RADIOCHEMICAL GUIDELINES AND PROCESS SPECIFICATIONS FOR REACTOR SHUTDOWN : THE EDF STRATEGY.

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ABSTRACT

Changes to French nuclear regulations made in June 2006 [1.] have made it necessary for EDF to modify its ruling principles. These modifications required the restructuring of radiochemical guidelines to better reflect their impact on nuclear safety, the environment and radioprotection.

In accordance with these aims, a new authoritative document has been produced. This ruling document identifies all parameters with a potential impact on nuclear safety, radiological releases to the environment and personnel dose rates. These diagnostic and control parameters have been identified for a reactor in production and for a reactor during shutdown.

For parameters related to a reactor in production, some indicators are used to evaluate impacts on availability, radioprotection and the environment during shutdown and on outage and to anticipate mitigation ways. On the other side, several parameters related to the stages of shutdown were also directly evaluated in order to minimize the impacts.

This paper describes the EDF methodology used to establish operational documents: radiochemical guidelines and process specifications, and includes the following:

- description of monitored parameters and their associated areas of risk;
- justification of target values, frequencies of inspection and the required actions for the monitored parameters.

The sizing methodology is based on theoretical studies and on EDF operational experience analysis.

By implementing in the operational and technical specifications requirements linked to nuclear safety, radioprotection and environment respect, EDF will benefit from an improved compromise between these areas as well as an increased focus.

1. Introduction.

From the beginning of its fleet exploitation, EDF has always used global radiochemical guidelines. Initially defined from the design technical specifications, the radiochemical guidelines were essentially dedicated to the "reactor in production" mode [2.]. Nevertheless, they have been quickly completed with expected and limit values in the other operation modes, and dedicated to the shutdown monitoring. These specific rules could be associated to radioprotection or environmental topics, even if it was not so directly indicated.

With the modification of the French nuclear regulations made in June 2006 and the publication of the Act on Transparency and Security in the Nuclear Field (TSN Act) [1.], it has been necessary to restructure the radiochemical guidelines to better reflect their impact on nuclear safety, the environment and radioprotection.

2. Organisation of the EDF Strategic Principles

To issue its authoritative documents, EDF follows a procedure with different levels of requirements depending on the applicability and on the management level of redaction. Four classes of authoritative documents are identified and elaborated for the EDF nuclear production fleet.

The first class of documents is dedicated to the strategic orientations: it defines trends to follow and the objectives to reach. It is the strategic plan for the EDF fleet.
The second class, the ruling memoranda, collect theoretical knowledge and operational feedback to define the requirements needed to achieve the objectives defined in the strategic plan.
The third class of documents identifies principles shared by these stated requirements and defines the activities and limits necessary to meet them. These documents are further elaborated upon to give the operational documents, which constitute the fourth class of authoritative documents.

The application of this methodology can be illustrated for the radiochemical guidelines (Figure 1). For example, the strategic plan, written by the Director of the Division for Nuclear Production, established the main goals concerning the EDF fleet with respect to safety, radioprotection, the environment and availability.
The ruling memorandum (class 2) for "the radiochemical measurements for a Pressurised Water Reactor (PWR) in operation", written by the Division for Nuclear Production (DPN), identifies all the parameters with a potential impact on nuclear safety, on the radiological releases in the environment and on the personnel dose rates. This ruling document quantifies the wanted results on these topics, in accordance with the new French nuclear regulations [1.]. It takes also into account the availability aim for the EDF plant fleet. This ruling document is sent to the French Nuclear Safety Authority (ASN) for information.

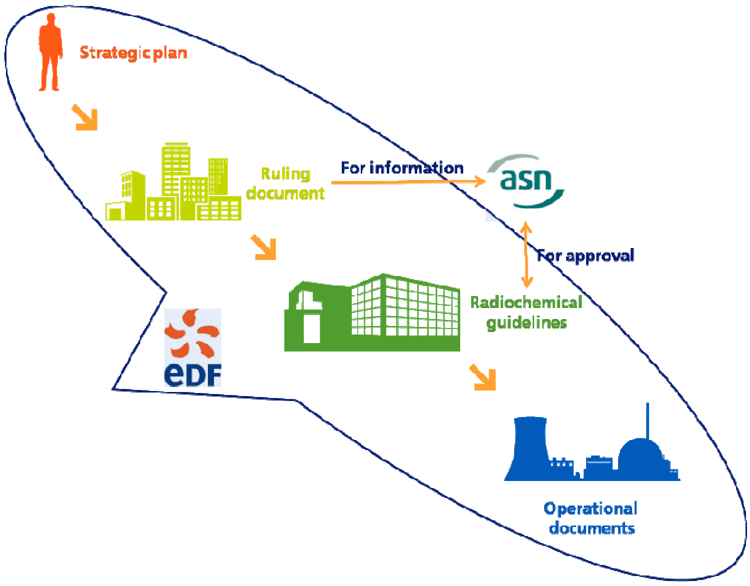

Figure 1 : EDF authoritative principles and organization.

The Division for Nuclear Engineering (DIN) is in charge to elaborate the chemical and radiochemical guidelines (documents from class 3) in accordance with the requirements and aims furnished by the ruling memorandum. All the specifications: are merged in a single document. There is no specific presentation between "safety" parameters or not, between parameters used for control or for diagnostic. Only Operating Technical Specification (OTS), linked to safety parameters are specifically identified. This document includes, for each parameter, the limit value(s), the expected value, the measurement frequency and the corrective actions associated to the limit value(s) and expected value. For some parameters, it is possible to get many limit values; for example depending on the delays for the corrective actions. For each plant series (CP0, CPY, 1300MW, N4) of the EDF NPP fleet, one document is written. These documents are then forwarded to the French regulator (ASN) for the approval of all safety-related parameters (i.e. those identified as "Technical Specification of Operation"). After having received approval, the documents are given to the Nuclear Power Plants (NPPs) for their implementation in the form of operational documents. Local requirements are taken into account during this translation. In the same time, a computer transcription is implemented in the EDF database MERLIN to help to apply these guidelines.

3. Objectives of the Radiochemical Guidelines.

In accordance with the requirements issued in the ruling memorandum, the radiochemical specifications are intended to:

- Specify to the operator the radiochemical parameters subject to periodic inspections- these inspections being performed for reasons of impact, potential or certain, and with associated times, short- or long-term
- Ensure the absence of negative trends of the radioactivity outside of expected values
- Ensure the compliance with the dimensioning assumptions design basis developed in the Safety Report
- Limit the activities of radionuclides to control discharges and radiation exposure of workers

These principles lead to the need monitor the primary, secondary and auxiliary systems in all modes and configurations of the reactor operation and thus require radiochemical measurements in order to help:

- Ensure nuclear safety,
- Ensure the integrity of the three barriers,
- Control containment for auxiliaries and intermediates circuits,
- Control the source term related to radiation protection,
- Control the source term related to the activity of the effluents.

Parameters are justified according to their impact on the following key objectives of TSN Act [1.]:

- Nuclear safety
- Radiation protection of workers
- Environmental considerations (such as effluent discharges and waste).

Event if the radiochemical specifications concern all modes and configuration of the reactor operation, this paper will focus on the radiochemical specifications dedicated to the shutdown and the outage management. It does not deal with radiochemical guidelines, which applies to "reactor in production" mode and/or associated to other objectives.

4. General Considerations Concerning Monitored Parameters.

4.1. Definitions.

A parameter is defined with consideration of the following attributes:

- The indicator; a measured or calculated variable, or combination of, which can be used to characterize the state of a fluid contained within a circuit;
- The sampling system; the system being monitored;
- The operating mode of the reactor.

Radiochemical monitored parameters on NPPs are compared to Expected Values and Limit Values, these are defined as follows:

- Expected Value; a value that is recommended not to be exceeded for the operation mode in which the specification applies. A fault is presumed when an Expected Value has been exceeded. It is desirable for corrective action to be put in place to restore, as early as possible, the previous compliant value. Generally, the Expected Value is determined by the analytical technique available to measure the parameter and its associated quantification and detection limits. It also takes into account a reasonable margin to detect a significant change of the parameter, allowing operators to initiate corrective action and minimize the risk of reaching the limit value.
- Limit Value; a value that must be respected in the operation mode in which the specification applies. Corrective action is performed within a set period of time when this value has been exceeded. It is possible that a limit value is associated with several graded thresholds according to the evolution kinetics of degradation and the reaction time needed to correct this drift. The limits take into account uncertainties from the analytical means. So, measurement results have to be directly compared to limits, regardless of accuracy.

Parameters used for radiochemical monitoring are segregated into the following two definitions:

- Control Parameters; these are associated with Limit Values, above which corrective action is normally required. They have a direct link with the objective of preventing and limiting the consequences of nuclear safety-related issues as well as other concerns: containment health, radiation protection, the environment, technical hazards, maintenance and plant availability.
- Diagnostic Parameters; these are taken into account in the overall analysis of the operation of a unit and are complementary to the control parameters. Their monitoring and, in some cases, the actions undertaken in case of non-compliance with an associated value (Expected Value and/or Limit Value) allow to anticipate the prevention and the mitigation of issues on nuclear safety-related issues. They allow

also to characterise the chemical and/or radiochemical situation, with the aim of prevention and mitigation of these issues.

4.2. Description of the Different Monitored Parameters.

Of all radionuclides in a nuclear power plant, only a few are likely to be used as indicators. The nature of the following radionuclides discourages their use as indicators:

- Radionuclides having too short a period (of the order of a few minutes) to be collected and measured
- Radionuclides with too long period (of the order of several decades) do not have generally a sufficient counting statistics to be used. Moreover for most of them, the equilibrium state is not reached to get a relevant characterisation of the situation;
- Radionuclides that have physico-chemical or nuclear properties that require long and difficult preparations (organic extraction, etc..). Radionuclides requiring metering equipment incompatible with operating configurations (e.g. for pure beta emitters) are similarly difficult to analyse.

Taking into account these aspects, the parameters, which can be monitored, are presented below according to the chemical families to which they belong.

4.2.1. Fission Products.

The fission products (FP) are normally confined within the clad of the fuel rods (first barrier). However, their presence in the primary coolant during normal operation and shutdown cannot be totally avoided due to the presence of negligible quantities of fissile nuclei on the surface of the rods. Much of the contamination of the primary circuit, with respect to fission products, originates from cracked or leaking fuel rods.

The activity in FP measured in the Reactor Coolant System (RCS) results from the accumulation of [11.]:

- The activity induced by natural uranium present as an impurity in the zirconium alloy constituting the fuel cladding or as residual contamination on the cladding during its production,
- The activity released by cracked or leaking fuel rods,
- The activity due to residual contamination of fissile materials from previous cycles,
- The activity due to fission of nuclei associated to current contamination with the release of fissile materials from disseminating fuel defects during the current cycle.

In addition, this source term in fission products can depend on the type of leaking fuel rods, the progress of the campaign, the operating power and burn-up of the leaking fuel rods.

The main FP, monitored in the primary fluid, are classified into several categories:

- Inert gases: ^{133}Xe, ^{133m}Xe, ^{135}Xe, ^{138}Xe, ^{85m}Kr, ^{85}Kr, ^{87}Kr, ^{88}Kr.
- Iodines: ^{131}I, ^{132}I, ^{133}I, ^{134}I, ^{135}I.
- Caesiums: ^{134}Cs, ^{137}Cs.
- Non-soluble fission products: ^{151}Sm, ^{89}Sr, ^{90}Sr.

4.2.2. Activated Corrosion Products.

The category of the “Activated corrosion products” designed two categories of radionuclides:

- these resulting from the direct activation of the RCS in-flux constituent materials and then degraded with release in the primary coolant;
- and all the radionuclides resulting from the activation of corrosion and erosion products, released by corrosion from out-of-flux materials into the primary fluid.

The physical and chemical changes of the primary fluid during shutdown cause usually a significant increase of the volume activity of activated corrosion products [4.]. Indeed, temperature changes lead to the dissolution of the oxides formed on the surface of the circuit components and the passage in oxidising conditions leads to oxidation and dissolution of activated metallic elements deposited on the surface of fuel cladding [4.]. These radionuclides are responsible for about 85-90% of doses integrated during outage [5.], hence the importance of controlling their behaviour during shutdown.

The corrosion product source term is very dependent, qualitatively and quantitatively, on the cleanliness of circuits, on operation modes, on maintenance operations, as well as on materials used (types of alloys, presence of stellite, control rods, seals, etc..) [4.][6.].

