## SOURCE TERM REDUCTION STRATEGIES FOR EDF PWRs

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#### ABSTRACT

The reduction of doses is a strategic purpose for EDF in link with the stakes of productivity gains, nuclear acceptability and respect of regulation. This consists not only in improving the reactor shutdown organization (time spent in control area, biological shielding, ...) but also in improving the radiological state of the unit and the efficiency of the source term reduction operations.

Since 2003, EDF has been running an innovative project called "Source Term Reduction" federating the organization in the different EDF research and engineering centers in order to :

- participate to the long term view about RP issue (international feedback analyses),
- develop contamination prediction tools (OSCAR) suitable for the industrial needs (operating units and EPR conception),
- develop scientific models useful for the understanding of contamination mechanisms in order to simplify the strategic decision processes,
- carry on with the updating and the analysis of contamination measurements feedback in corrosion products (EMECC and CZT campaigns) and fission products and actinides,
- carry on with the operational support at short or middle term by optimizing startup and shutdown processes, passivation or zinc injection procedures and by improving purification media or material characteristics.

This paper will describe the main R&D developments and the actions brought or planned to be brought into operation on EDF units as a part of this project.

### I – Introduction

The reduction of doses is a strategic purpose for EDF in link with the stakes of productivity gains, nuclear acceptability and respect of regulation. During the last 15 years, EDF dosimetry has significantly decreased :

- collective dosimetry : from 2.44 H.Sv in 1991 up to 0.66 H.Sv in 2008,
- personal dosimetry : in 1992, 1200 operators received doses higher than 20 mSv/y and in 2008, no operator received doses higher than 18 mSv/y.

EDF strategy consists not only in improving the reactor shutdown organization (time spent in control area, biological shielding, ...) but also in improving the radiological state of the unit and the efficiency of the source term reduction operations (corrosion and fission products).

Since 2003, EDF has been running an innovative project called "Source Term Reduction" (S.T.R.) federating the organization in the different EDF research and engineering centers [1].

A general overview of this project is given in section II. Sections III to V describe the different technical parts of the source term reduction program planned by EDF as much in terms of anticipation (prediction tools and modeling) as in terms of operational solutions (operation procedures, purification management or material improvements).

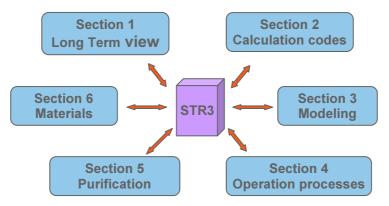
## II – General project description

The main working axes of the «Source Term Reduction» are :

• participation to the long term view about RP issue (international feedback analysis),

- development of contamination prediction tools (OSCAR) suitable for the industrial needs (operating units and EPR conception),
- developments of scientific models useful for the understanding of contamination mechanisms in order to simplify the strategic decision processes,
- update the analysis of contamination measurements feedback in corrosion products (EMECC and CZT campaigns) and fission products and actinides,
- operational support at short or middle term by optimizing operation processes, and especially startup and shutdown processes, passivation or zinc injection procedures and by improving purification media or material characteristics.

In order to meet these general needs, the "Source Term Reduction" project has been divided into 6 different parts as shown in picture 1.



*Picture 1 : Project segmentation* 

# III – Long-Term view

A technical watch is continuously achieved by EDF in order to :

- provide a dosimetric level evaluation of French and foreign units and compare EDF performances with international results,
- list different practices, carried out by foreign operators, which are supposed to have a significant impact on source term and thus, on radiation protection (design and operation).

This technical monitoring is mainly possible thanks to EDF/EPRI collaboration and several missions and technical discussions with other operators specifically on primary chemistry in operation or during shutdown transients.

Thanks to well-focused assessments about contamination and national and international feedback analysis, the knowledge accumulation over time highlights efficient units and their efficiency key factors and leads to progress lines detection and long-term planning.

