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Modelling of the contamination transfer in nuclear reactors: The OSCAR code - Applications to SFR and ITER

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1 INTRODUCTION

Predicting the radioactive contamination of nuclear reactor circuits is a significant challenge for plant designers and operators. The major stakes are to decrease personnel exposure to radiations, to optimize plant operation, to limit activity of wastes and to prepare decommissioning. To address this challenge, the French strategy has been focusing on performing experiments in test loops representative of Pressurized Water Reactor (PWR) conditions, measuring the PWR contamination and developing a simulation code namely OSCAR (Outil de Simulation de la Contamination en Réacteur – tOol of Simulation of Contamination in Reactor). The OSCAR code has been developed by the CEA in collaboration with EDF and AREVA NP for more than 40 years. It has resulted from the merging of two former codes [Genin *et al.*, 2010]: PACTOLE for Activated Corrosion Products (ACPs) and PROFIP for fission products and actinides [Beslu and Leuthrot, 1980]. Thus, the OSCAR code has been designed to predict the contamination transfer in PWR reactor coolant systems.

In the same way, ACPs in the Primary Heat Transfer System (PHTS) of the International Thermonuclear Experimental Reactor (ITER) and in the primary system of Sodium-cooled Fast Reactors (SFRs) can be a major concern as contributors to the radioactive source term of potential released activities to the environment in case of accident and to the Occupational Radiological Exposure (ORE) during the normal operation and decommissioning. Although the design and operating conditions of these reactors are different from those of PWRs, an application of the OSCAR code to SFRs, so-called OSCAR-Na, and another one to ITER, so-called OSCAR-Fusion, have been developed to treat the behaviour of corrosion products thanks to the modularity of the OSCAR code.

After a presentation of the OSCAR code, the specificities of OSCAR-Fusion and OSCAR-Na are described. Issues and R&D needs related to the modelling of the ACP transfer in these nuclear reactors are proposed.

2 THE OSCAR CODE

The process governing the ACP contamination of a PWR primary system involves many different mechanisms that react with each other. The source term is the uniform corrosion of metallic alloys which leads to the formation of an oxide layer. This oxide layer limits ion diffusion but does not eliminate them: ions are directly released in the primary coolant. Ions, generated by oxide dissolution as well, are transported by the coolant. When the coolant becomes supersaturated in corrosion products, ions precipitate on the walls or on particles in the bulk. Particles are also generated by erosion processes. Transported by the primary coolant, particles are deposited on the walls. Two types of ACP formation coexist. On the one hand, the activation of corrosion products occurs when they are deposited on the under-neutron flux surfaces. On the other hand, the corrosion of structural materials under neutron

flux is accompanied by a direct release of ACPs. Then, ACPs transported by the coolant contaminate the primary and auxiliary systems.

The OSCAR code is devoted to model this phenomenology, which is common to other types of nuclear reactors.

2.1 TRANSFER MODELLING

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

- The systems are discretized into as many control volumes or regions as necessary, defined according to their geometric, thermal-hydraulic, neutronic, material and operating characteristics.
- Six media can be defined in each control volume: metal, inner oxide, outer oxide/deposit, particles, ions and filter (ion exchange resins and particle filter).
- The following elements Ni, Co, Fe, Cr, Mn, Ag, Zn and Zr and their corresponding radioisotopes can be taken into account.
- A system of mass balance equations are calculated for each isotope (stable and radioactive) in each medium of each region using the following equation:

$$\frac{\partial m_i}{\partial t} = \sum_{Source} J_m - \sum_{Sink} J_m \quad (1)$$

where m_i is the mass of the isotope (i) in a given medium [kg], t is the time [s] and J_m is the mass flux between 2 media, 2 regions or 2 isotopes [$\text{kg}\cdot\text{s}^{-1}$].

The main transfer mechanisms taken into account are: corrosion-release, dissolution, precipitation, erosion, deposition, convection, purification, activation and radioactive decay. The first five mechanisms are described hereafter.

Corrosion of the base metal causes the formation of an inner oxide layer (mainly a chromite), of an outer oxide (a ferrite + metal Ni° or NiO in general) and a direct ion release into the bulk. The corrosion and release rates [$\text{kg}\cdot\text{s}^{-1}$] are given by:

$$J_{Cor}^{elt} = V_{Cor} \cdot S_w \cdot \alpha_{met}^{elt} \quad \text{and} \quad J_{Rel}^{elt} = V_{Rel} \cdot S_w \cdot \alpha_{rel}^{elt} \quad (2) \ \& \ (3)$$

where V_{Cor} and V_{Rel} are the surface corrosion and release rates [$\text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$] calculated by an empirical model as a function of chemistry, temperature and material (or by a power law, logarithmic law or constant value per stage), S_w is the wet surface area [m^2], α_{met}^{elt} is the element fraction in the metal and α_{rel}^{elt} is the element fraction involved in the release.