The main activated corrosion products are summarised below [3.]:

- ^{54}Mn, formed by neutron activation of ^{54}Fe released from metal structures.
- ^{58}Co, formed by neutron activation of ^{58}Ni, released as the main constituent of Inconel 600 and 690 alloys used for the steam generator tubes (also used in varying proportions in stainless steels).
- ^{59}Fe, formed by neutron activation of ^{58}Fe released from metal structures.
- ^{60}Co, formed by neutron activation of ^{59}Co, released as the main constituent of stellite, but also present as impurities in other metallic materials.
- ^{51}Cr, formed by neutron activation of ^{50}Cr released from structures in stainless steel and in 600 and 690 Inconel alloys.
- ^{63}Ni, formed by neutron activation of ^{62}Ni mainly released from structures with 600 and 690 Inconel alloy.
- ^{110m}Ag, formed by neutron activation of ^{109}Ag from AIC control rods (Silver-Indium-Cadmium) and the buttering of silver seals (Helicoflex).
- ^{122}Sb, formed by neutron activation of ^{121}Sb from antimony-beryllium secondary neutron sources, from impurities of zircaloy cladding and from bumpers and levels of some pumps.
- ^{124}Sb, formed by neutron activation of ^{123}Sb from antimony-beryllium secondary neutron sources, from impurities of zircaloy cladding and from bumpers and levels of some pumps.
- ^{125}Sb, formed by neutron activation of ^{124}Sb.

4.2.3. Actinides

In principle, the actinides are contained in the fuel rod cladding. However, actinides may be found in the primary coolant in case of significant leakage of fuel cladding. This is called a disseminating default and usually results in the spread of fissile material.

In the absence of cladding failures, two sources for actinide traces are identified [13.][14.]. Their existence results from the fission of fissile material located in residual deposits on the outer surface of the rod from the fuel production, but also of fissile material available as impurities present in the fuel cladding material.

In case of defective fuel rods, fissile material can be released during a campaign. Its distribution is substantially uniform between the parties in the primary circuit flows and out flows, without excluding nuclear auxiliary circuits.

Studies on the physicochemical form of alpha-emitting radionuclides in the primary fluid show that they are probably in particulate form in the operating conditions of PWR plants.

All actinides are alpha emitters, but some also emit gamma radiation. The following radionuclides do not emit gamma radiation, and can be detected and quantified only by alpha spectrometry [15.]: ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am, ^{242}Cm, ^{243}Cm and ^{244}Cm.

4.2.4. Activated Products.

The category "activation products" is used for radionuclides resulting from the activation of impurities and pollutions in the makeup water and from the activation of the primary fluid itself. They are distinguished from "activated corrosion products" that result from the activation of corrosion products released from reactor components into solution.

In a PWR, the main products of activation are:

- ^{16}N, ^{14}C from the activation of the primary circuit water;
- ^{3}H from the activation of packaging products of the primary fluid, in particular ^{10}B;
- ^{41}Ar, ^{17}N from the activation of the air dissolved in the primary fluid.

Tritium, mainly in the form of tritiated water ($^{3}H_2O$) in the primary coolant and all systems related to the RCS, can be an excellent leakage tracer. It is easy to measure and shows no sampling bias (present as water, it presents no problem of deposit, absorption, or trapping, as other radionuclides in aqueous samples).

5. Description of a Typical EDF Reactor Shutdown.

Shutdown and start-up transients are associated with significant changes in physicochemical conditions of the primary fluid. In the initial state, when the reactor is under production, the primary coolant is hot and under basic and reducing conditions. On the contrary, at the final state of the shutdown procedure, the primary fluid is cold and under acid and oxidizing conditions. During the start-up procedure, the contrary modifications occur.

These changes may result in a number of risks which have to be controlled, including the following ones:

- H_2/O_2 dangerous mixtures: ignition, detonation;
- Integrity of materials: corrosion;
- Radiation protection: exposure of workers during the outage;
- Environment: control of effluents and emissions;
- Plant Availability: extended outages.

Thus, in terms of radiochemical aspects, the main objective of the procedure for cold shutdown is to reconcile the requirements on radiation protection, discharges limitation and outage duration.

Concerning a chemistry and radiochemistry point of view, the main steps of a typical EDF reactor shutdown can be summarised as follows [7.], from the mode Reactor in Production (RP) to the mode Reactor Completely Discharged (RCD):

1) Preparation of the power setback and shutdown (RP)
2) Reduction of power- Hot standing
3) Borification in hot standing / shutdown (NS / SG)
4) Cooling and depressurisation
5) Connection of the RHR sytem and bubble collapse (NS / RHR)
6) Cooling, oxygenation and purification of primary coolant
7) Last RCP is secured and the RCS is depressurised (MCS)
8) RCS opening
9) Draining of the RCS, pressure vessel head lift and filling of the reactor cavity
10) Fuel handling (RCS)
11) Draining of the RCS and bottom of RCS pipes transition (RCD)

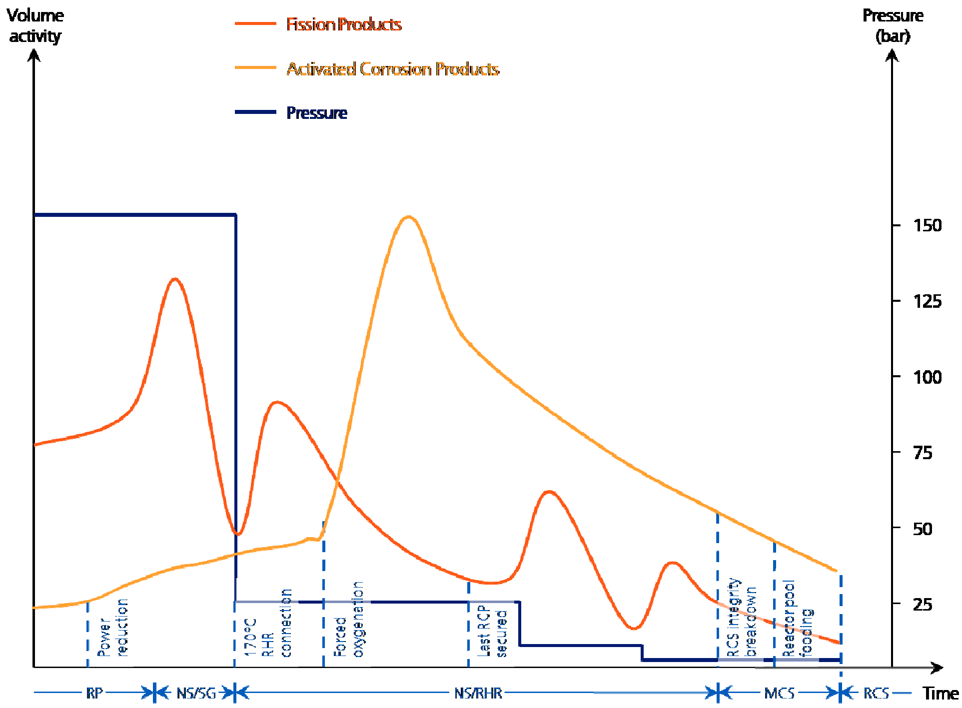

Figure 2 : Example of volume activity for fission and activated corrosion products during shtudown.

Shutdown transients are accompanied by significant modifications in physical and chemical conditions of the primary fluid. From the viewpoint of radiochemistry, the following sketch (Figure 2) shows an example of evolutions of the fission and activated corrosion products.

5.1. Behavior of Activated Corrosion Products.

By performing the oxygenation of the primary fluid, the operators try to control the modification of the primary fluid from reducing condition (under hydrogen) to oxidizing one (oxygenated coolant). To achieve this objective, it is necessary to control the solubilization of the activated corrosion products, to ensure their purification at the highest rate and to assure the absence of hydrogen in the RCS for its integrity breakdown. The procedure of forced oxygenation is currently performed on EDF fleet by injecting H_2O_2 at 80℃.

The procedure for cold shutdown with forced oxygenation is a compromise between radioprotection and the duration of outages. The advantages of forced oxygenation are exposed below:

- Avoidance of the risks associated with hydrogen-oxygen mixtures everywhere to preserve the integrity of the second and third barriers
- Control of the dissolution of activated corrosion products, as well as the release of activated materials
- Reduction of the activity of the coolant as quickly as possible
- Avoidance of the contamination of circuits by deposition of activation products or solid fission products, or by adsorption of molecular iodine
- More efficient fuel handling operations due to an improved water clarity and by a reduction in the dose rates at the surface of the pool
- Dose rate reduction for maintenance operations
- Avoidance of exceeding effluent regulatory limits for exhaust stack releases
- Minimisation of the volume of gaseous and liquid discharges.

Decontamination of circuits achieved via the solubilisation of out-of-flux deposits is impossible because most of the volume activity results from the dissolution of deposits up to the fuel clad [4.]. Many measurements of the surface activity (by using the EMECC devices [19.]), have demonstrate that any modification of the deposited activity on out-of-flux pipes occurs during the forced oxygenation. The risk of redeposition is reduced via the avoidance of temperature increases during the cooldown period. Moreover, following the forced oxygenation, primary coolant has to be kept sufficiently oxidizing until the pressure vessel head lift. Regular monitoring of dissolved oxygen concentration in the primary fluid is thus performed to prevent its decrease below a limit value.
For hydrogen, the objective is to control the release of corrosion products and to prevent the risk of a dangerous mixture with oxygen. The idea is to remove enough hydrogen before oxygenation, to achieve a concentration below the threshold defined to avoid the explosive mixture. The removal may be performed either by acting directly in the gaseous phases of the capacity (venting/nitrogen sweeping), or by acting on the hydrogen dissolved in the primary fluid (chemical degassing), or both in parallel. The thresholds to meet specifications are indicated in the Chemical Guidelines.

On the other hand, it may interesting to keep a dissolved hydrogen content ("heel") at lower temperature, to maintain a reducing environment and to control the solubilization of out-of-flux deposits. This approach requires that to keep the ability to removal hydrogen quickly and with control, until the oxygenation, for example by using chemical hydrogen degassing.

5.2. Behavior of Fission Products.

Throughout the different steps of a shutdown, the evolution of fission products is regularly monitored by gamma spectrometry measurements of the primary fluid.

The selected indicators are ^{133}Xe and ^{131}I. ^{133}Xe is chosen as it is the main contributor to the total noble gas activity and ^{131}I is selected because it has a long half-life and a high radiotoxicity. The main radiochemical objectives during the shutdown are the limititation of radioactive gaseous discharges to the environment at the time of oxygenation and the limitation of risk of air contamination in the reactor building during the opening of the primary circuit.

Thermohydraulic modifications occur in the primary circuit during the power setback, and then, during various pressure or temperature decreases. These modifications facilitate the release of fission products from the pellet-cladding gap and increase the activity of fission products in primary coolant in case of a leak of the fuel clad (Figure 2).

Noble gases are chemically inert and can be eliminated only by physical degassing of the primary fluid. Iodine speciation is characterized by a complex dependence to the chemistry of the primary coolant [16.][17.]. Studies have shown that various forms of iodine can exist in the primary fluid: ionic or molecular ones, soluble or not. The ionic species can be retained by the ion exchange resins, whereas the molecular iodine can be adsorbed onto the walls of the primary circuit and desorbed at the air sweeping of the surfaces.

6. Shutdown and Outage Preparation.

Through the monitoring of the integrity of fuel cladding, the radiochemical guidelines establish a strategy in order to limit the consequences during shutdown and outage of the fission products presence. The objective is to help to organize the outage by adapting the actions to perform the outage with performance depending on the potential risks associated to fission products presence in the primary coolant.

The monitoring strategy is mainly based on an early detection of fuel failure, completed with a graduated strengthening of the monitoring of parameters associated to the consequences and with adapted management strategies as soon as the activities reach a defined threshold.

In accordance with these objectives, the main parameters associated with the shutdown and outage preparation are the following ones:

- ✓ Characterisation of the leaking fuel assembly: ratio $^{134}Cs/^{137}Cs$ and $^{135}Xe/^{85m}Kr$.
- ✓ Behavior of fission products released from the leaking fuel rod during shutdown: ^{133}Xe and ^{131}I volume activity, reactor in production but after power transition.
- ✓ Risk of workers exposure to iodine: ^{131}I volume activity for a reactor in production, in normal operation.
- ✓ Risk of workers exposure to α emitters: ^{134}I volume activity reactor in production, in normal operation.

