**Materials and Corrosion in Light Water Reactors**

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Abstract – *Since the beginning of nuclear industry, corrosion issues have been a major concern. Less than two years after start-up, stress corrosion cracking occurred on the stainless steel tubing of the steam generator of the prototype for the Nautilus in USA (1953); more recently, last year in 2022, several French Pressurized Water Reactors were shut down due to stress corrosion cracking of stainless steels pipes in a safety injection circuit. We propose to underline the complex corrosion mechanisms linked to the nuclear reactor environments (high temperature and high-pressure water environments) and to present briefly the three main corrosion phenomena occurring in Light Water Reactors (LWRs), after a short overview of the materials of the boiling water reactors (BWRs) and pressurized water reactors (PWRs):*

1. *general corrosion of zirconium cladding which limits the life-time of fuel elements to generally 3 cycles;*
2. *flow-accelerated corrosion (FAC) of carbon steel components, which is the only corrosion phenomenon that has led to several deaths in PWRs;*
3. *stress corrosion cracking (SCC) of nickel base alloys (“the Coriou effect”) and of stainless steels including irradiation-assisted stress corrosion cracking (IASCC); SCC phenomena has led to the replacement of major components like steam generators or pressurized vessel heads.*

*Finally, the corrosion future will be discussed as BWRs and PWRs are extending their period of operation up to 60 and 80 years, and even more.*

**Keywords:** Light Water Reactor, Zirconium, carbon steel, stainless steel, nickel base alloy, flow-accelerated corrosion, stress corrosion cracking

I. Introduction

The nuclear industry works on how to improve and optimize the safety and security, the performance and the durability of its facilities. Whatever the materials, they are altered more or less rapidly on contact with the environment in which they are used. So accurate and reliable knowledge of the behavior, strength and properties of the materials constituting these nuclear facilities over their lifetime is acquired and taken into account for meeting safety requirements. Corrosion of nuclear materials, i.e. the interaction between these materials and their environments, is a major issue for plant safety, but also for the economic competitiveness of the nuclear industry **[1]**.

 Since the beginning of nuclear industry, corrosion issues have been a major concern. Less than two years after start-up, stress corrosion cracking occurred on the stainless steel tubing of the steam generator of the prototype for the Nautilus in USA (1953) **[2]**. Since the 1950s, many efforts have been spent for selecting right materials for the various environments encountered in nuclear power plants, to develop prediction and mitigation of corrosion phenomena in nuclear systems in order to prevent failures, and to increase safety and lifetime of these systems. Over time, substantial progresses have been made towards understanding, preventing, monitoring and modeling the interactions between materials and their environments. After a brief presentation of fundamentals of corrosion phenomena and a short overview of the main materials used in nuclear power plants (NPPs), this paper underlines three main corrosion issues occurring in NPPs:

* general corrosion of fuel cladding made of zirconium alloys,
* flow-accelerated corrosion (FAC) of carbon steels in the steam/vapor circuits,
* stress corrosion cracking (SCC) of nickel base alloys (“the Coriou effect”) and of stainless steels in pure or primary waters, respectively for boiling water reactors (BWRs) and for pressurized water reactors (PWRs).

II. Overviews

Nuclear industry requires a selection of materials having outstanding properties in various and sometimes very specific environments. In addition, the irradiation leads to a selection of alloys with specific properties with respect to irradiation, which induces alloy transformation (evolutions of the microstructures, of mechanical characteristics…) and compatibility with neutron flux.

***II.A. Corrosion Basics***

Corrosion is defined as a “*physicochemical interaction between a metallic material and its environment that results in changes in the properties of the metal, and that may lead to significant impairment of the function of the metal, the environment or the technical system, of which these form a part*” **[3]**. Metals and alloys corrode because we use them in environments where they are not chemically stable, as shown by the Pourbaix diagrams **[4]**.

The definition of corrosion by ISO standard includes a note “*This interaction is often of an electrochemical nature*” **[3]**. In water, like in BWRs and PWRs, the phenomena is always an electrochemical one. It means that the overall corrosion process necessarily involves at least two simultaneous reactions: an oxidation (or anodic) reaction and a reduction (or cathodic reaction), which are coupled through the exchange of electrons. Examples of anodic (oxidation) reactions include:

Fe → Fe2+ + 2e− (1)

Cr → Cr3+ + 3e− (2)

While examples of cathodic (reduction) reactions include oxygen reduction if oxygen is dissolved in water or proton reduction in acid solutions or in solutions without oxygen or other oxidants:

O2 + 2H2O + 4e− → 4OH− (3)

2H+ + 2e− → H2 (4)

So a corrosion cell involves two simultaneous reactions (anodic and cathodic) that must proceed at the same time and the same rate, but not in the same place **[5]**.