Dissolution of an oxide occurs when the concentration of a soluble species in the coolant is lower than its equilibrium concentration. The dissolution flux of the element elt , J_{dissol}^{elt} [$\text{kg}\cdot\text{s}^{-1}$] may be written:

$$J_{dissol}^{elt} = \frac{S_w}{1/h + 1/V_{dissol}} \cdot (C_{equil}^{elt} - C^{elt}) \quad (4)$$

where h is the mass transfer coefficient of ions in the fluid [$\text{m}\cdot\text{s}^{-1}$], V_{dissol} is the dissolution velocity (dissolution surface reaction rate coefficient) [$\text{m}\cdot\text{s}^{-1}$], C_{equil}^{elt} is the equilibrium concentration of the element elt [$\text{kg}\cdot\text{m}^{-3}$] and C^{elt} is the bulk concentration of the element elt [$\text{kg}\cdot\text{m}^{-3}$].

The equilibrium concentration of each chemical element and the oxide speciation are calculated by the OSCAR chemistry module, PHREEQCEA (a version of the PHREEQC code [Parkhurst and Appelo, 2013] extended to the PWR temperature range) in combination

with a thermodynamic database developed by the CEA [Plancque *et al.*, 2011]. PHREEQCEA determines the composition of the ideal solid solution (mixed oxides and any pure solid phases possibly in excess) and the equilibrium concentration of each element in relation to the chemical conditions (pH, H₂, O₂), the coolant temperature and the masses of the metallic element of the outer oxide/deposit or the inner oxide in each control volume.

Soluble species precipitate when their concentration in the coolant reaches their equilibrium concentration. The expression of the precipitation flux is similar to that of the dissolution flux.

Erosion of a deposit results from the coolant friction forces. The erosion flux, J_{eros} [kg.s⁻¹], is given by:

$$J_{eros} = \frac{E}{\Psi} \cdot m_{erod} \quad (5)$$

where E is the erosion coefficient [s⁻¹] based on the [Cleaver and Yates, 1972] model and depending on the shear stress at the wall and the dynamic viscosity of the coolant, Ψ is the erosion resistance [-] and m_{erod} is the mass of the deposit that can be eroded [kg].

Deposition rate of particles takes into account turbulent diffusion [Beal, 1970], sedimentation, thermophoresis and boiling deposition [Ferrer, 2013]. The deposition flux, J_{depos} [kg.s⁻¹], may be expressed as follows:

$$J_{Deposition} = S_w \cdot V_{depos} \cdot C^{part} \quad (6)$$

where V_{depos} is the deposition velocity of particles [m.s⁻¹] and C^{part} is the particle concentration in the fluid [kg.m⁻³].

2.2 CALIBRATION AND VALIDATION

As the dissolution velocities and the erosion coefficient are not well known in PWR conditions, they are calibrated.

The validation of the OSCAR code is based on a large operating experience feedback unique in the world: the so-called EMECC campaigns (about 400 campaigns of the γ surface activity measurements in 72 different PWRs in France and abroad since 1971 using the EMECC device [Eimecke and Anthoni, 1988]) [Dacquait *et al.*, 2012].

The last version of OSCAR, OSCAR V1.4, is validated in power operating and cold shutdown conditions for PWR primary and auxiliary systems, thus covering a large range of chemical and thermo-hydraulic conditions (water temperature from 20 °C to 350 °C, laminar and turbulent, reducing and oxidizing, alkaline and acid conditions).

3 OSCAR APPLICATION TO ITER: OSCAR-FUSION

The PHTSs (IBED, NBI, VV) of the ITER Tokamak Water Cooling System (TCWS) are also subject to contamination by ACPs. Indeed, a part of these systems are under neutron flux and their materials (mainly stainless steel and copper alloy) corrode.

Thanks to similarities between ITER PHTSs and PWR primary and auxiliary systems, mainly the coolant, since 1995 the PACTOLE code and then the OSCAR code have been used to calculate the ACP contamination transfer in the ITER PHTSs. Nevertheless, some differences, mainly the presence of CuCrZr alloy for the plasma facing components and the neutron flux, involved adaptations of PACTOLE and OSCAR, called PACTITER [Luigi *et al.*, 1998] and OSCAR-Fusion respectively. OSCAR-Fusion is currently the reference version, in particular thanks to its powerful chemistry module PHREEQCEA.

3.1 TRANSFER MODELLING

As the coolant of ITER PHTSs and PWRs is the same, the transfer modelling of the OSCAR-Fusion code is the same as the one of the OSCAR code (see section 2.1).