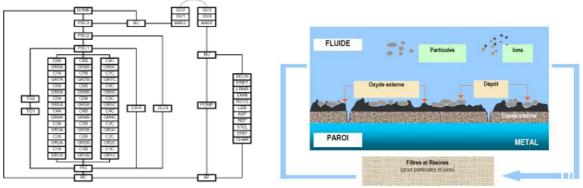
### IV - Prediction tools and modeling developments

### IV.1 – Prediction tools

Considering the cost of a specific experimentation and the difficulties encountered for analyzing the impact of a single parameter among all varying parameters impact in operation, calculation codes are an interesting and useful tool to predict contamination levels in the primary coolant. These codes allow to aggregate the whole scientific knowledge obtained for more than 40 years in order to :

- give, in a short-term, a support to operating process optimization (start-up, shutdown, hydrogen concentration, pH, zinc injection impact) and to design studies (EPR, SGR),
- lead, in a long-term, to knowledge transfer towards common tools used in operation.

For example, the OSCAR V1.1 code [2], developed by CEA for EDF and AREVA NP, allows to simulate, in a single code, corrosion and fission products behavior in the primary coolant operating at high temperature level. OSCAR V1.1 is fitted with a more efficient thermo-chemical database (nickel solubility data at high and low temperature) and a complete release module for corrosion products. For fission products, OSCAR V1.1 benefits from a thermo-mechanical module ALCYONE (more accurate calculation of the behavior of fuel inside defective rods).



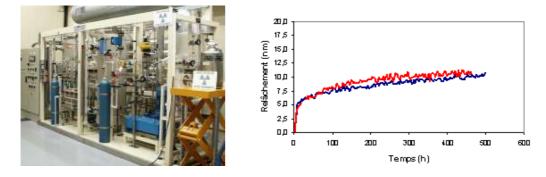
Picture 2a : 900 MWe-series RCS+CVCS modeling with OSCAR Picture 2b : metal / oxide / water modeling with OSCAR

Further OSCAR developments are planned to :

- improve corrosion products contamination prediction during shutdown transients (temperature and chemical conditioning effect, oxygenation peak and purification) and extend calculations to auxiliary systems as RHRS,
- take into account EPR design and operation characteristics (B/Li management, stellites reduction, ...),
- improve fission products contamination prediction by a model calibration procedure based on the feedback available for defective fuel assemblies,
- integrate a module for actinide behavior modeling (uranium oxide dissolution, actinides precipitation/dissolution mechanisms in the primary coolant, link between actinides and corrosion products behavior).

### *IV.2 – R&D modeling developments*

At the same time, R&D groundwork is carried out not only in order to prepare the further codes evolutions but also to improve contamination mechanisms knowledge for possible short-term decision in case of incident in operation (metal species releasing modeling with the help of the BOREAL loop, colloidal particles characterization and sorption research, nickel oxide solubility and dissolution kinetic measurements in collaboration with CEA).

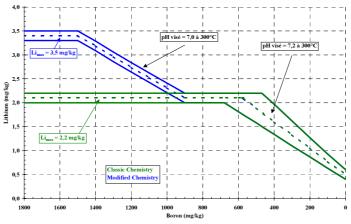


*Picture 3a* : BOREAL loop *Picture 3b* : Releasing curve obtain with BOREAL loop (Boron : 1000 ppm ; Lithium : 2 ppm ; H2 : 30 cc ; T = 300 °C)

#### V - Operational procedures improvements

#### V.1 – Operation, shutdown and start-up procedures

Reactor operation procedures can have a significant effect on source term. It is well known that chemistry management has a relevant role on corrosion product contamination. A "modified chemistry" (based on a maximum authorized lithium concentration of 3.5 mg/kg) has been run on several French units for 5 years (1300 MWe pilot unit – Cattenom 2 – and N4-series) and will be extended in 2010 to 1300 MWe units on which new fuel management (Galice) will be used. Therefore, the feedback will be sufficient to analyze the impact of boron/lithium management and pH on source term and thus radiation protection and to compare to international results.



Picture 4 : Boron / Lithium management in french fleet.

Moreover, in addition to the chemistry, new fuel management (N4 "Alcade", 1300 MWe "Galice", 900 MWe "Parité MOX") influence study on contamination and dose rates is also planned (especially cycle length extension and power increase impact). OSCAR calculations will also be considered with a view to consolidate the feedback analysis.