During corrosion, the local electrochemical potential at an anode is different from that at a cathode. In addition, the local electrochemical reactions at anodes and cathodes result in significant chemical changes in their vicinity that encourage and maintain their spatial separation. Furthermore, this potential gradient attracts oppositely charged ions, a process known as electro-migration. Additionally, the requirement for electro-neutrality in the electrolyte (meaning that an anion cannot exist in solution without a corresponding cation) gives rise to diffusion in the electrolyte. To illustrate these phenomena, the overall process for corrosion of iron, with oxygen as the cathodic reactant, is shown schematically in Fig. 1.

O2

Electron flow

Cathode

Anode

O2 + 2H2O + 4e- => 4OH-

Fe => Fe2+ + 2e-

Aqueous solution

Cations (e.g. Na+)

Anions (e.g. Cl-)

*Fig. 1. Schematic diagram showing spatial separation of anode from cathode with corresponding migration of ions in solution (from S. Lyon [5]).*

Corrosion damage (e.g. metal loss) generally occurs at the anodic locations while at the cathodic locations no corrosion damage occurs. For alloys subject to general corrosion, the locations of anodes and cathodes tend to move randomly over the surface of the metal and, on average, metal thinning occurs relatively uniformly. However, for many corrosion resistant alloys, which are covered by a passive oxide film (called passive alloys), the anode tends to become strongly localized thus giving rise to localized corrosion damage: pitting corrosion, crevice corrosion, intergranular corrosion, stress-corrosion cracking…

***II.B. Nuclear Materials***

Material used in nuclear industry are generally divided into 3 main categories [6]:

* The first category concerns materials developed specifically for nuclear applications like fuel materials and neutron absorbers.
* Some specific alloys developed for nuclear applications, but used also in other industries compose a second category. It includes graphite (used as moderator in gas cooled reactor), zirconium alloys (even if zirconium alloys without hafnium is only used as clad material in nuclear power plants due to their low capture neutron cross sections), oxide dispersion-strengthened (ODS) ferritic-martensitic steels which are developed for cladding of Gen IV reactors or vanadium alloys for fusion systems.
* The third category includes standard alloys (stainless steels, aluminum alloys, nickel base alloys, titanium alloys…) which are used not only in nuclear environments. Structural alloys used in nuclear power plants, are in this third category, as those shown in the ternary diagram Fe-Cr-Ni of Fig. 2 where carbon steels, low alloyed steels, stainless and nickel based alloys are shown **[2]**.

In NPPs, these materials are subject to various degradation processes, driven by applied loadings (radiation, thermal, mechanical, chemical). In this paper, we focus on corrosion, degradation by the environment.



*Fig. 2. Ternary diagram of the main structural materials used in NPPs (from R.W. Staehle – [2])*

This document is written to give an overview of some of the materials degradation issues that may be key for reactor service. A detailed description of all the possible forms of degradation is beyond the scope of this short paper and has already been described in other documents (**[1]** for example). The intent of this short paper is to present an overview of current materials main issues regarding safety or availability in the existing reactor fleet. So, as explained already in the introduction, three distinct areas have been selected because they are major issues in PWRs and BWRs: (i) zirconium cladding, (ii) Flow Accelerated Corrosion of carbon steels and (iii) Stress Corrosion Cracking of austenitic alloys (stainless steels and nickel base alloys).

III. Zirconium cladding

The cladding material of the fuel rods used in the Light Water Reactors is a zirconium alloy (Zr), which is oxidized on contact with water (in PWR primary water: 155 bar, 300-330 °C) following the electrochemical reaction:

Zr + 2H2O → ZrO2 + 2H2 (5)

leading to the formation of zirconia (ZrO2) and hydrogen, part of which is dissolved in water and the other part may react with zirconium to form hydrates.

