The OSCAR code: a simulation tool to assess the PWR contamination for decommissioning

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1 Introduction

Knowing the contamination state of the end-of-life nuclear reactor systems by Long-Lived RadioNuclides (LLRNs) is a key stage for the decommissioning process. Indeed, the initial state is necessary to optimize the dismantling operations and the radioactive waste management. To address this issue, the contamination state is usually characterized using different types of techniques: in-situ gamma spectrometry, γ camera scanning, dose rate measurements, α, β, γ measurements of samples obtained by smears or scrapings and then chemical separation processes, scaling factor approach… To reduce the amount of these measurements and thus the Occupational Radiation Exposure (ORE) and the decommissioning costs, a method is to assess the level of contamination by numerical simulation. Furthermore, at the design stage of a new reactor, its decommissioning has to be taken into account and a simulation tool, such as the OSCAR[a] code, allows to predict the radioactive source term at the end of life of a future reactor.

2 The OSCAR code

The contamination of nuclear systems can have three origins:

- Activated Corrosion Products (ACPs): uniform corrosion of materials leads to the release of Corrosion Products (CPs) into the coolant, which are activated under neutron flux.
- Actinides and Fission Products (AFPs): in case of rod cladding failure, Fission Products (FPs) and possibly actinides are released into the primary coolant.
- Coolant Activation Products (CAPs): the coolant, its additives and impurities are activated when passing through the core.

Deposited on the out-of-flux surfaces, these radioactive products contaminate the nuclear systems.

The OSCAR code is devoted to the simulation of the transfer of ACPs, AFPs and CAPs in the reactor coolant and auxiliary systems. It calculates the masses and activities of isotopes (CPs, ACPs, AFPs and CAPs) in the solid, liquid and gaseous phases of nuclear circuits as a function of time during normal operation over several decades and during transients down to a few seconds as well. The OSCAR code has been developed by the CEA (the French alternative energies and atomic energy commission) in collaboration with EDF and Framatome since the 1970’s. It has resulted from the merging of two former codes in 2008: PACTOLE for ACPs and PROFIP for AFPs [1] [2]. Thanks to the modularity of the OSCAR code, an application to Sodium-cooled Fast Reactors (SFRs), so-called OSCAR-Na, and another one to fusion reactors (ITER/DEMO), so-called OSCAR-Fusion, have been developed to treat the behavior of ACPs in these nuclear reactors [3].

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[a] OSCAR acronym for “Outil de Simulation de la ContAmination en Réacteur” in French or “tOol for Simulating ContAmination in Reactors” in English.

2.1 The OSCAR modelling

The OSCAR code modelling is based on a control volume approach; briefly:

- The nuclear systems are nodalized into as many control volumes (regions) as necessary, defined according to their geometric, thermal-hydraulic, neutronic, material and operating characteristics (see the nodalization of a typical Pressurized Water Reactor (PWR) in Figure 1).

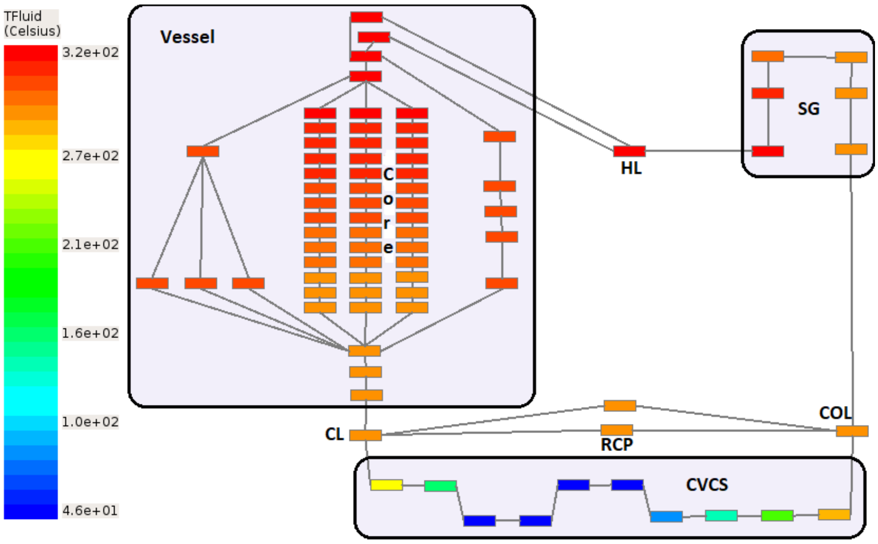


Figure 1. OSCAR - RCS (Reactor Coolant System) and CVCS (Chemical and Volume Control System) nodalization of a typical PWR with a third-core reload fuel management (HL: Hot Leg / SG: Steam Generator / COL: CrossOver Leg / RCP: Reactor Coolant Pump / CL: Cold Leg).