Furthermore, zinc injection has been implemented in France since 2004 at Bugey 2 – Cycle 22 in order to reduce dose rates (curative aim). In 2005, zinc injection was also initiated at Bugey 4 – Cycle 23, during the previous cycle of the steam generator replacement (preventive aim). The results [3] allows to conclude that there is no contraindication to add zinc in terms of contamination and dose rates. The positive effect of zinc on contamination observed at Bugey 2 and Bugey 4 is not as significant as observed in other foreign units. An encouraging slight decrease trend can be underlined but needs to be confirmed. Based on Bugey results and on international feedback, EDF decided to implement zinc injection on 8 new units from 2009 to 2011, not only to control contamination and dose rates and confirm Bugey results, but also to manage a potential AOA risk on future core design and to mitigate PWSCC.

Finally, new practices feedback as chemical dehydrogenation and bubble collapsing at  $130^{\circ}$ C (put in practice on EDF units since 2004) or fast cooling at  $-40^{\circ}$ C/h (experimented on several 900 MWe series units) will be studied (statistical analysis of dose index, collective dose and CZT campaigns measurements).

### V.2 – Measurement campaigns

EDF strategy in terms of contamination monitoring is based on two complementary ways. Usually, routine dose rate measurements are achieved by plant radiation protection teams on primary loops (cold, hot and cross-over legs) in order to calculate a RCS index and to be able to compare performances for each unit.



However, this current program is not complete enough to allow an accurate analysis when specific experiments are carried out (modified chemistry, zinc injection, new fuel managements, steam generator replacement) or in case of specific contamination : <sup>122</sup>Sb and <sup>124</sup>Sb (secondary source assembly), <sup>110m</sup>Ag (control rods and primary pumps) or <sup>60</sup>Co (mainly stellites) hot spots. In that case, EMECC campaigns [4] (achieved by CEA) have been commissioned for more than 30 years on French fleet units in order to better characterize contamination mechanisms. At the same time, EDF has also commissioned and financed EMECC campaigns on foreign units (Doel, Sizewell, Trillo during the 4 last years) with the contribution of several European operators in order to compare different good international practices.

Picture 5 : EMECC device

In addition to EMECC campaigns, EDF have been carrying out a new dose rate measurement program since 2006 based on a semi-conductor CZT probe. Comparison with EMECC results, more accurate but also more difficult to handle, shows that the CZT device is able to satisfactorily quantify the main radionuclides contribution to equivalent dose rate [5]. The first feedback analyses confirms that CZT is a pertinent tool to understand contamination mechanisms. Technical modifications are planned to improve sensitivity.

Finally, following actinides contamination at Cattenom 3 during cycle 8, several measurement campaigns have been planned in order to better understand actinides behaviour in the primary and auxiliary circuits. One goal is a new actinides behaviour model, which could be introduced in OSCAR calculation code. The mini-CVCS campaign, planned at Cattenom 3 after cycle 15, will give information about reliability of measurements achieved via the sampling circuit compared to RCS circuit.

# VI – Purification improvements

In parallel with the control of source term, an important optimization program for purification is under way at EDF as much in terms of new equipments as in terms of purification management optimization.

### VI.1 – Technological survey and R&D developments on filters and resins

A technological survey of new purification devices available in the marketplace is in progress by means of meetings with filter and resin suppliers. These discussions are an efficient way to communicate about EDF needs with a view to be proactive before suppliers.

Concerning filters improvements, several specific new technologies are planned to be tested. For example, the possibility to use filters based on an ultrafiltration membrane technology is studied as a possibility of pleated media filter replacement currently in place in the CVCS purification system. Firstly, experimentations in laboratory (CEA) are in progress. Then, a specific test directly in the nuclear plant is planned by the end of the year 2010 (during operation for 1 month). This technology is expected to be interesting for obtaining a thinner filtration threshold than this available at the moment. A further technical and economic feasibility study will be done on the basis of the tests results in order to decide finally if this technology has an interest in being used in EDF NPPs.

At the same time, silica-free filters are currently homologated for use in NPPs. Indeed, reduction of silica source term is important both for boron release and to avoid the negative impact of a possible zinc silicate precipitation on fuel cladding during zinc injection. These new filters are proposed to be experimented in a nuclear plant before 2011.

In parallel, experimentations with resins are also carried out in laboratory and in nuclear plant. First of all, two types of cation exchange resins (gel and macroporous) were characterized in laboratory (EDF/R&D) in 2008 [6]. A new study has been launched in view to improve the knowledge on anionic exchange resins performances (exchange capacity, boron retention, iodine selectivity) as well as exchange kinetics and flow rate impact. Moreover, chemical conditioning (and more precisely peroxide hydrogen) impact on resins damages and lifetimes is planned to be done during this project.