6.1. Shutdown Preparation.

Reactor in production, criteria are defined on the volume activity of the primary coolant to detect fuel defect. If these criteria are met, French regulation requires the leaking fuel rod(s) are identified and removed during the next outage. Refueling with a leaking rod is forbidden.

There are multiple reasons why leaking rod(s) should be identified as quickly as possible with respect to shutdown preparations at EDF:

- The need for the anticipation of new fuel loading studies, if necessary;
- The need to plan and forecast operations during the shutdown and schedule the necessary phases of degassing, purification, and sipping tests.

6.1.1. Characterization of the Leaking Fuel Assembly [11.]

The two main criterias used to characterise a fuel assembly with an associated leaking fuel rod are the burn-up and the type of the fuel (UOx or MOX).

Burn-up Estimation.

The estimation of the burn-up of the failed fuel is based on the determination of changes in the concentration of appropriate fission products. The radionuclides with the best nuclear characteristics (period, fission yield) to determine the burn-up of the leaking rod are ^{137}Cs and ^{134}Cs. The determination of the ratio $^{134}Cs/^{137}Cs$ is internationally recognized as an effective indicator,for activity levels, of each isotope, below 30 MBq/t.

In simple cases of single defect or multiple defects but with similar characteristics, the ratio of caesium can therefore determine the burn-up of the leaking fuel rod(s) and thus guide the analysis to a type and an expected number of defects. It is as such a diagnostic parameter. In cases of multiple defects with very different characteristics, the interpretation of the caesium ratio is not so reliable.

Type of Fuel.

The fission yields of the main fission products released from a fuel pellet are not highly dependent on the original fissile nucleus (^{235}U or ^{239}Pu), except for krypton isotopes. Indeed, the yield of fission isotopes kryptons is lower in the case of thermal fission with an isotope of plutonium than with an isotope of uranium. So there is less ^{85m}Kr products in MOX fuel than in UO_2 fuel. However, in the case of ^{135}Xe, the fission yields are essentially equivalent. Accordingly, the activity ratio of $^{135}Xe/^{85m}Kr$ (isotopes with compatible periods) is higher in the case of a MOx Than in a UOX. Under the assumption that activities in the primary coolant are comparable with the activities in the fuel, it is possible to discriminate defects from UO_2 assemblies from those from MOX assemblies by comparing the ratio resulting from the activities of ^{135}Xe and ^{85m}Kr in the primary fluid.

6.1.2. Behaviour of fission products released from the leaking assembly during shutdown

The monitoring of the behaviour of the fission products during power transients, reactor in production, contributes to the diagnostic of the fuel clad integrity. It is also a good way to access to interesting information concerning the characteristics of the leaking fuel assembly. This information may also be used to prognostic some trends on the fission products releases during the reactor shutdown. Such evaluation can be helpful to anticipate and adapt the shutdown strategy.

The objective of the measurements performed upstream of the shutdown transient is to anticipate the meeting of radiochemical criteria that punctuate the course of the shutdown. By monitoring the activity of the primary fluid, it is possible to adjust the shutdown strategy.

The Expected Values, set for ^{133}Xe and ^{131}I (without flow rate purification correction) upstream of the transient, help to obtain the oxygenation criteria.

6.2. Outage Organisation.

Reactor in production, one of the goals of the specification is the radiochemical monitoring of the degradation of the primary barrier i.e. the fuel rod cladding. The consequences of loss of water tightness of the primary barrier may be a release of gaseous fission products contained in the pellet-clad space or, for significant breaks, the penetration of liquid water in the pellet-clad gap. This can then erode the fuel, causing the release of solid particles of fuel in the primary circuit. This dissemination results in a risk of contamination by alpha-emitting particles and should thus be controlled.

6.2.1. Risk of workers exposure to iodine

To anticipate the risk of iodine presence in the circuits during the outage, the level of ^{131}I volume activity, reactor in production, is carefully monitored. There is no limit value associated to this risk in the radiochemical guidelines. Nevertheless, the stable level of ^{131}I activity is used as diagnostic parameter to plan and implement specific dispositions during maintenance operations to protect workers against iodine ingestion. As these dispositions can have a strong impact on outage organization and duration, it is important to anticipate as soon as possible.

6.2.2. Risk of workers exposure to Alpha emitters

The evolution of volume activity in ^{134}I is a sensitive indicator of the release of fissile material into the primary circuit resulting from major defects of the fuel rods. Control of this parameter also provides an indirect image of the residual contamination of circuits with alpha emitters. A quantitative link both parameters is very difficult to establish. Nevertheless, it is used to detect any significant spread of fissile material (along with actinides) which could lead, during maintenance operations performed on the circuits, to a potential risk of internal exposure of workers on work places. It is thus linked to site management and radiation protection specific actions about the risk of internal exposure to alpha emitters. It is therefore relevant to radiation protection and it is a decision parameter.

The assessment of dissemination is based on the short period of this radionuclide (53 minutes). Indeed, only very little ^{134}I quantities can be released from the cladding defects. Most of the activity of this isotope is so assumed to come from fissions of fuel material directly in contact with the water in the primary after dissemination.

Thus, this parameter provides an indirect picture of the residual alpha contamination of primary and auxiliaries circuits. Thresholds are set so as to keep the circuits in a state compatible with alpha radiological maintenance operations. The shutdown threshold, reactor in production, is set with the objective to limit reasonably the spread of fissile material in the primary fluid. Beyond this limit, the specific actions, that could be implemented during maintenance operations to protect workers, are considered incompatible with a simple, safe and sturdy management of the outage.

In addition to measurements of volume activity for ^{134}I, and of solid fission products (^{140}La, ^{140}Ba, etc.. or ^{239}Np), gross Alpha measurements are also performed to detect for the presence of a leaking fuel undergoing changes. As such, it constitutes a diagnostic parameter of the spread of fissile material.
The Gross Alpha activity is a diagnostic parameter also used in the radioprotection guidelines. Depending on the level of ^{134}I volume activity reached during the production campaign, the campaign can be identified as with "potential alpha risk". Such a classification implies a specific organization of work places and appropriate dispositions for monitoring and for implementing dedicated protection of workers against internal exposure. These dispositions can have a strong impact on outage organization and duration.

7. Shutdown Monitoring.

7.1. Mitigation of the Consequences of a Fuel Clad Defect

7.1.1. Anticipation of Later Criteria.

During power setback, a monitoring is necessary to adjust the shutdown strategy. The compliance with the expected values on the fission products activities help to anticipate the obtaining of the criteria for oxygenation and RCS opening. During this phase, the degassing ways available in RCS are various and more effective before the pressure drop. For example, the degassing by the gas phase of the pressurizer to BRS or the possibility of storage for decay via the GWTS are not available anymore after oxygenation.
The following criteria are diagnostic parameters.

Measurements at NS/SG Operation Mode.
For reactors with a high level of xenon activity, increases of the ^{133}Xe volume activity may be observed during the shutdown transient and then for the successive depressurizations of the primary circuit.
The prediction of the evolution of the ^{133}Xe activity is difficult until oxygenation because it depends, among other things, on the size, on the number and on the type of defects present in the core. Before the shutdown, it is nevertheless recommended to aim for a value equal to twice the limit value before oxygenation. This value, initially arbitrarily defined, has been confirmed by operational feedback: if this value is met, most of the time, there are no particular problems to meet the criterion for oxygenation (except in special cases related to the size and type of defects).
The expected value is a recommendation and should be considered as a way to stay focused on the following criteria on xenon activity for oxygenation of the primary coolant and for the opening of the RCS.
The compliance with this expected value determines the availability of degassing methods of the primary circuit to implement to meet the activities required to perform the oxygenation.

Concerning iodine, its speciation is characterised by a complex dependence on the environment. The elimination of iodine is favored by using a suitable environment (reducing conditions and high pH) before oxygenation and an associated maximum purification with respect to speed and efficiency.
Under the conditions corresponding to the oxygenated state of the primary fluid (after oxygenation), iodine is found preferentially in its molecular form (I_2). Many studies have confirmed that a large proportion of iodine in solution before oxygenation could be adsorbed on metal surfaces after oxygenation. Iodine, in its soluble form, can be eliminated only with ion exchange resins. The volume activity during stable operation results from a balance between the source term (depending on the condition of the cladding) and the elimination of activity through purification and through radioactive decay. A high purification rate and regularly monitored, highly efficient, resin beds are employed to thus aid the reduction of the ^{131}I activity. The expected value is a recommendation and should be considered as a way to stay focused on the following criteria on iodine activity for oxygenation of the primary coolant and for the opening of the RCS.
To keep available the degassing ways of the primary circuit, it is necessary to comply with this expected value, implemented to meet the activities required to performed oxygenation.
These criteria are often met more easily than those relating to ^{133}Xe.

To secure last RCP.
To meet easily the criteria on fission products for the RCS opening, limit values are defined to secure the last RCP. These are based on the most efficient purification that takes place on a homogeneous fluid with at least one primary pump kept in service.

7.1.2. Oxygenation of the Reactor Cooling System.

^{133}Xe Criteria.
The definition of the limit value associated with oxygenation of the primary fluid is related to a problem of radioactive discharges to the environment. Indeed, after oxygenation of the primary coolant, it is no longer possible to use the hydrogenated effluent storage decay capacity from GWTS. Indeed the gaseous phase of the CVCS tank normally oriented to the Gaseous Waste Treatment System (GWTS) is swung to the Nuclear Auxiliary Building Ventilation System (NABVS). The NABVS is equipped with "iodine filters" in the form of a spool piece. Noble gases present in the CVCS are then discharged directly to the chimney of the NAB. As soon as the gaseous phase of the CVCS tank is connected to the exhaust stack, the releases of radioactive gases have to comply with the regulatory requirements about flow activity of radioactive gases and hence with the threshold of the PRMS nuclear measuring channel chain.

To control releases of noble gases in the chimney, oxygenation cannot be induced until the ^{133}Xe volume activity of the primary fluid is reduced below a limit value. This limit value corresponds to the pre-alarm for the threshold of PRMS nuclear measuring channel chain. To determine the limit value, the balance of ^{133}Xe activity is calculated between the liquid phase of the CVCS tank and the discharge stack. This is defined as a control parameter.

^{131}I Criteria
As for ^{133}Xe, the definition of the limit value for ^{131}I associated with oxygenation of the primary fluid is also related to a problem of radioactive discharges to the environment. The iodine present in the gaseous phase of the CVCS tank is discharged via the same path as for the noble gases. Even if the NABVS can be equipped with "iodine filters", the sizing of the limit value, as for the noble gases, is aimed to meet the regulatory requirements related to discharges of radioactive iodine.

Moreover, as it has been already explained, the iodine speciation is strongly correlated to the physical chemistry of the environment. The elimination of iodine is favored by a soluble speciation available before oxygenation, since iodine is found preferentially in molecular form (I_2) after oxygenation. It is therefore important to reduce as much as reasonably possible the ^{131}I activity before inducing oxygenation. It may also be interesting to monitor the activity in ^{131}I during the post-oxygenation purification phase to detect any sudden "fall" of activity that may be indicative of a significant adsorption of iodine on the surfaces of the circuits. This is defined as a control parameter.

Recommendations

The achievement of these criteria on the ^{133}Xe and ^{131}I activities will be promoted before the connection of the RHR Indeed, there is a risk of increase of fission products activities following the collapse of the bubble if the pressuriser gas phase has not been enough diluted by circulation of dinitrogen. In addition, the degassing of the primary fluid is more effective in biphasic conditions, since the degassing can be performed by using the pressuriser.

7.1.3. RCS Opening.

The EDF current operating conditions allow three possible configurations for the RCS opening. The integrity may be broken (Figure 3) at the following locations:

- The manway of the pressuriser with water filled until the middle level of the pressuriser (configuration 1);
- The manway of the pressuriser with water filled until the vessel mating surface (configuration 2);
- The direct pressure vessel head lift, with water filled until the vessel mating surface (configuration 3).

For each of these configurations, the volumes of gas that may be degassed in the reactor building, are different and have to be taken into account in the definition of the criteria.

^{133}Xe Criteria.

When the integrity of the primary circuit is broken, the reactor building ventilation is performed with the CSV system. The volume activity in the atmosphere should not exceed a limit, which could be responsible for achieving the pre-alarm for the threshold of PRMS nuclear measuring channel chain. Such excess would lead to close the reactor building after isolation of the CSVS. It is really important since, after such isolation, the atmosphere is no longer stirred and homogenized, and therefore the evacuation for workers non-equipped with respiratory system is required.