- Several media can be defined in each region (see Figure 2):
  - Immobile media: Metal (stainless steel, alloy 600…), Inner oxide (passive Cr-rich layer) and Deposit/Outer oxide (porous Fe-Ni-rich layer);
  - Moving media: Particles, Agglomerates and Solubles (dissolved species);
  - Trapping media for the coolant purification in the CVCS: Filter for Particles and Agglomerates, Ion-Exchange Resins (IER) for Solubles.

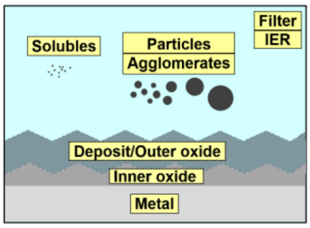


Figure 2. OSCAR V1.4 - Media.

- The following elements and their corresponding radioisotopes can be taken into account in OSCAR V1.4:
  - CPs: Ni, Co, Fe, Cr, Mn, Zn, Ag, Sb, Zr and Cu
  - FPs: Xe, Kr, I, Cs and Sr
  - Actinides : U, Pu, Np, Am, Cm…
  - CAPs : ^{16}N and ^{41}Ar

  The radioactive half-lives of the radioisotopes range from some seconds to a million years.

- The OSCAR code calculation kernel solves a system of mass balance equations for all isotopes (stable and radioactive) in all media of all regions at each adaptive time step (from one second to several days) using the following equation:

$$\frac{\partial m^i}{\partial t} = \sum_{sources} J^i_{transfer} - \sum_{sinks} J^i_{transfer} \qquad \text{Eq. (1)}$$

where m^i is the mass of isotope i in a given medium [kg], t the time [s] and $J^i_{transfer}$ a transfer mass rate of isotope i between 2 media or 2 regions or 2 isotopes [kg.s^{-1}].

For ACPs, all the transfer mechanisms taken into account in OSCAR V1.4 are presented in Figure 3 (further details in [4]).

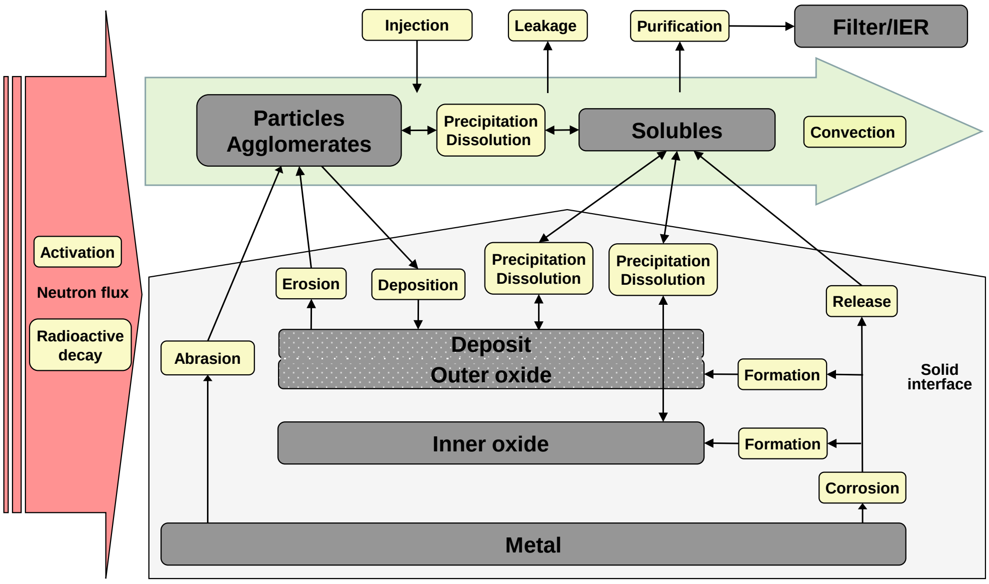


Figure 3. OSCAR V1.4 - Mass transfer mechanisms involved in the ACP contamination.

