defective rod configuration and allows calculating the release in the primary circuit for gaseous and volatile radionuclides in case of a rod clad defect. Validation against a unique experimental database including defective PWR rods as well as analytical irradiations in a research reactor shows that the difference between calculated and measured fuel released fractions is lower than a factor of 2 for UO_2 and MOX fuel up to 50 GWd/t.
- Mechanistic erosion and deposition models have been calibrated for corrosion products and applied to actinides. For that, a specific computation scheme has been used, which allows calculating the contamination of corrosion products as well as of fission products and actinides in a PWR primary circuit with the same data set. This provides a fair simulation of actinides during nominal operation, with very few adjusted parameters. It confirms the coupled behavior of corrosion products and actinides in the primary circuit. However, the uranium dissolution rate could not be precisely assessed, as it has very little impact on alpha contamination.
- The alpha activity increase in the primary coolant during end-of-cycle shutdown is correctly simulated considering enhanced erosion, as well as the correlation between alpha activity and corrosion product ^{60}Co in particulate form along the shutdown process. Enhanced erosion accounts for deposit embrittlement due to corrosion layer partial dissolution.

The qualification of the OSCAR code should be pursued on more reactor cases to consolidate the validation domain.

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