During the breakdown of the primary integrity, xenon present in the gas phase is diluted in the air of the reactor building. The kinetics of the phenomenon and the transfer mode are not well known. That is why, conservatively, the entire content of xenon in the gas phase of the primary circuit is assumed to be instantly diluted into the entire volume of the reactor building. Depending on the configuration and on the plant design, the volume of the gaseous phase will be changed (Figure 3).

On one hand, it is easier to perform measurements in the water phase. On the other hand, the measure in the gas phase is relevant only if an equilibrium is established between liquid and gaseous phases. That is why, only a limit value on the volume activity in the liquid phase is retained at EDF. Balance between aqueous and gaseous phase is thus calculated to get the limit value in the primary coolant. Depending on the version of the radiochemical guidelines, a total degassing or equilibrium between both phases is assumed to establish the balance.
^{133}Xe volume activity in primary coolant for RCS opening is a control parameter.

^{131}I criteria.

When the integrity of the primary circuit is broken, the ^{131}I volume activity in the reactor building shall not exceed a level that would lead to the sounding of the alarm of iodine mobile radiation monitor installed in the reactor building.

With a methodology comparable to that for xenon, the limit value in the primary coolant is calculated by balancing between the aqueous phase and the gaseous phase in the RCS in accordance with the different configuration for the RCS opening and then between the gaseous volume, which will been diluted in the air of the reactor building.

Nevertheless, for iodine, it is not possible, as for xenon, to assume a total degassing for the soluble iodine from aqueous phase of the primary circuit to gaseous one, before breaking the integrity of the primary circuit. Indeed, the chemical nature of this specie leads, unlike the case of a noble gas such as xenon, to distinguish the various chemical forms of iodine in solution [16.][17.]. It is therefore necessary to make the additional assumption that the degassing of iodine is only partial. It is essential to make the application with a value of the partition coefficient which is both realistic and conservative with respect to the phenomena and physicochemical conditions that exist in the primary circuit of EDF's PWRs. For iodine, the evaluation of the partition coefficient is difficult to consider. At EDF, a value of 10^{-3} is assumed for the partition coefficient of iodine in the shutdown conditions.
^{131}I volume activity in primary coolant for RCS opening is a control parameter.

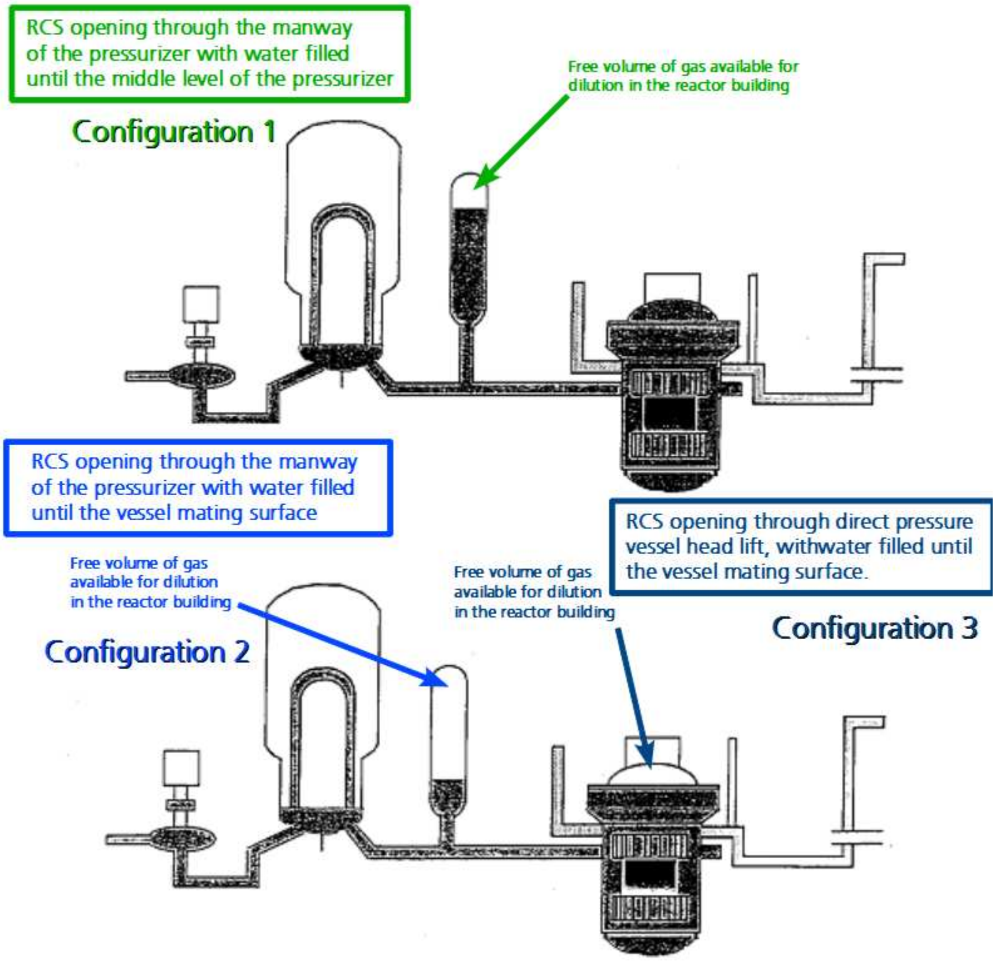

Figure 3 : Different configurations for the RCS opening.

7.2. Control of Activated Corrosion Products.

When the outage requires the RCS opening (fuel refueling outage particularly), the oxygenation of the primary fluid is inevitable. For a good control of the effects of the oxygenation on the radiation field, it is recommend to perfom a forced oxygenation of the primary fluid [8.][9.]. This oxygenation often induces a sudden increase in the volume activity (Figure 2).

The monitoring of the activity of activated corrosion products in primary coolant is conducted throughout the different phases of the shutdown:

- During the cooldown and until the oxygenation of the primary fluid
- Throughout the peaking of activities resulting from the forced oxygenation
- From the peak at the end of the oxygenation until the filling of the reactor cavity

The effectiveness of the purification treatment, through the CVCS, is also monitored to ensure the efficiency and purification rate is maintained as high as possible [10.].

All the following criteria are defined considering that, for cold outage with refilling of the reactor cavity, it is necessary to minimise the volume activity of the reactor pool in order to limit dose rates during fuel handling operations.

7.2.1. To Secure the Last RCP.

After securing the last primary pump, it is conservatively assumed that the activity in crossover legs and in the SG shells is no longer purified. The resulting "trapped" activity is potentially mixed with the activity of the Reactor Cavity Treatment System Tank and with the activity of the Reactor Cavity after going through the bottom of the primary pipes. This "trapped" activity determines partially the collective and individual integrated doses for maintenance operations around the water circuits and the reactor cavity.

The volume activity of the reactor cavity after SG draining and following the mixture, results from:

- The volume activity remaining in the primary circuit after purification (pressure vessel and RHRS)
- The volume activity trapped in the crossover legs and SG tubes after securing the last RCP
- The volume activity provided by the Reactor Cavity Treatment System Tank (possibly negligible)

The balance of volume activity is established between these three volumes. The volume activity of the reactor cavity resulting from SG draining and following the mixture has to be combinable with the later limit values required for flooding the reactor cavity after mixing the previous described volumes. This calculation allows to define the limit value required to secure the last RCP and associated to the volume activity trapped in the crossover legs and SG tubes.

Limit values are applied to ^{58}Co and gross gamma activities since ^{58}Co is assumed to be the more important contributor to dose rates and to take into account the ^{58}Co contribution to the gross gamma measurement (1Bq of ^{58}Co is equivalent to 1,6 Bq of gross gamma).

For the evaluations of radioactive releases in case of accident with loss of RHRS in cold shutdown, the criterion on ^{58}Co activity to secure the last RCP is a decoupling hypothesis. This is indeed an input data to the evaluation of released activities from the reactor vessel in case of such accident.

Both uses (dose rates control and input data for evaluations of radioactive releases in case of accident) of criterion on ^{58}Co volume activity defined itself as a control parameter.

7.2.2. To Flood the Refueling Cavity.

The objective of the following criteria is to limit, in advance, (just before the filling) the volume activity of the reactor cavity. To achieve this objective, limit values on ^{58}Co and gross gamma activities are set for the primary fluid, considering the resulting dose rates evaluated by design code calculations.

Requirements (Limit Values) and recommendations (Expected Values) have been established on the ^{58}Co which is, for now, the most abundant element. The specification for gross gamma activity is twice that for ^{58}Co activity, to take into account the possible presence of other radionuclides such as silver or antimony. These two thresholds can prevent an excessive activity in the reactor pool.
It is a control parameter.

7.3. Control of activated products: tritium.

Considering the involvement of workers in the reactor building during outage with a filled reactor cavity, the contamination of ambient air with tritium have to be sufficiently limited to minimize the integrated doses and to reduce the potential discharges of tritium through the sweeping ventilation of the building [18.].

Given the free volume in the reactor (or fuel) building and air flow recirculation, a homogeneous steady state is quickly reached with regards to tritium (after a few hours) between ambient atmosphere and the pools fluid. Thus the activity of tritium in the atmosphere depends only on the activity of reactor (or fuel) pool water, the humidity and the difference between the ambient temperature of the atmosphere of the reactor (or fuel) building and the temperature of the pool.

During shutdown and outage, the water exchanges between RCS, auxiliaries, reactor pool and spent fuel pool allow the spread of the primary coolant tritium into these pools after the opening of the pressure vessel. The evaporation from reactor and spent fuel pools is a significant source of tritium gas. It is thus necessary to prevent spread during outages to ensure the respect of regulatory limits concerning gaseous tritium discharges. Established from the operational feedback from EDF fleet, the respect of the expected value must maintain the tritium inventory low and relatively constant in the RCT tank and the spent fuel pool.

8. Conclusion.

Changes to French nuclear regulations made have made it necessary for EDF to restructure its radiochemical guidelines to better reflect their impact on nuclear safety, the environment and radioprotection.

To illustrate the methodology used at EDF to elaborate its radiochemical guidelines, the strategic radiochemical parameters used to plan and to monitor the shutdowns and outages of its reactors have been described. It is mainly based on the identification of all the parameters with a potential impact on nuclear safety, radiological releases to the environment and/or personnel dose rates. These diagnostic and control parameters have been identified for a reactor in production and for a reactor during shutdown.

By implementing in the operational and technical specifications (Table 1), all requirements linked to nuclear safety, radioprotection and environment respect, EDF benefits from an improved compromise between these areas as well as an increased focus.

Reactor in production	Parameter with criteria	Parameter without criteria	Objective
Characterization of leaking assembly	$^{134}Cs/^{137}Cs$		BU evaluation
	$^{135}Xe/^{85m}Kr$		Identification of fuel type
Behavior of fission productions released from leaking rod		^{133}Xe - ^{131}I	
Outage organisation	^{131}I		Evaluation of the risk of iodine exposure
	^{134}I		Evaluation of the risk of Alpha emitters exposure
Shutdown step	**Parameter with criteria**		**Objective**
Power setback	^{133}Xe - ^{131}I		
Oxygenation of RCS	^{133}Xe - ^{131}I		Control of gaseous discharges
Secured last RCP	^{58}Co - γ_{global}		Control of dose rates
	^{133}Xe - ^{131}I		
RCS opening			
Config 1	^{133}Xe - ^{131}I		Control of the radiological exposure to fission products
Config 2 or Config 3	^{133}Xe - ^{131}I		
Pressure vessel head lift	^{133}Xe - ^{131}I		
Filling of refueling pool	^{58}Co - γ_{global}		Control of dose rates
	^{3}H		Control of dose rates & gaseous discharges

Table 1 : Synthesis of the radiochemical parameters implemented in the radiochemical guidelines and dedicated to the shutdown and outage monitoring.

9. References.

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Radiochemical guidelines and process specifications for reactor shutdown

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The EDF Strategy

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NPC2012, Paris, September 24

The French regulatory context

- June 2006 : publication of the Act on « *Transparency and Security in the Nuclear Field* » (TSN Act).