For AFPs, the main transfer mechanisms taken into account in OSCAR V1.4 are summarized in Figure 4 (further details in [1]).

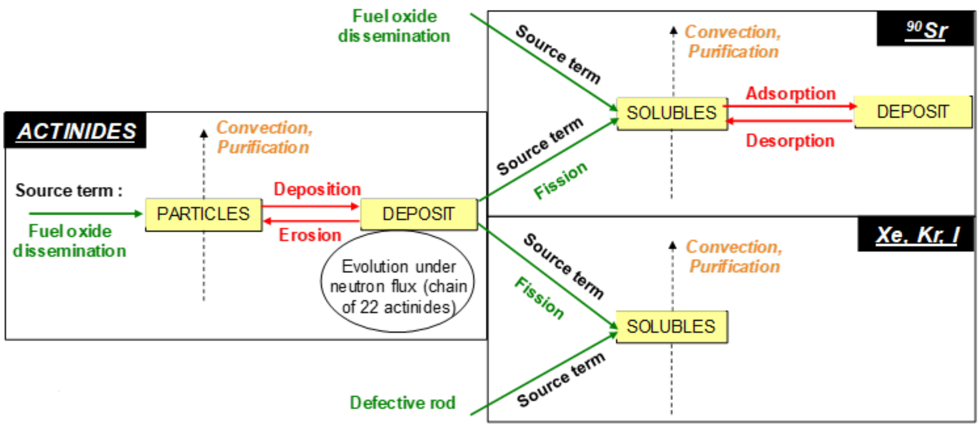


Figure 4. OSCAR V1.4 - Main mass transfer mechanisms involved in the AFP contamination.

2.2 The OSCAR validation

The validation of the OSCAR code consists in comparing OSCAR simulation results with contamination measurements in PWRs. For ACPs, the validation of OSCAR is based on an operational experience feedback unique in the world, the so-called EMECC[b] campaigns, which consists in measuring γ activities deposited on PWR primary and auxiliary systems using the EMECC device [5]. To date, 420 EMECC campaigns have been performed by the CEA in 74 different French and foreign PWRs since 1971 [6]. In addition to the surface activities, the OSCAR results are compared to other on-site measurements: volume activities and chemical element concentrations[c].

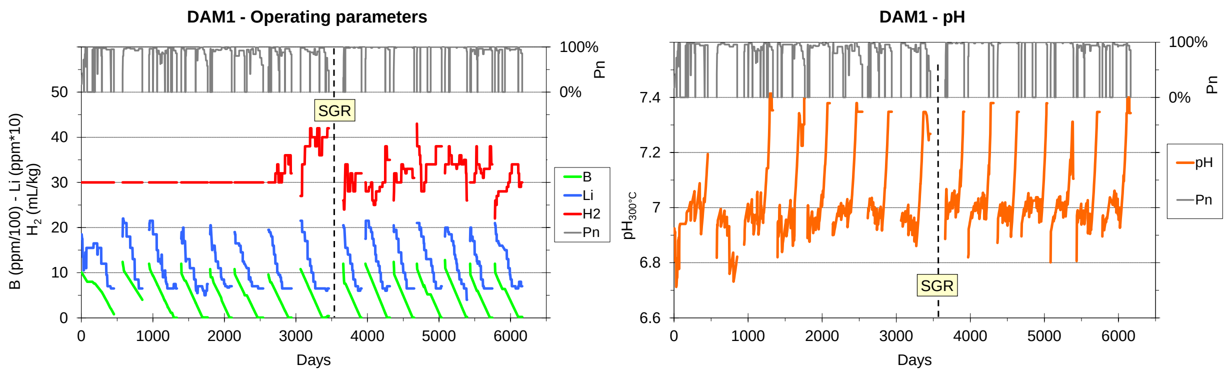
[b] French acronym for « Ensemble de Mesures et d'Étude de la Contamination des Circuits ».

[c] On-site dose rate measurements are not considered because not only they reflect the surface and volume activities but also they depend on the activities of radionuclides, on the measured components, on the measurement conditions and on the ambient dose rates.