 Owing to its effects upon clad mechanical behavior (formation of zirconium hydrates) and upon fuel rod thermal behavior (formation of zirconia), Zr alloy corrosion is a factor that limits the in-reactor residence time of fuel assemblies. Typically, the maximum allowable oxide thickness is approximately a hundred micrometers.



*Fig. 3. Zircaloy-4 and M5TM oxidation kinetics in an autoclave at 360 °C, with 10 ppm Li and 650 ppm B in water. The kinetics appear as cyclic (reproduced from [1]).*

Zr alloy oxidation kinetics exhibit a succession of cycles (with a decreasing oxidation rate) and transitions in the kinetic curve (which bring the cycles to an end). The major difference between alloys lies in the thickness of the oxide that has grown between two transitions in the kinetic curve (the so-called transition thickness). The lower the average oxidation rate, the higher the transition thickness. For example, alloy M5TM, which displays a higher resistance to corrosion, shows the highest transition thickness (~ 3.1 µm for M5TM against ~1.7 µm for Zircaloy-4), as shown on Fig. 3 under PWR primary chemistry conditions **[1,7]**.

Zr alloy oxidation kinetics depend upon many factors including:

* The solution chemistry (pollutants, redox potential, pH…). Nevertheless, under chemical conditions very close to those observed in PWRs any slight rise of the Li content beyond the current value of 2.2 ppm, does not display a significant effect on corrosion. In contrast, under chemical conditions very far from those encountered in PWRs (no boron, high lithium - very basic conditions), a strong increase of Zr corrosion rate can rapidly takes place.
* Temperature rise accelerates oxidation kinetics: the oxidation rate is nearly twofold because of a temperature rise of 20 °C.
* Boiling on the clad wall results in a local increase of temperature and a local alteration of water chemistry leading to a local increase of the corrosion rate.
* The reactor environment, especially irradiation, strongly accelerates Zr alloy oxidation kinetics compared to kinetics obtained in laboratories.

Fig. 4 regroups and consolidates measurements of oxide thicknesses on Zr alloy fuel clads after exposure in PWR cores. For higher burnups, the thickness of the oxide films formed on Zircaloy-4 is far higher than that measured on M5TM: for a burnup higher than 55 GWd/tU, the thickness may reach 100 µm for Zircaloy-4 while it does not exceed 25 µm for M5TM.



*Fig. 4. Onsite and hot-laboratory eddy-current measurements of oxide thicknesses on fuel rod clads of Zircaloy-4 (1.3 % Sn), M5TM (Alloy 5 – 1%Nb) and Alloy 2 (0.5 % Sn) arising from PWRs (reproduced from [1])*.

The new Zr alloys (M5TM from Framatome, ZirloTM from Westinghouse) may help to increase nuclear power plant competitiveness, as they may lead to extend operating cycles, to increase the coolant temperature as well as heat flow. Nevertheless, following Hiroshima accident during which the production of hydrogen due to the oxidation at high temperature of zirconium cladding - reaction (5) – leads to explosions, main efforts are made to develop new Accident Tolerant Fuels (ATF) with new cladding materials (better behaviour at high temperatures) and new fuels (high thermal conductivity) **[8]**.

IV. Flow Accelerated Corrosion

Flow Accelerated Corrosion (FAC) is the main degradation mechanisms of the steam/water circuits **[9, 10]**. Some catastrophic failures due to FAC occurred in NPPs and lead to injuries and casualties:

* In December 1986, five workers were killed due to the FAC rupture in the condensate system of Surry Unit 2 (PWR commissioned in 1973, Virginia, USA).
* In August 2004, a piping rupture took place at a condensate line of Mihama-3 (PWR commissioned in 1976, Japan) and killed five workers and resulted in six others being injured.

Many complementary reported events of FAC degradation occurred in steam/water circuits of all types of NPPs. Large efforts have been and are still made to prevent and\or master the risks linked to the FAC.