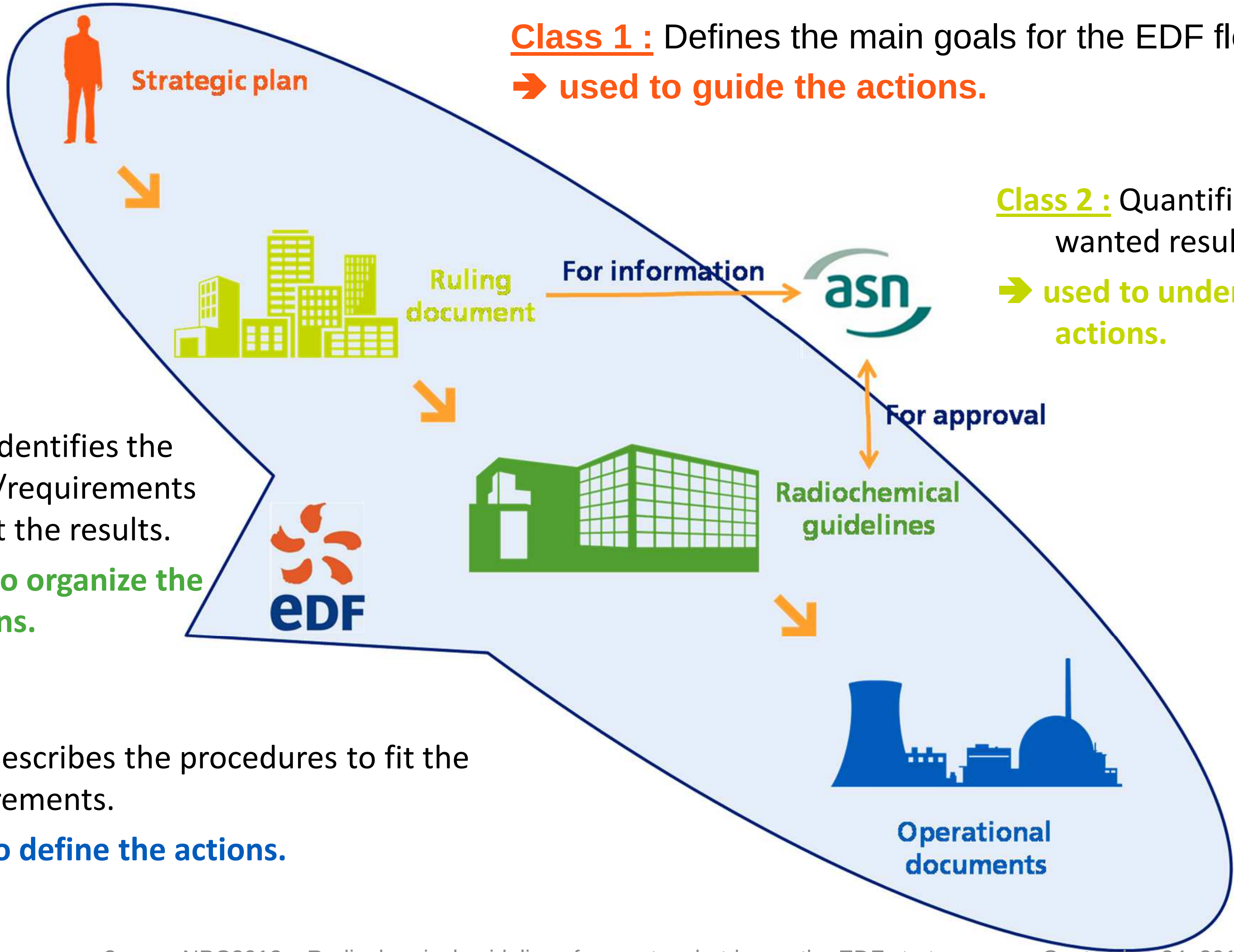
↳ Modification of the prerogative of the French Safety Authority.

  - ✓ Nuclear safety
  - ✓ Environment
  - ✓ Radioprotection

- Necessity for EDF to restructure its ruling documents.

↳ To take into account all impacts of the operational procedures on the key objectives of the TSN Act.

Organisation of the EDF strategic principles



Class 1 : Defines the main goals for the EDF fleet.

➔ **used to guide the actions.**

Class 2 : Quantifies the wanted results.

➔ **used to understand the actions.**

Class 3 : Identifies the ways/requirements to get the results.

➔ **used to organize the actions.**

Class 4 : Describes the procedures to fit the requirements.

➔ **used to define the actions.**

Objectives of the radiochemical guidelines

- Radiochemical specifications are intended to:
  - ✓ Ensure the compliance with the dimensioning assumptions design basis developed in the Safety Report, in accordance with the normal operation limits (incidental & accidental operation are not considered).
    ➔ Nuclear safety
  - ✓ Limit the volume activities to control discharges.
    ➔ Environment
  - ✓ Limit the volume activities to control radiation exposure of workers.
    ➔ Radioprotection
  - ✓ Optimize the reactor operation **➔ Availability**
- That requires to specify :
  - ✓ The radiochemical parameters subject to periodic measurements,
  - ✓ The expected and limit values associated to those parameters,
  - ✓ The mitigation and corrective actions;
  - ✓ In primary, secondary and auxiliary systems,
  - ✓ In all modes and configurations of the reactor normal operation.

Monitored Parameters (1)

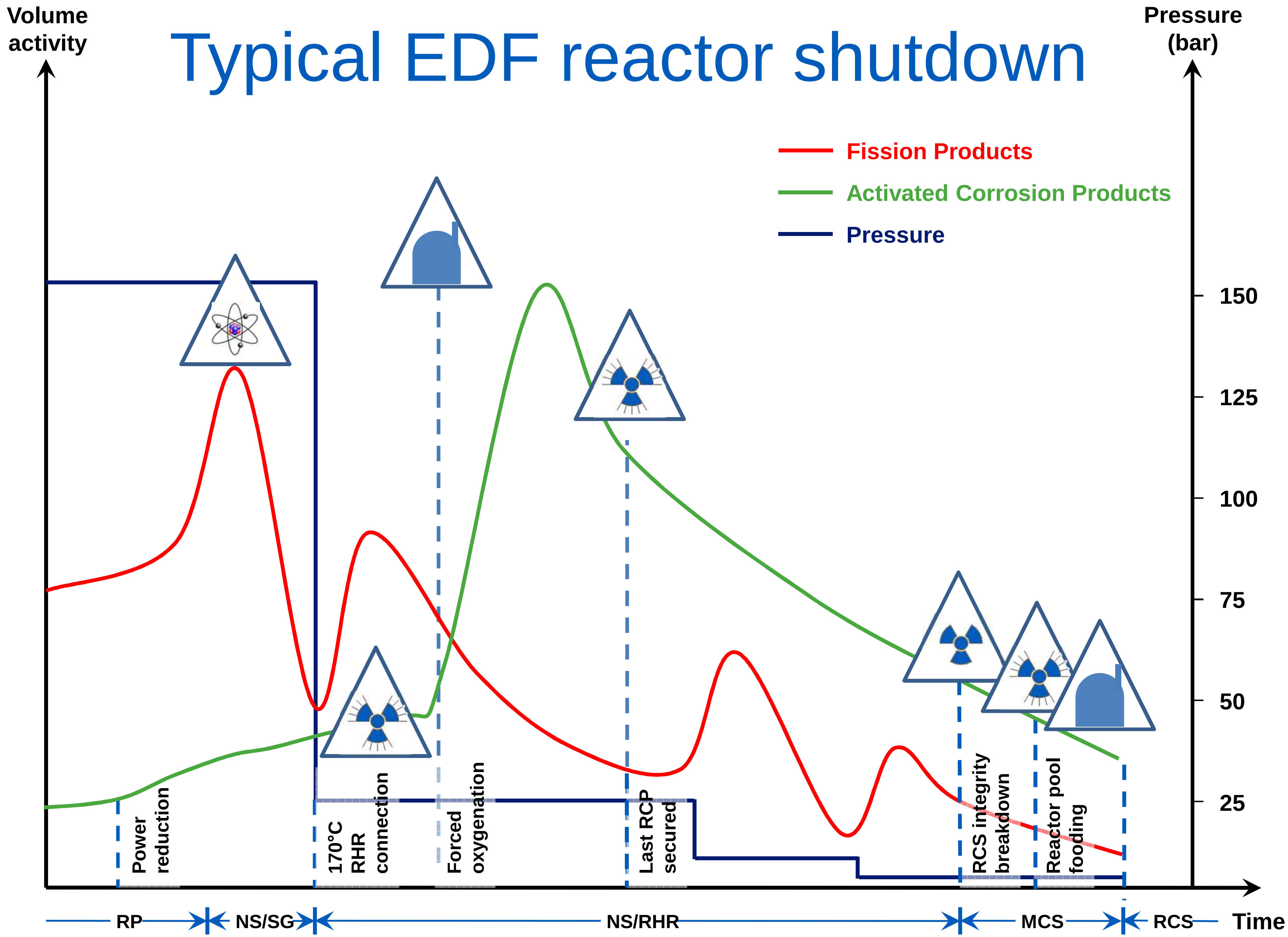
- Parameter is defined by :
  - ✓ A measured or calculated variable.
  - ✓ A sampling system.
  - ✓ The operating mode of the reactor.

- Parameters classification :
  - ✓ Control parameters : ➔ Directly related to the objectives.
    - ➢ Limits values.
    - ➢ Corrective actions or compliance required to go on the shutdown procedure.
  - ✓ Diagnostic parameters : ➔ Contribute to the overall analysis.
    - ➢ Limit and/or expected values.
    - ➢ To characterize the chemical and/or radiochemical situation.
    - ➢ To anticipate the prevention and the mitigation of issues.

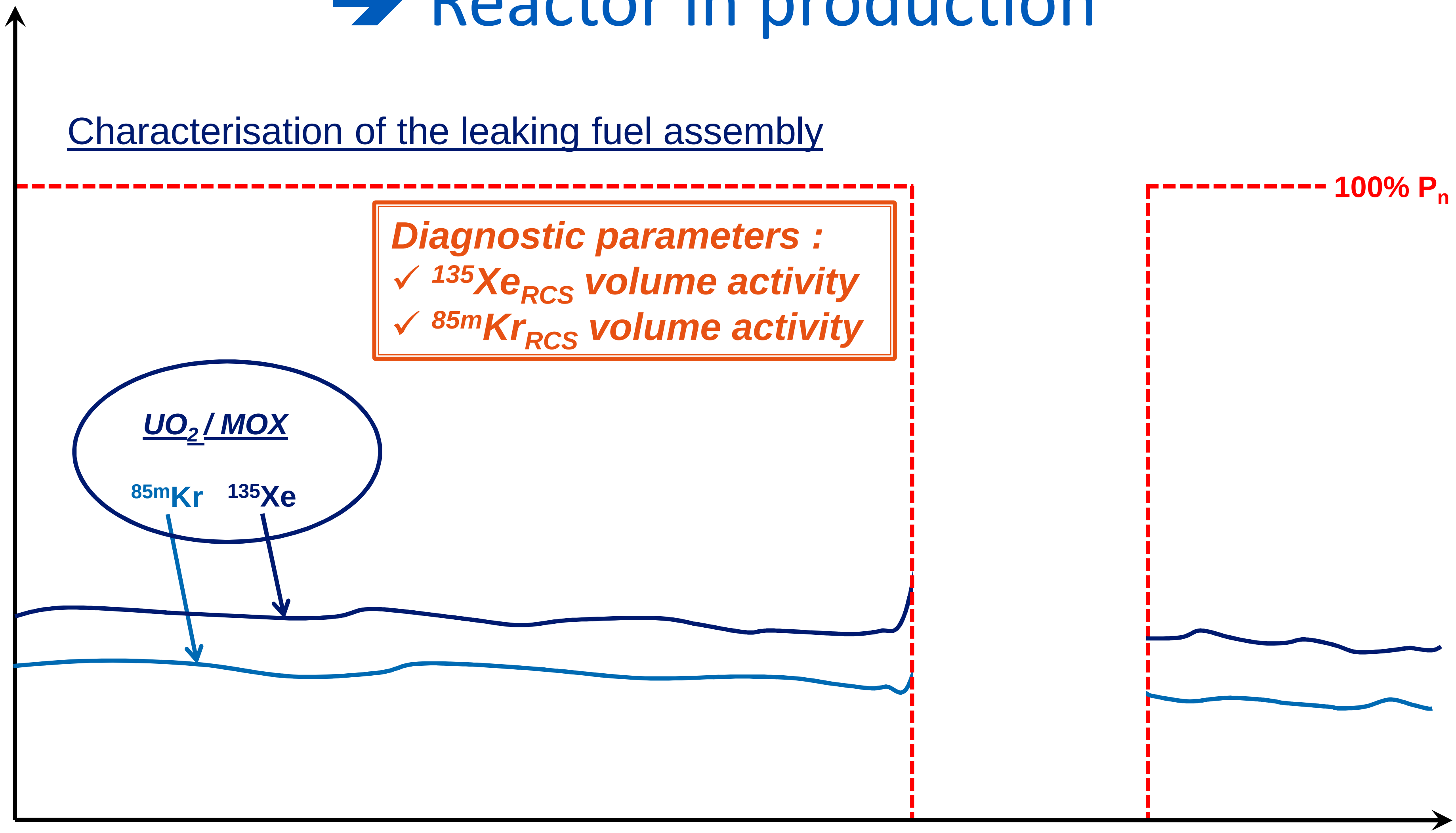
Monitored Parameters (2)

- Chemical nature of the monitored parameters :
  - ✓ Fission products : $^{133/135}Xe$, $^{131/133}I$, ^{134}I, $^{134/137}Cs$
    - ➢ Monitoring of fuel clad integrity.
    - ➢ Control of the radiological exposure & of gaseous discharges.
  - ✓ Activated corrosion products : $^{58/60}Co$, $^{122/124}Sb$, ^{110m}Ag
    - ➢ Control of dose rates during outages.
  - ✓ Actinides : $^{238/239/240}Pu$, ^{241}Am, $^{242/243/244}Cm$
    - ➢ Monitoring of fuel clad integrity : defect degradation.
    - ➢ Control of alpha emitters exposure.
    - ➢ Compliance with discharge requirements.
  - ✓ Activated products : ^{3}H
    - ➢ Control of dose rates during outages.
    - ➢ Compliance with discharge requirements.