To illustrate the capacity of the OSCAR code to calculate the ACP surface activities of a real PWR, the results of the OSCAR simulation of DAMPIERRE-1 are compared to the EMECC measurements carried out at the very beginning of several outages of this unit.

DAMPIERRE-1, a 900 MWe PWR commissioned in 1980, underwent a Steam Generator Replacement (SGR) at the outage of cycle 8. The Inconel 600MA SGs have been replaced by Inconel 690TT SGs. The average Co content of the SG tubes was 270 ppm before SGR and 130 ppm after SGR. From the 6th cycle, the fuel assembly grids made of 718 alloy were replaced by Zircaloy grids.

The first 15 operating cycles with an aimed $pH_{300°C}$ of 7.0 have been simulated. The simulated operating parameters and the $pH_{300°C}$ calculated using OSCAR/PHREEQCEA are presented in Figure 5. PHREEQCEA, the chemistry code coupled to OSCAR, is a version of the PHREEQC code [7] extended to the PWR temperature range in combination with a thermodynamic database developed by the CEA [8]. PHREEQCEA calculates the values of the parameters of the OSCAR dissolution/precipitation model, i.e. the equilibrium concentration of each element in the coolant with respect to the considered oxide, the composition of the ideal solid solutions (mixed oxides) and pure solid phases (solid speciation), the pH at temperature and the dihydrogen and dioxygen partial pressures.



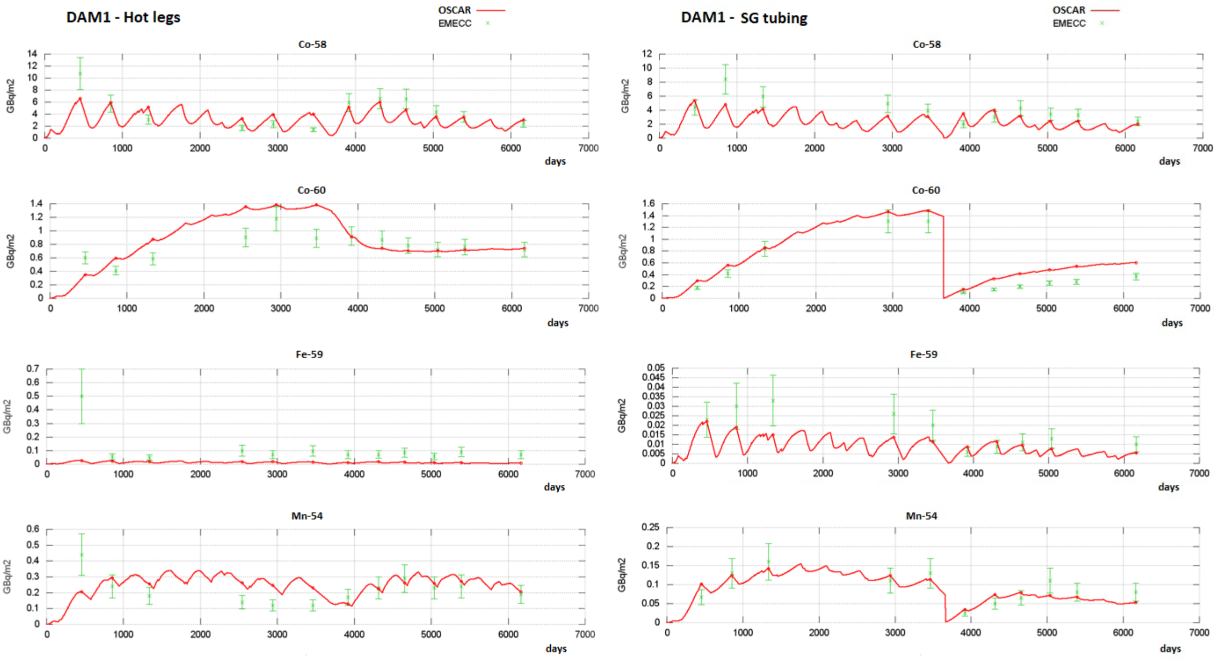
(a) (b)

Figure 5. OSCAR - Simulated operating parameters for DAMPIERRE-1 - (a) Nominal power Pn and B, Li and H_2 concentrations (b) Nominal power Pn and calculated $pH_{300°C}$

Figure 6 presents the comparison of the ^{58}Co, ^{60}Co, ^{59}Fe and ^{54}Mn surface activities calculated using OSCAR V1.4 with the ones measured at the very beginning of several outages (cycles 1 to 3, 6 to 13 and 15) of DAMPIERRE-1 using the EMECC system for the hot legs and for the SG tubing cold side.