## VI.2 – Purification management optimization

Subsequently, purification management is another important way to improve purification efficiency. As an example, national market for filters and resins, launched in 2007 and 2008, include several filter and resin manufacturers and shows the importance of stabilizing both supplying assurance and costs control. Moreover, operational guidelines for filters and resins operation have been written in order to provide to nuclear plant operators reference documents. They incorporate a description of filters and resins characteristics, a description of their role and the main operating conditions, the replacement criteria and the main guidelines to operate these materials. These support documents are currently used in French fleet and meetings are organized each year with chemistry teams of each unit in order to collect operational feedback and to update recommendations.

Furthermore, EDF/R&D has been working for several years on a modeling tool (OPTIPUR) with a view to optimize the management of auxiliary circuits demineralization devices in operation. This useful tool will be used in the future, among others, for studying the best compromise between resin cost and lifetime with an eye to the national resin market renewal.

Finally, the usual practice for shutdown purification consists in using a full resin volume deep bead column for several successive shutdown transients. The opportunity to use only one resin per shutdown transient with a lower resin volume is studied (experimentation planned in 2009). This practice would avoid releasing of radioactive contaminant firstly retained during the previous shutdown (especially colloidal silver species).

# VII – Material characteristic improvements

For EDF, source term reduction is also closely correlated to the type of materials used in the primary and auxiliary circuits and to their surface state.

# *VII.1 – Contamination source reduction*

A particular attention has to be paid on the reduction of cobalt-based material (Stellites) used for their excellent mechanical performances but, nevertheless, at the root of <sup>60</sup>Co contamination. As a part of this project, a specific design study for EPR showed that cobalt-based material reduction could reduce dose rates by 15% and a similar work still needs to be achieved for the whole fleet. In a same way, the impact of antimony (secondary source assembly and primary pump buttering and bearing) and silver (AIC control rods, pump seals) sources on dose rates will be studied from a feedback analysis. Considering these results, replacement material and their influence on dose rates will be examined in order to define a general suppression or reduction policy for EDF.

# *VII.2 – Steam generator tubes*

Considering steam generator tubing importance in contamination mechanisms (metal species releasing over a large exchange surface), an accurate control of manufacturing process is achieved in order to maintain a good behaviour whatever process modifications. As a part of this project, a significant influence of SG tubes surface state on releasing has been demonstrated with the help of BOREAL loop. This characterization facility allows to test process improvements or modifications with a view to alert to nonconformity behavior.

Moreover, the definition of SG tubes specifications could also be a good way to assure a low releasing rate (surface roughness, metal hardening, impurities, grain size, microstructure or oxide composition). Nevertheless, studies carried out until now have not led to univocal specifications which could be used to link SG tubes behavior to their surface state. Further works will be realized on surface state and oxide characteristics in order to have a better understanding of mechanisms involved in releasing (testing loop TITANE and BOREAL, XPS and SIMS characterizations).

### VII.3 – SG tubes surface treatments

SG tubes surface pre-oxidation has been identified for a long time as a mean of dosimetry reduction. Optimal physicochemical conditions during (pre-critical or not) hot functional tests have been analytically studied by EDF/R&D department. These results, combined with experimentations in BOREAL loop lead to propose a specific procedure both in the case of startup procedure after SG replacement and first startup of a new unit (EPR). This analytic study will be completed as a part of this project by a theoretical international feedback (collaboration with Sizewell B) and additional experimental tests in BOREAL loop in order to validate definitively analytic conclusions.

At the same time, gaseous oxidation could be a positive technique to reduce metal species releasing in the primary circuit by the generation of a protective oxide layer similar to this created with a chemical procedure. An experimental test with the help of the BOREAL loop is also planned in 2010 in order to improve mechanisms and efficiency knowledge.

## VIII – Conclusions

Source term reduction is an important matter of concern for EDF French fleet. A specific project has been launched for 6 years in order to federate R&D and engineering actions with a view to decrease contamination level and dose rate applied to the French fleet. The project program deals with investigations about innovative technologies, tools (calculation codes) or procedures in one hand, and practical answers in operation in the other hand.

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