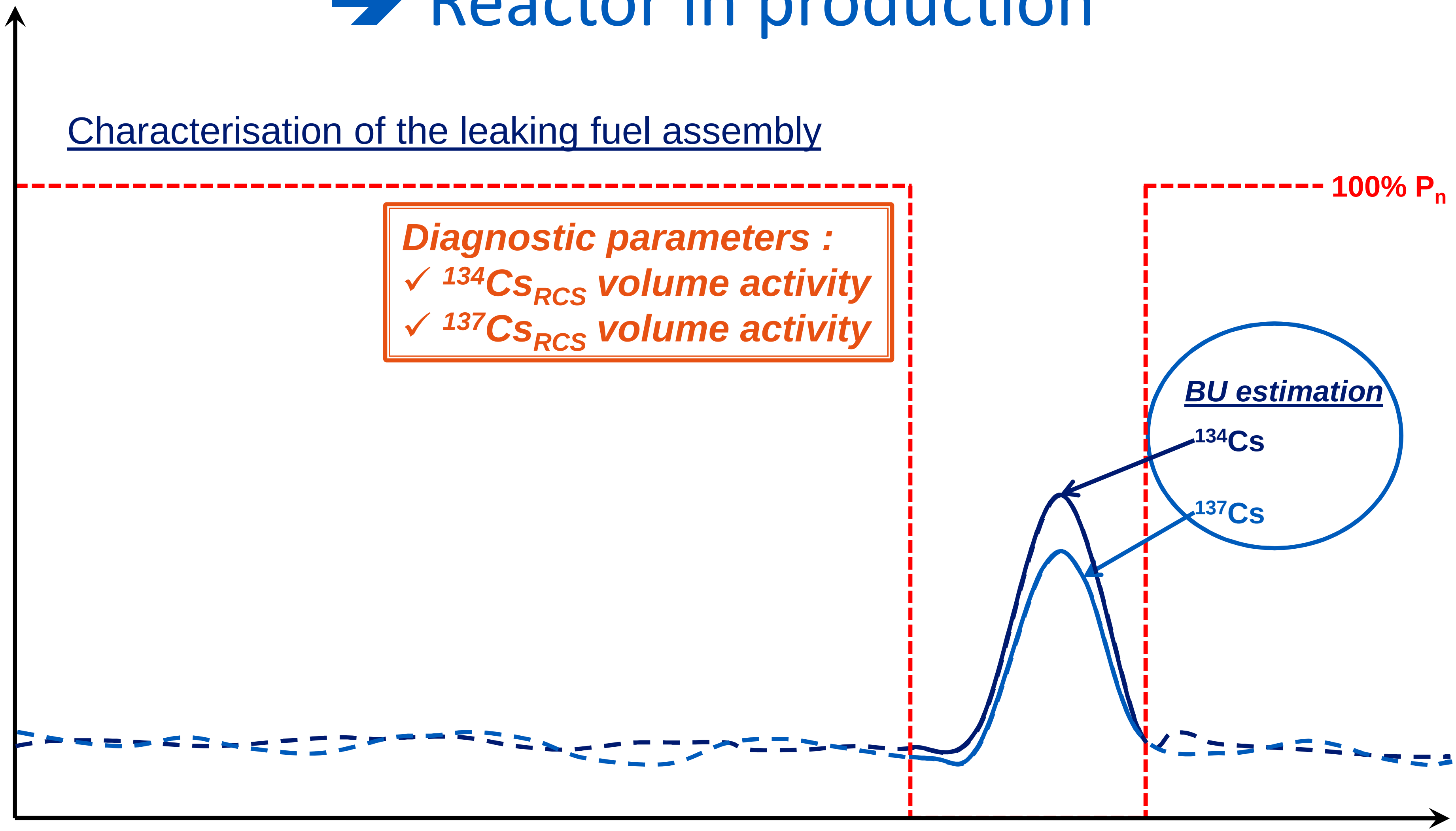
Typical EDF reactor shutdown



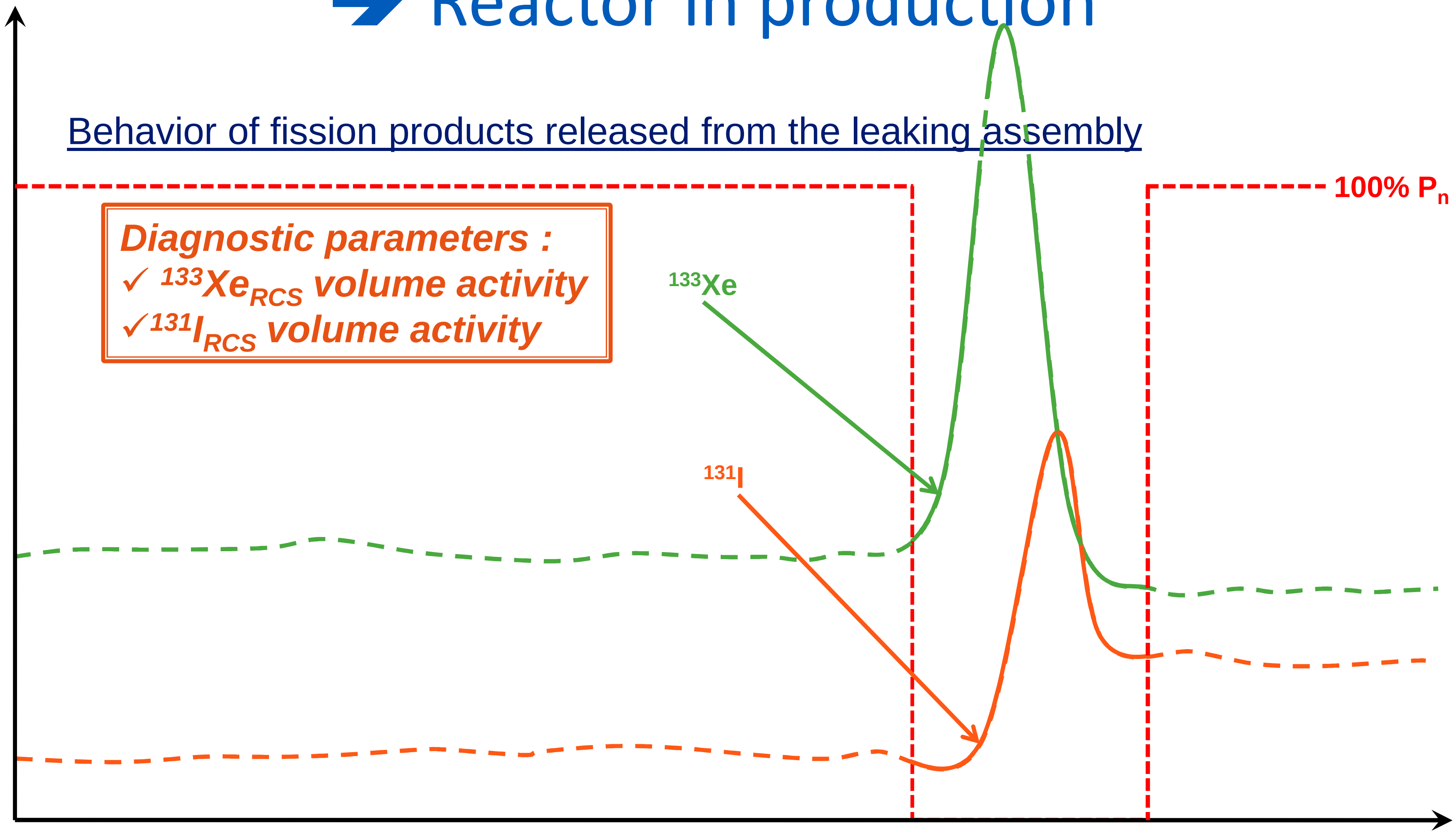
Shutdown and outage preparation
➔ Reactor in production



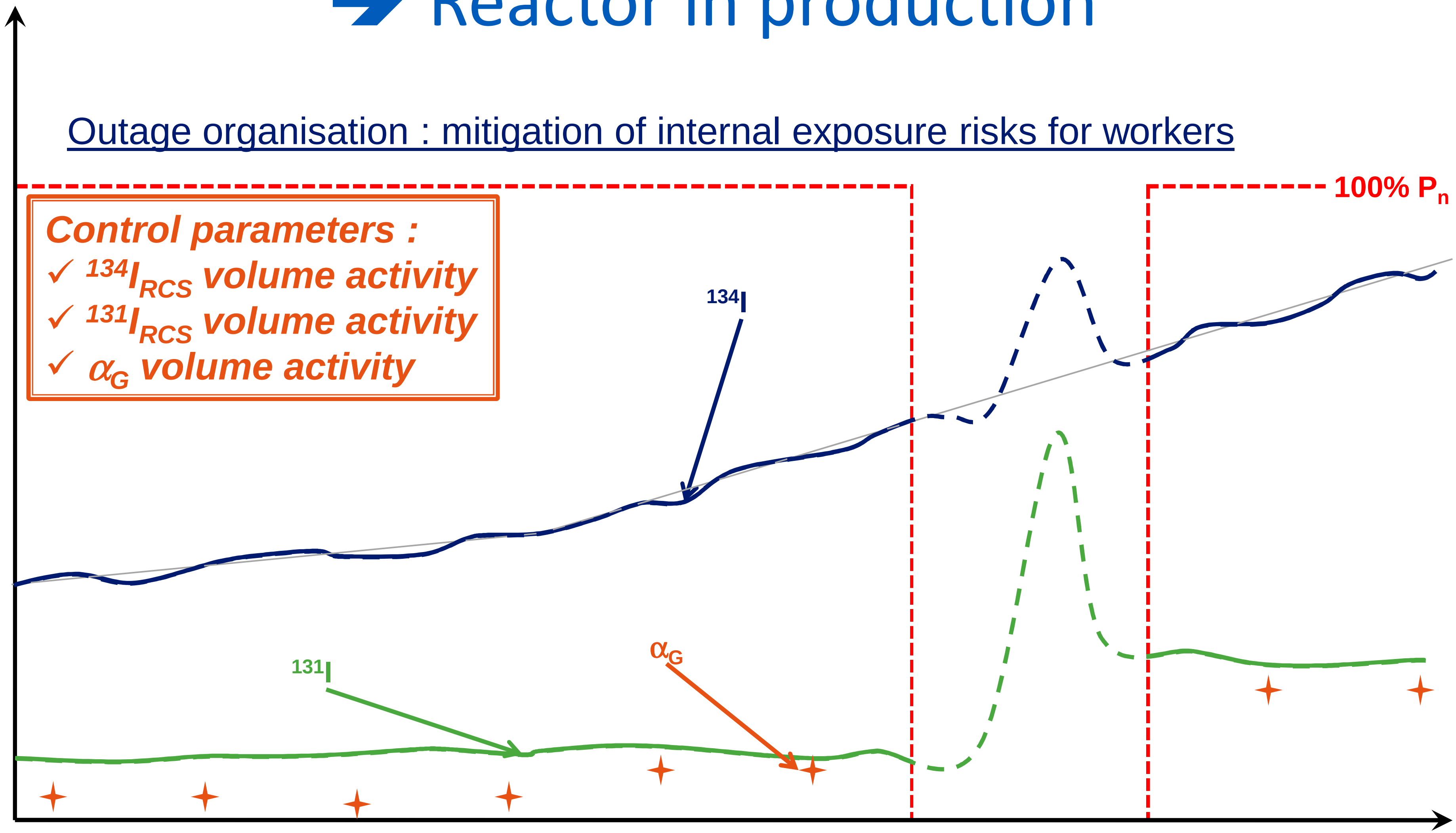
Shutdown and outage preparation
➔ Reactor in production