The simulation correctly reproduces the levels and the trends of the surface activities of the primary system before and after SGR [9]:

- Before SGR, the ^{58}Co surface activity tends to decrease with cycles. After SGR, it increases for two cycles and then decreases.
- Before SGR, the ^{60}Co surface activity increases for the first 7 cycles and then stabilizes. After SGR, it is stagnant or even decreases on the primary pipes and it increases at a lower rate than before SGR on the new SG tubing.



(a) (b)

Figure 6. DAMPIERRE-1 – OSCAR V1.4 calculation/EMECC measurements comparison for ^{58}Co, ^{60}Co, ^{59}Fe and ^{54}Mn activities deposited on the hot legs (a) and the SG tubing (b).

For AFPs, to illustrate the capacity of the OSCAR code to calculate their activities correctly, the result of the simulation of CATTENOM-3 is compared to on-site measurements.

CATTENOM-3 is a 1300 MWe PWR in which 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 (cycles 9 and 10).

Figure 7 presents the comparison of the ^{134}I primary volume activity during several power operating cycles calculated using OSCAR V1.4 with the one measured, considering or not uranium dissolution in reducing conditions for the OSCAR calculations. Due to its short half-life, ^{134}I primary activity can be used as a tracer of fissile material under neutron flux during the disseminating and subsequent cycles. It comes mainly from ^{235}U and ^{239}Pu fissions in the deposited fissile material.

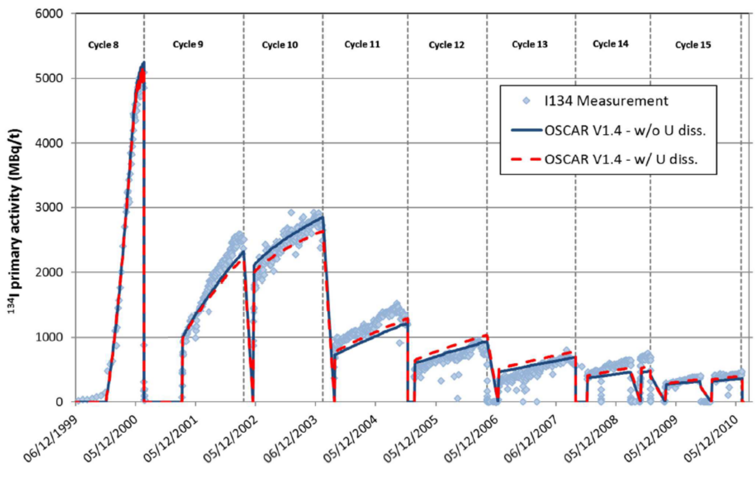


Figure 7. CATTENOM-3 – OSCAR V1.4 calculation/Measurement comparison for ^{134}I primary activity during power operating cycles.

The OSCAR results are close to the ^{134}I primary activity measurements. 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 mainly due to the evolution of the fissile material under neutron flux and to the transfer from the out-of-core surfaces due to erosion/deposition mechanisms (for further details see [10]).

3 OSCAR for decommissioning

As the OSCAR code has been originally devoted to an industrial objective, which is the reduction of the ORE for operating PWRs, the development of the OSCAR code has been focused on the main gamma emitting radionuclides such as ^{60}Co, ^{58}Co, ^{110m}Ag or ^{124}Sb. These radionuclides and their relatively short half-life are generally not relevant for decommissioning concern, which requires to know the activities of LLRNs to be declared and compared with the radwaste acceptance criteria of disposal facilities.