Cu data was added in PHREEQCEA thermodynamic database based on an experimental campaign performed by CEA to obtain Cu solubility data for ITER divertor PHTS conditions [Luigi *et al.*, 1998]. The three following nuclear reactions involving Cu, found to be the most important ones for safety and ORE analysis, were then included in OSCAR-Fusion: $^{63}\text{Cu}(n, \gamma)^{64}\text{Cu}$; $^{65}\text{Cu}(n, 2n)^{64}\text{Cu}$; $^{63}\text{Cu}(n, \alpha)^{60}\text{Co}$. For the other most important corrosion products (Ni, Fe, Co, Cr and Mn), the activation rates were calculated by considering the ITER fast neutron flux.

3.2 CALIBRATION AND VALIDATION

In order to get realistic values of release rates, experiments were carried out using the CEA CORELE loop in thermohydraulic and chemical operating conditions envisaged for ITER PHTS [Luigi *et al.*, 2007].

Nevertheless, the validation of OSCAR-Fusion is based on the validation of OSCAR, which covers a large range of conditions (see section 2.2).

As for the ITER PHTSs, OSCAR-Fusion can be used to predict the ACP contamination transfer in the water cooling system of DEMO (DEMONstration power station) or of other nuclear fusion reactors.

3.3 ISSUES AND R&D NEEDS

In addition to the “Corrosion issues in thermonuclear fusion reactors and facilities” detailed by N. Baluc [Feron, 2012], the following issues and R&D needs related to the ACP transfer and its modelling in a simulation code such as OSCAR-Fusion can be highlighted:

- Simulation of the pulsed mode within a reasonable time for each ACP whatever their radioactive half-life (short or long). The pulsed mode is a succession of burn, hot and cold stand-by, baking and shutdown phases (about 400,000 burn phases of 400 s over about 20 a).
- Validation of OSCAR-Fusion against experiments in ITER/DEMO PHTS conditions.
- Optimization of the cooling water chemistry conditioning for each PHTS and each operating phase. The water chemistry specification of each PHTS for plasma operation is based on the Boiling Water Reactor water chemistry, which is mainly controlled by its cation conductivity [Gopalapillai *et al.*, 2012], rather than on the PWR water chemistry, which is mainly controlled by its pH and H₂ concentration, for instance [Fruzzetti *et al.*, 2014].
- Determination of the corrosion rates of the different materials (stainless steels, Cu alloys and the impact of Cu swirls, RAFM steel) in the different conditions.
- Assessment of the impact of manufacturing processes (surface finish) of the materials, which is a key parameter for the contamination level [Dacquait *et al.*, 2006].
- Assessment of the impact of the corrosion at different material junctions like CuCrZr and stainless steels.
- Assessment of the fraction or the quantity of ACP deposit released from the loop after a large LOCA

4 OSCAR APPLICATION TO SFR: OSCAR-NA

The OSCAR-Na code has been developed to calculate the contamination by corrosion products in the primary circuit of SFRs [Genin *et al*, 2016].

In SFRs, contamination of the primary circuit is due to the release of radioactive nuclides produced in the reactor core by neutron reactions on the constituent elements of the stainless steel fuel cladding and subassembly wrappers. Due to both bulk corrosion (i.e. surface loss) of the activated cladding and preferential release of highly soluble elements, a small fraction of the radionuclides is released into the flowing sodium and deposits throughout the primary circuit. This leads to contamination of the piping and components such as primary pumps or Intermediate Heat eXchangers (IHX). The main radio-contaminants are ^{54}Mn and ^{60}Co .

4.1 TRANSFER MODELLING

The OSCAR-Na code is based on the OSCAR code. For each region and each isotope considered, a mass balance is calculated in the sodium and in the metal. The modeling assumes that the transfer between steel and sodium is primarily by solution and precipitation of metallic elements, rather than by particle detachment and particle deposition.

The solution/precipitation model at the steel/sodium interface is based on mass transfer theory [Poley and Skyrme, 1978]. The mass flux Φ between sodium and steel is obtained by equating the fluxes at both sides of the interface:

$$\Phi = D \cdot \left. \frac{\partial C}{\partial x} \right|_{x=0} + u \cdot C_i = K^{eff} \cdot \left(\frac{C_i}{\beta} - C' \right) \quad (7)$$

where C and C' are the nuclide concentration (g/m^3) in the steel and bulk sodium, respectively (the subscript i indicating interfacial values at $x = 0$). In OSCAR-Na, C' is calculated through a complete mass balance around the primary circuit, and C_i is obtained by numerical solving of the general diffusion equation in the steel:

$$\frac{\partial C}{\partial t} = D \cdot \left(\frac{\partial^2 C}{\partial x^2} \right) + u \cdot \frac{\partial C}{\partial x} - \lambda \cdot C + R \quad (8)$$

with eq. (7) as boundary condition at the interface (λ is the decay constant of the considered radionuclide, and R is the rate of production by neutron activation).