Shutdown and outage preparation
➔ Reactor in production

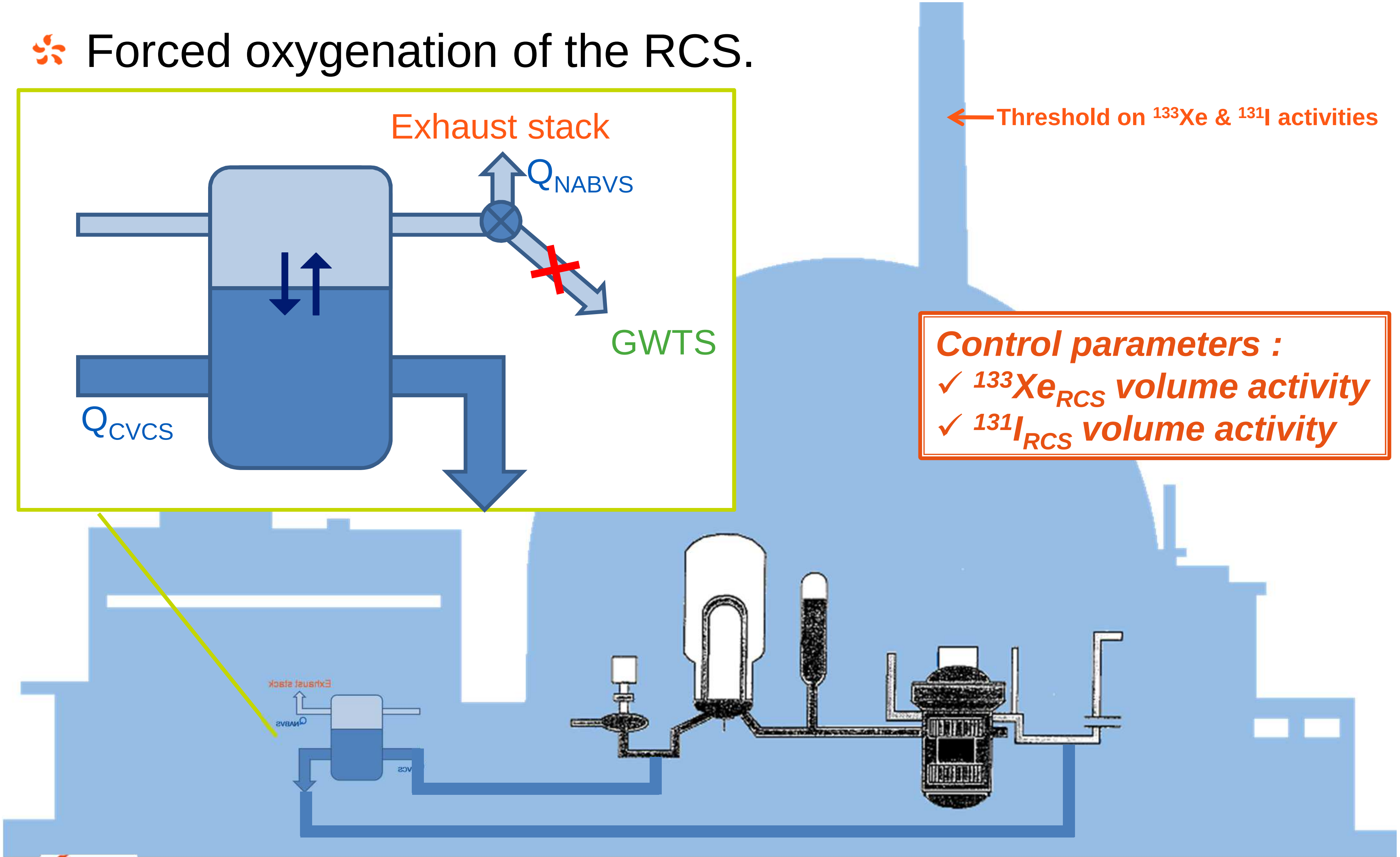


Shutdown and outage preparation
➔ Reactor in production



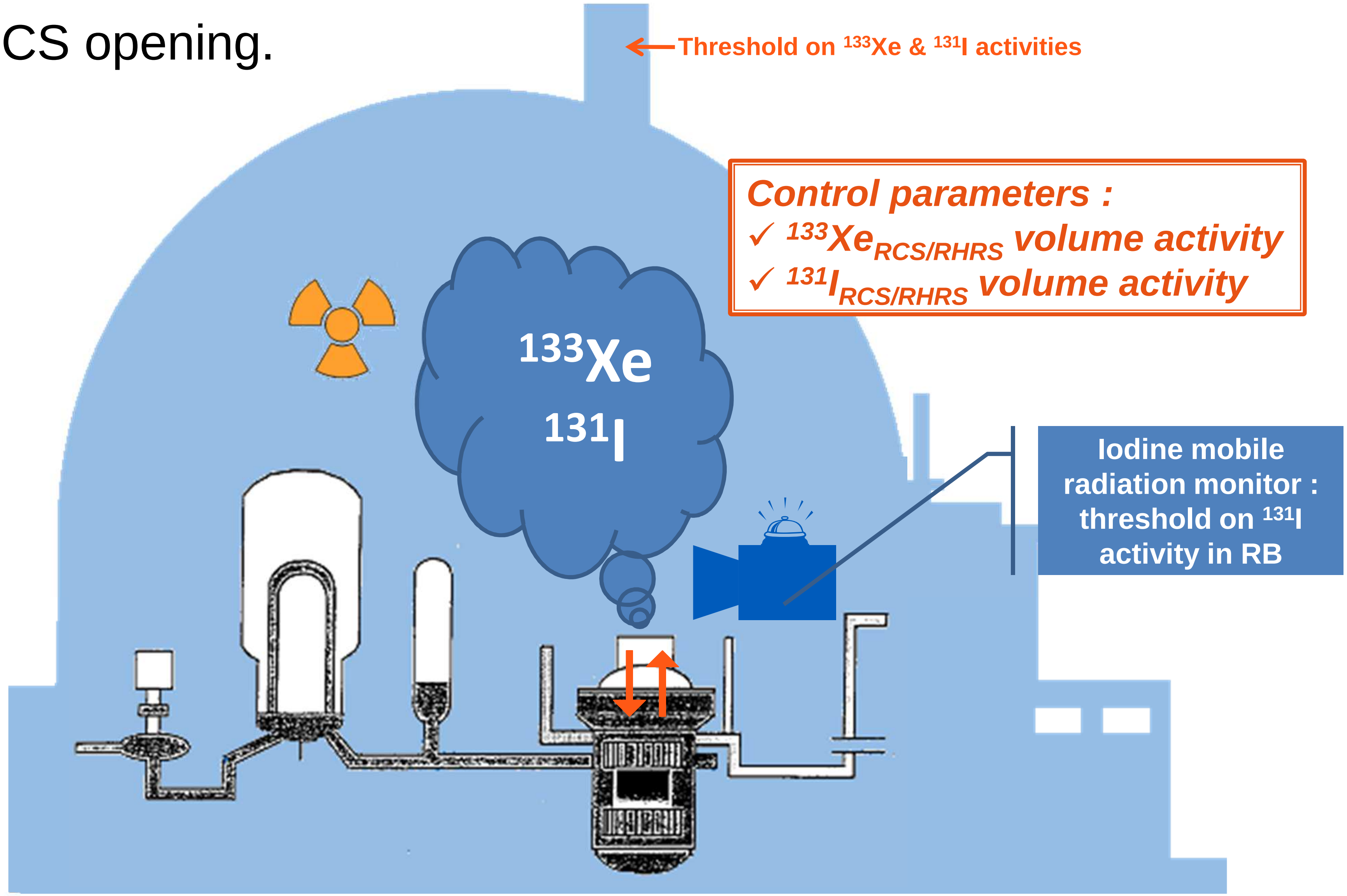
Shutdown monitoring : control of FP (1)

- Forced oxygenation of the RCS.



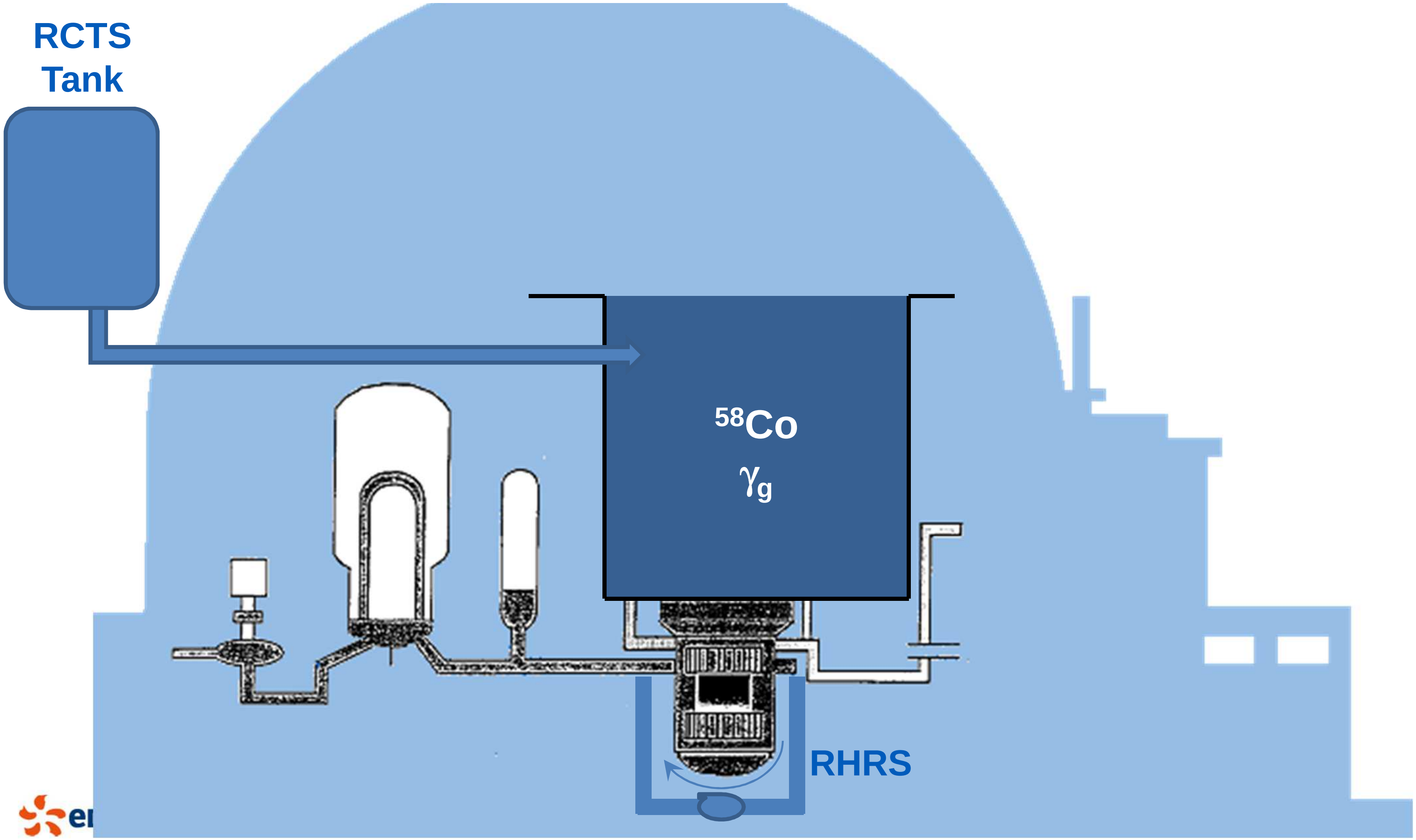
Shutdown monitoring : control of FP (2)

- RCS opening.