Nevertheless, the OSCAR code can already calculate the behavior of LLRNs of some ACPs and AFPs. For the ACPs, the OSCAR V1.4 code takes into account the following radionuclides of interest for decommissioning: ^{55}Fe (activation product of Fe, half-life of 2.7 y[d], X emitter), ^{59}Ni (activation product of Ni, half-life of 7.6x10^{4} y, X emitter), ^{63}Ni (activation product of Ni, half-life of 101 y, pure β emitter), ^{93}Zr (activation product of Zr[e], half-life of 1.6x10^{6} y, β emitter) and ^{108m}Ag (activation product of Ag, half-life of 438 y, γ emitter) and of course ^{60}Co (activation product of Co, half-life of 5.3 y[d], γ emitter).

As it is generally difficult to measure LLRNs, the most common radiological characterization method is the scaling factor approach, which consists to use ratios of activities of easy-to-measure nuclides (generally ^{60}Co for ACPs and ^{137}Cs for AFPs, the so-called key nuclides) and of difficult-to-measure nuclides such as ^{55}Fe or ^{63}Ni as ACPS, ^{135}Cs or ^{141}Am as AFPs. The capacity of the OSCAR code to assess the RCS and CVCS contamination in the light of the scaling factor approach was demonstrated for WWER-440 type reactors in [11]. Calculated scaling factors for ACPs by using OSCAR are comparable with measured ones (see [11] for further details).

To show the contribution of the OSCAR code to the contamination assessment of a 900 MWe PWR to be dismantled, the surface activities of some radionuclides of interest for decommissioning have been calculated using OSCAR V1.4. The simulated PWR (see the RCS and CVCS nodalization in Figure 1) is equipped with Inconel 600 SG tubes (Co content of 300 ppm). The first 20 cycles are simulated and each cycle lasts 282 days with a shutdown of 25 days (shutdown of 75 days at cycle 10). The simulated chemistry is a coordinated B-Li chemistry at aimed $pH_{300°C}$ of 7.2 and the hydrogen concentration is 30 mL/kg. One-third of the fuel elements is reloaded at the end of each cycle. From the 5th cycle, the fuel assembly grids made of 718 alloy are replaced by Zircaloy grids. The CVCS purification flowrate is 22 t/h with a purification efficiency of 99%.

For the easy-to-measure radionuclides such as ^{58}Co (activation product of Ni, half-life of 71 d, γ emitter), ^{60}Co and ^{54}Mn (activation product of Fe, half-life of 312 d, γ emitter), the calculated surface activities of the 900 MWe PWR type reactor using OSCAR V1.4 are within the range of the measured surface activities of the French PWR fleet using the EMECC device (see [4] for further details).

Because of the difficulty to measure them, few or no measurements of LLRNs deposited on the out-of-flux surfaces of PWR systems are available. Nevertheless, for ^{63}Ni and ^{55}Fe some measurements were performed.

Figure 8 compares the calculated ^{63}Ni and ^{55}Fe surface activities on the SG tubing of a 900 MWe PWR using OSCAR V1.4 with the measured ones on SG tubes extracted from the replaced SGs of two 900 MWe PWRs (SGR at cycle 10 for one and 11 for the other). The comparison for ^{60}Co between the OSCAR simulation and the EMECC measurements on the French 900 MWe PWR fleet is also presented in Figure 8 and the calculated ^{59}Ni as well.

The calculated ^{63}Ni, ^{55}Fe and ^{60}Co surface activities on the SG tubes are close or even equal to the measured ones, showing that OSCAR calculates these radionuclides well. ^{59}Ni was not measured for the SG tubes of the 2 units, but considering that the calculated ^{63}Ni activity is right and that ^{59}Ni and ^{63}Ni are 2 radioisotopes of the same element (Ni) and are formed by the activation of the same element (Ni), the calculated ^{59}Ni activity of about 1 MBq/m^{2} is very likely the right level.

[d] ^{55}Fe and ^{60}Co are not LLRNs but their contribution to the activity (and dose rate for ^{60}Co) at the beginning of the radwaste repository is high.

[e] ^{93}Zr could be a FP as well.