Thus, the main parameters of the model are, for each element:

- D (m^2/s): diffusion coefficient in the austenitic stainless steel. A higher diffusion coefficient can also be considered in the ferrite layer which forms near the steel surface due to nickel depletion.
- K^{eff} (m/s): effective mass transfer coefficient between interface and bulk sodium. It corresponds to the limiting value between the diffusion rate through the sodium laminar boundary layer (k) and the dissolution or precipitation rate (k_a) at the interface: $K^{eff} = \frac{k \cdot k_a}{k + k_a}$.
- β : dimensionless chemical partition coefficient. It is related to the concentration $C'_{\acute{e}q}$ in the sodium at equilibrium with the steel surface by $C'_{\acute{e}q} = C_i / \beta$. For a pure element, β varies inversely with respect to the solubility.

- u (m/s) is the interfacial velocity or moving rate of the sodium/steel interface, due to dissolution and precipitation. It is positive or negative in case of bulk corrosion (dissolution) or bulk deposition (precipitation) respectively.

The other transfer models considered in the OSCAR-Na code (convection, purification, neutron activation and radioactive decay) are the ones of the OSCAR code for PWR.

The OSCAR-Na code uses a numerical method for solving the diffusion equation in the steel and the complete mass balance in sodium for all elements, allowing the calculation of the metal/sodium interface shifting and of the flux of each element through this interface.

4.2 CALIBRATION AND VALIDATION

The key parameters (D , K^{eff} , β and u) of the solution/precipitation model have been assessed from a literature review or adjusted to match some experimental data.

The simulation of the French SFR reactor PHENIX using OSCAR-Na was able to assess the correct amount of contamination and the correct contamination profiles on heat exchanger surfaces for ^{54}Mn , ^{58}Co and ^{60}Co radionuclides, compared with measurements (see Figure 1, further details in [Genin *et al.*, 2016]). And the simulation of tests performed in the STCL loop confirms OSCAR-Na modeling ability to calculate weight losses due to corrosion of 316 SS in sodium circuits (see [Genin *et al.*, 2017]).

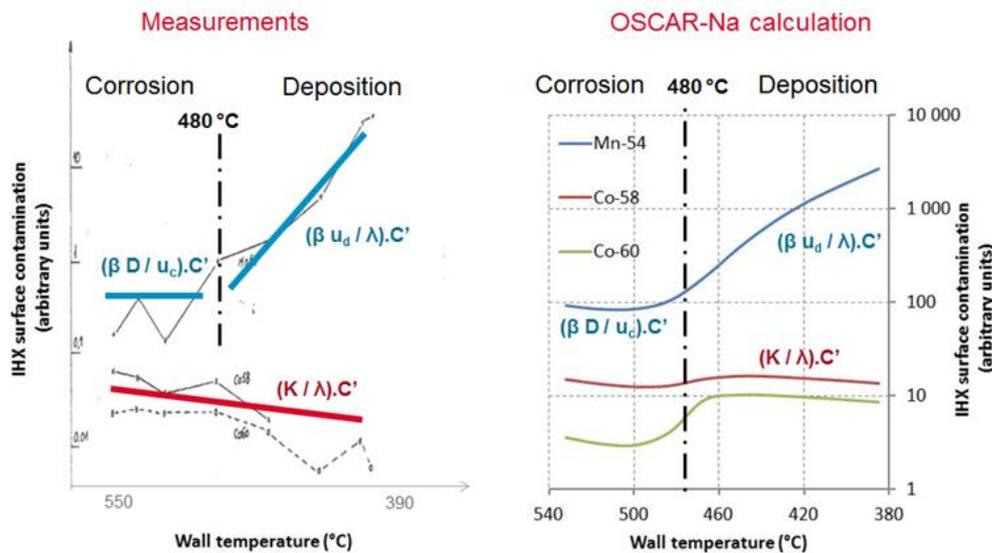


Figure 1 : Comparison measurements/OSCAR-Na calculation - PHENIX reactor IHX contamination.

4.3 ISSUES AND R&D NEEDS

In addition to the “Corrosion issues in sodium-cooled fast reactor (SFR) systems” detailed by E. Yoshida and T. Furukawa [Feron, 2012], the following issues and R&D needs related to the contamination transfer and its modelling in a simulation code such as OSCAR-Na can be highlighted:

- Obtaining of data on oxide equilibrium concentrations (only pure element solubility in sodium are known) and on co-precipitation.
- Obtaining of further data on diffusion coefficient in steel.
- Assessment of the impact of the ACP transfer under particle form.

- Carrying out of further OSCAR-Na validation work against operating experience with SFRs and experimental loops.
- Modelling of the contamination by fission products.

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