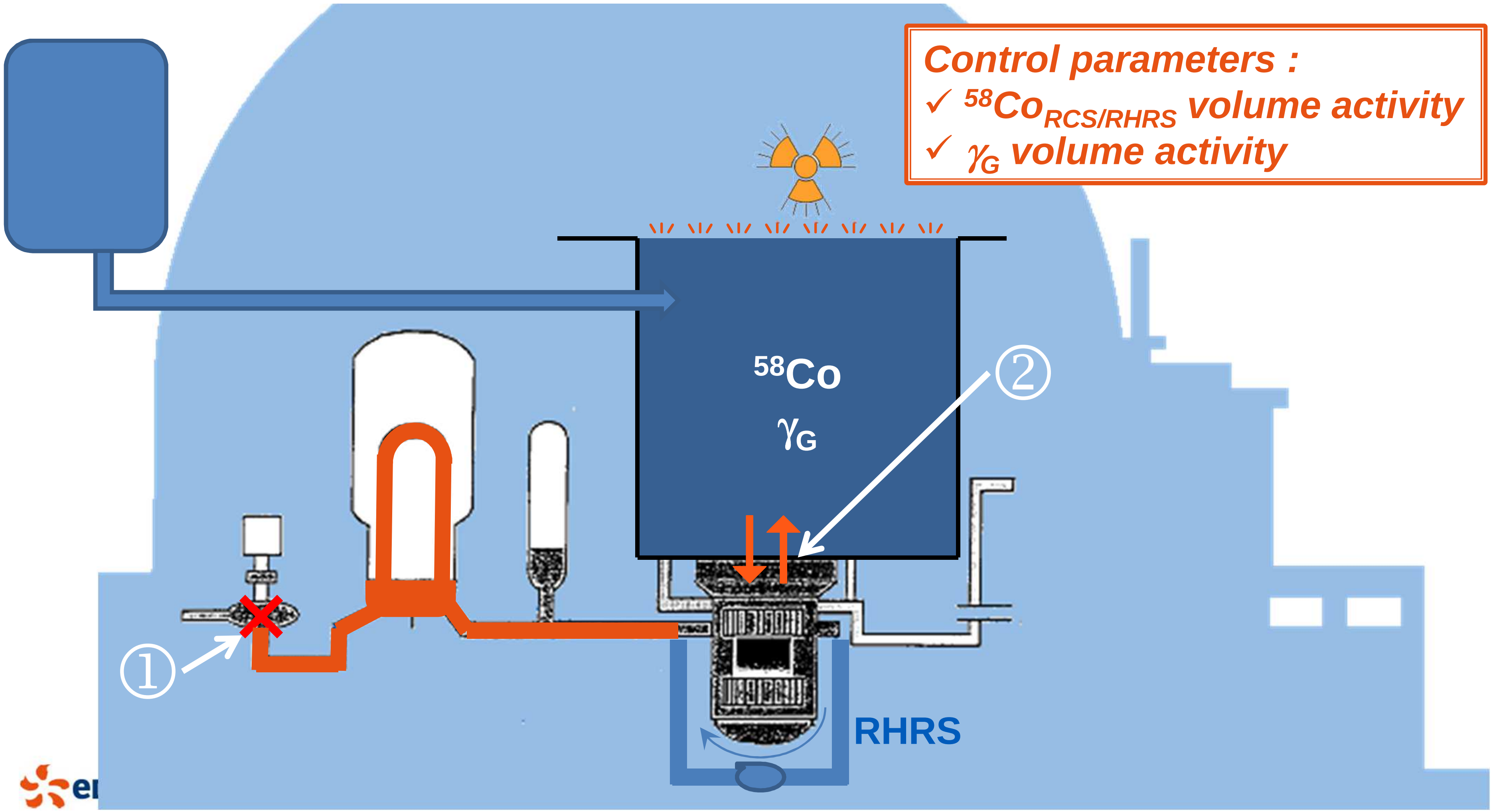
Shutdown monitoring : control of ACP

- To secure the last Reactor cooling pump.



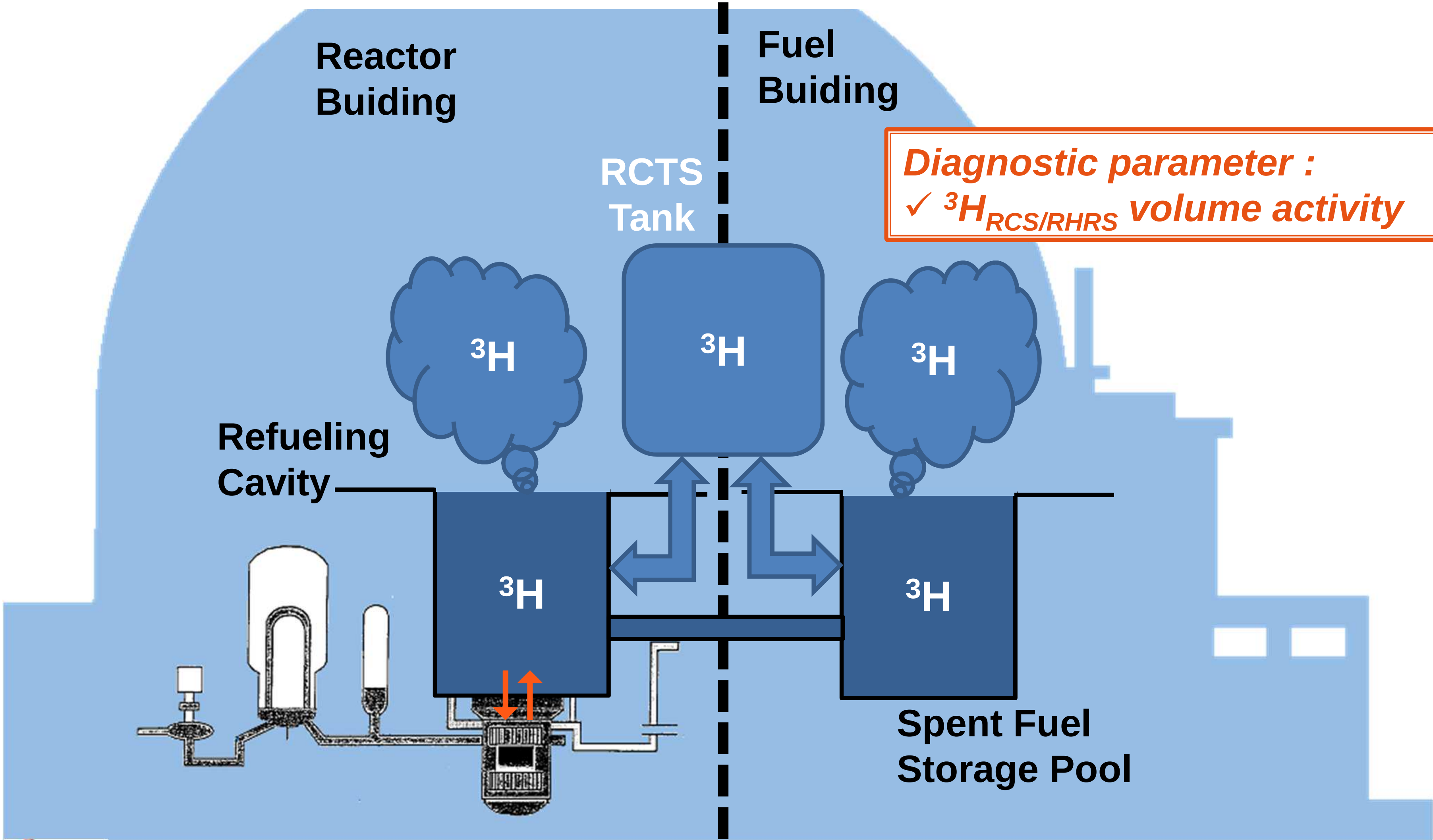
Shutdown monitoring : control of ACP

- To secure the last Reactor Cooling Pump ①.
- To flood the Refueling Cavity ②.



Shutdown monitoring : control of AP

- Expected value on ^{3}H activity to flood the Refueling Cavity.



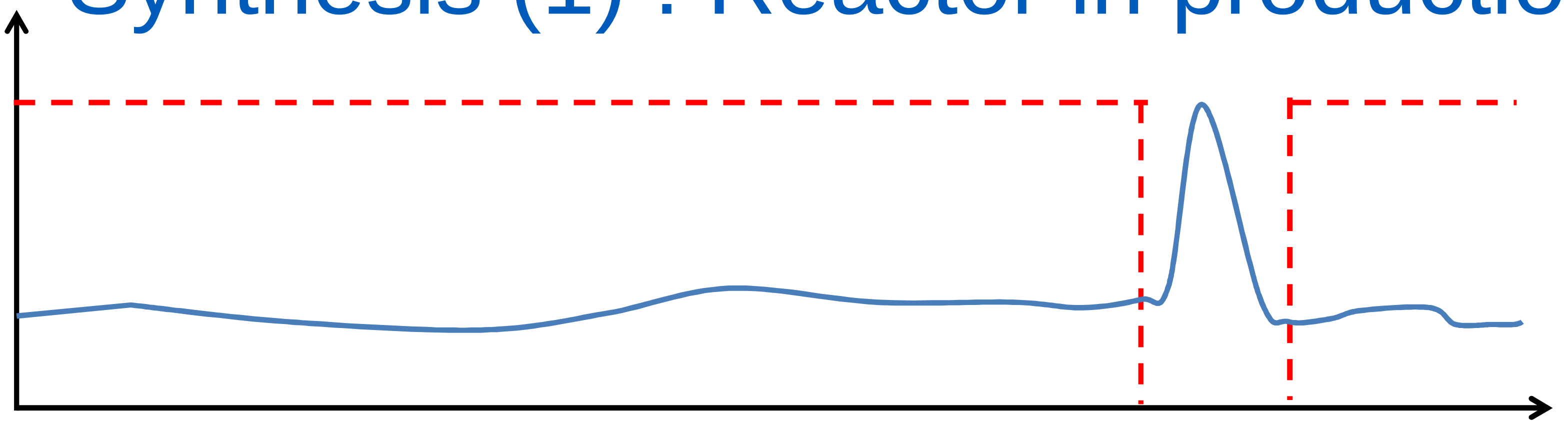
Conclusion

- EDF Radiochemical guidelines include :
  - ✓ parameters with impacts on:
    - ➢ Nuclear safety,
    - ➢ Radiological releases to the environment,
    - ➢ Personnel dose rates,
    - ➢ Operation performances and reactor availability.
  - ✓ diagnostic and control parameters,
  - ✓ parameters identified for reactor in production and during shutdown.

- For each parameter, all the impacts on TSN objectives have to be considered :
  - ✓ To benefit from an improved compromise between all fields,
  - ✓ To stay focused on all the risks,
  - ✓ To anticipate as early as possible the correction and required actions,
  - ✓ To identify and quantify all impacts from any modification.

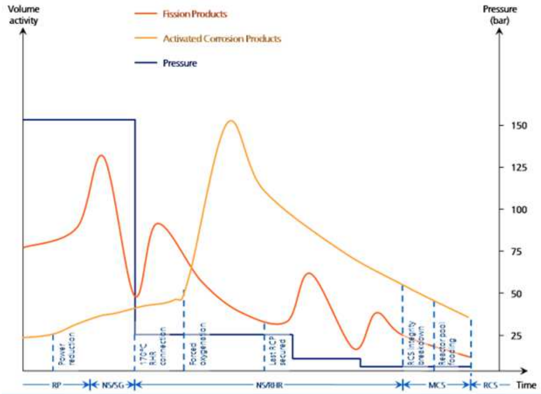
Thank you for your attention.

Synthesis (1) : Reactor in production.



Reactor in production	Parameter	Objective
Characterization of leaking assembly	$^{134}Cs/^{137}Cs$	Burn-up evaluation
	$^{135}Xe/^{85m}Kr$	Identification of fuel type
Behavior of fission products released from leaking rod	$^{133}Xe - ^{131}I$	
Outage organisation	^{131}I	Evaluation of the risk of iodine exposure
	^{134}I	Evaluation of the risk of alpha emitters exposure

Synthesis (2) : Shutdown monitoring.



Shutdown step	Parameter	Objective
Power setback	$^{133}Xe - ^{131}I$	
Oxygenation of RCS	$^{133}Xe - ^{131}I$	Control of gaseous discharges
To secure last RCP		Control of dose rates
	$^{133}Xe - ^{131}I$	
RCS opening / Pressure vessel lift	$^{133}Xe - ^{131}I$	Control of radiological exposure to fission products
Filling of refueling pool	$^{58}Co - \gamma_{global}$	Control of dose rates
	^{3}H	Control of dose rates & gaseous discharges