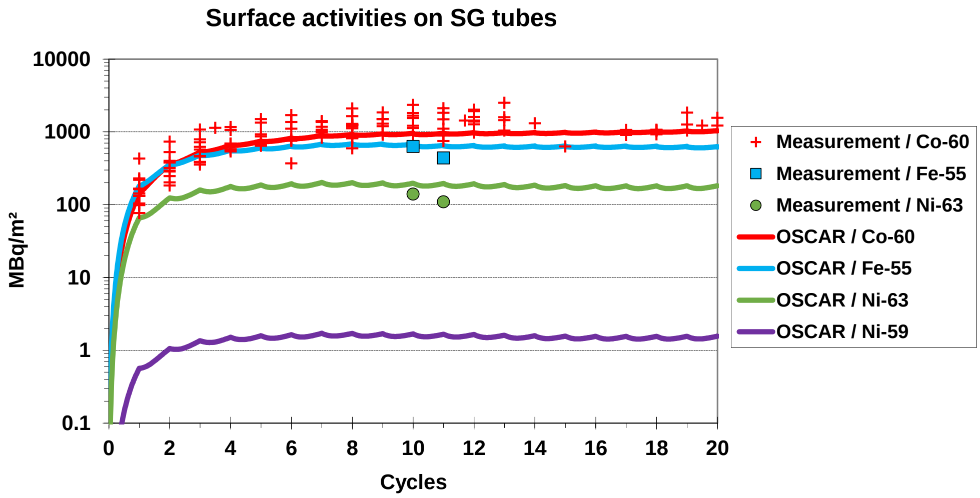


Figure 8. OSCAR V1.4 calculation/Measurements comparison for ^{60}Co, ^{55}Fe and ^{63}Ni surface activities on SG tubes.

The OSCAR code also allows to calculate the total surface activities. Figure 9 presents the total activities deposited on the CVCS and RCS out-of-flux components. The ^{59}Ni activity level is the lowest one, but it remains at this level for a dozen of thousand years and after a repository of 1000 years, it is about 10 times higher than the ^{63}Ni activity.

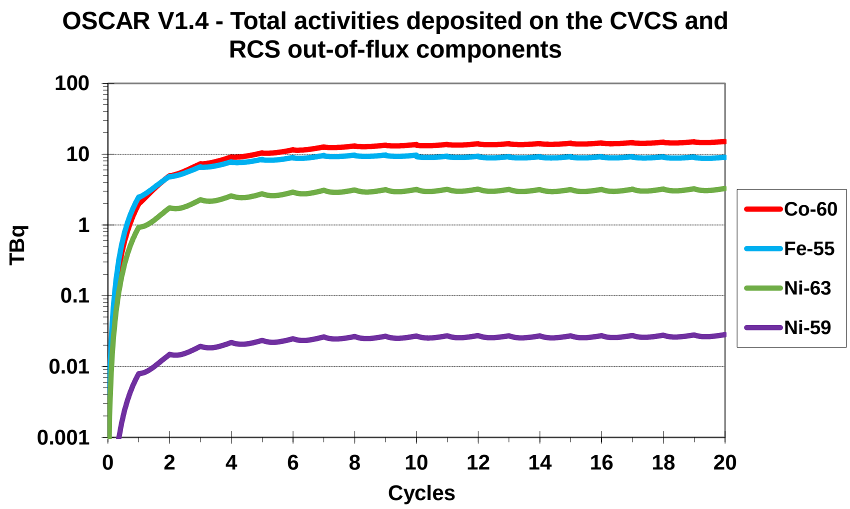


Figure 9. OSCAR V1.4 - Total surface contamination of the CVCS and RCS out-of-flux components of a 900 MWe PWR for ^{60}Co, ^{55}Fe, ^{59}Ni and ^{63}Ni.

For FPs, the OSCAR code calculates the activities of 78 nuclides within 27 filiation chains, including the relatively LLRNs of I (^{129}I) and Cs (^{134}Cs, ^{135}Cs and ^{137}Cs), which are volatile. However, iodine and cesium are considered to be fully soluble in the primary coolant and not to interact with the primary walls. If this assumption is valid in reducing conditions during power operation, it may not be the case in other conditions such as in oxidizing conditions during shutdown (adsorption of I). On the other hand, the activity of ^{90}Sr in the primary coolant and the one adsorbed on the walls are computed in the OSCAR code (see Figure 10 and [1] for further details).

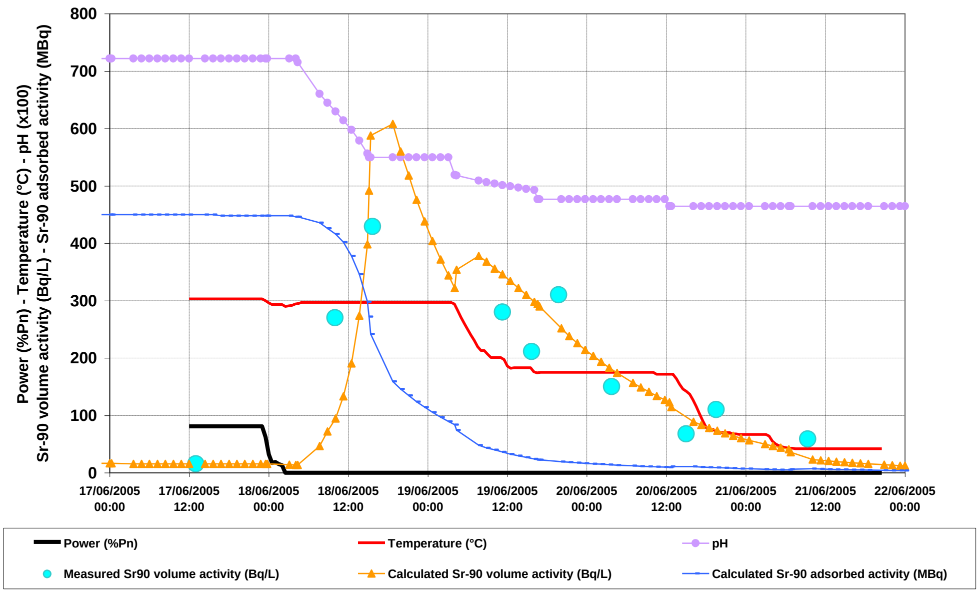


Figure 10. OSCAR calculation/Measurement comparison for ^{90}Sr primary volume activity during end of cycle shutdown (from [1]).

For actinides, the OSCAR V1.4 code calculates the activities deposited on the primary system in case of fuel oxide dissemination (see Figure 7 and [10] for further details). A 22-nuclide filiation chain is considered, including the 6 main α emitters: ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am, ^{242}Cm and ^{244}Cm.

For CAPS, the two very short-lived radionuclides calculated using the OSCAR V1.4 code (^{16}N and ^{41}Ar) are irrelevant for decommissioning.

As shown, several LLRNs are already taken into account in the OSCAR code, however, it is not the case for some of them important for decommissioning, such as ^{94}Nb (ACP), ^{151}Sm (FP), ^{99}Tc (ACP/FP) or ^{14}C (CAP). But thanks to its modularity, it is possible to add new chemical elements and their long-lived radioisotopes in the OSCAR code. To do so, this consists in:

- Identifying the source mechanism: corrosion/release, fuel release, coolant activation.
- Identifying their behavior and the corresponding data in the primary coolant in power operating and shutdown conditions: dissolution/precipitation (solubility), adsorption/desorption (equilibrium ratio), deposition/erosion (zeta potential [12][f])… It is generally the most difficult point.
- Determining the neutronic data: activation rate, filiation chain, branching ratio, capture cross section, decay constant.
- Adding the new data in the databases of the OSCAR/PHREEQCEA codes and programming a new transfer mechanism if required.
- Comparing the results of OSCAR calculation with measurements. This final stage can be also a difficult point if few or even no measurements are available.

4 Conclusion

The OSCAR code is a powerful simulation tool to assess the contamination state of nuclear systems by ACPs, AFPs and CAPs. Thanks to the EMECC measurements, its validation experimental database for PWRs is very large. The OSCAR code is generally used for reactors in operation or to be commissioned. Nevertheless, for radwaste management and decommissioning purposes, it can treat several long-lived ACPs or AFPs, such as ^{63}Ni, ^{90}Sr or actinides, therefore allowing to reduce the number of measurements of LLRNs, which are generally complicated (smears, scrapings, chemical separation processes, sophisticated laboratory equipment…). Even for the most common radiological characterization method,

[f] A comprehensive transport model for colloids is being developed for the OSCAR code.

which is the scaling factor approach, the OSCAR code can be useful to determine the activity ratios. The OSCAR code does not treat all relevant LLRNs yet, however, thanks to its modularity, it is possible to add missing chemical elements and their long-lived radioisotopes.

The OSCAR code can thus allow a plant operator to reduce the ORE and the decommissioning costs.

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