It is important to keep in mind that FAC of carbon steel is a chemical process. There is no mechanical removal under FAC conditions. High FAC rate occurs at locations with high mass transfer rates, corresponding to highly turbulent flow conditions found at elbows, bends, valves, diaphragms…

The FAC process consists of an extension of the generalized corrosion process considering the effect of the water flow at the oxide/solution interface. In the absence of oxygen, like in PWR secondary circuits, iron is oxidized to give magnetite and hydrogen according to the following reaction:

3Fe + 4H2O → Fe3O4 + 4H2  (6)

In BWR water/steam systems where oxygen is present, the corrosion reaction leads to the formation of hematite (Fe2O3) and the reduction of oxygen. The kinetics of the corrosion reactions increase with temperature and flow rates. Therefore, FAC occurs mainly **[9, 10]**

* on steels materials (carbon steels or low alloyed steels) covered with a protective oxide layer,
* in liquid water (single or dual phased flow),
* at a minimum fluid velocity (around 0.5 m/s) because at high flow rate the dissolution of the oxide protective layer is faster which leads to a dramatic increase of the corrosion rate.

According to observations of FAC in plants and in laboratories, the main parameters on which the process depends in a critical way are the following:

- Water chemistry: FAC kinetics decrease when pH increase in PWR conditions but is not dependent of the chemicals added as alkalizing agents (ammonia, morpholine or other amines), as shown on Fig.5. Dissolved oxygen has beneficial effects (decrease of FAC kinetics) due to the fact the solubility of magnetite is much higher than the solubility of hematite.

- Temperature influence is more complex with a maximum around 150°C as shown on Fig. 5.

- Hydrodynamic or mass transfer conditions (i.e. flow velocity, surface roughness geometry, steam quality, flow pattern...): FAC corrosion rates increases with the Reynold number.

- Steel composition: the most beneficial alloying element is chromium; further alloying elements like molybdenum and copper also have a beneficial effect. An alloy steel with a nominal chromium content as low as 1% (w/w) will have low or negligible FAC rate in NPP conditions. With 0.1 % of chromium, the FAC rate is 2.5 times lower than without chromium **[9]**.

*Fig. 5. Temperature effect on the relative FAC rate of carbon steel at a pH25°C of 9.0 under morpholine and ethanolamine chemical conditionings. Results are also compared to BRT-CICERO™ calculation for ammonia in one-phase flow conditions [1].*

Nowadays, some models (*BRT-CICERO*™ in France, CHECWORKS™ in USA for instance) exist to calculate the margin of piping components. They are used in addition with non-destructive examinations to operate in secure way and to increase the availability of NPPs. For new components and new plants, it appears necessary to take into account FAC at first step of detailed design. For example, the next EPR reactor at Flamanville in France will incorporate many lines with high chromium and molybdenum contents.

V. Stress Corrosion Cracking

Stress Corrosion Cracking (SCC) is one of the most feared forms of corrosion. It appears as a cracking, the propagation of which may be relatively fast, and take place after a more or less long incubation phase, with no major precursor sign. Moreover, it often occurs in poorly aggressive environments like pure water at high temperature. Three main factors are thus involved: the material and the environment (as always in corrosion), as well as the stress, as shown by the Venn diagram of Fig. 6.



*Fig. 6. Venn diagram as an illustration of the three main factors of stress corrosion cracking (reproduced from [1])*

Nowadays, no material can be considered as sensitive or insensitive to SCC. However, materials may be more or less sensitive in a given environment. Cracking initiates when the stress goes beyond a limiting value. In addition, residual stresses should not be omitted when assessing the stress level, which justifies stress-relieving treatments systematically, performed. Cracking induces brittle fracture (or brittle failure) in materials. Cracks may be transgranular or intergranular, depending on the material and its environment **[11, 12, 13]**.

SCC of austenitic alloys (stainless steels and nickel base alloys) in NPPs has been a major issue since the beginning of the development of nuclear energy. As said in the introduction, less than two years after start-up, SCC occurred on the stainless steel tubing of the steam generator of the prototype for the Nautilus in USA (1953). More recently, last year in 2022, several French PWRs have been shut down due to stress corrosion cracking of stainless steels pipes in a safety injection circuit. It is more than interesting to look at the SCC historical perspective **[2]**. After the SCC failures of stainless steels on Nautilus prototype, H. Copson at INCO (USA) demonstrated that SCC on Fe-Cr-Ni alloys, would stop above about 40% Ni when exposed to boiling MgCl2. Therefore, he proposed to use Inconel 600® (15% Cr, 7% Fe and 78% Ni) for steam generator tubing. As soon as the choice of Alloy 600 was known for PWR steam generator tubes, the CEA’s “Corrosion Department”, headed by Henri Coriou, started to investigate the behavior of this new material in an aqueous environment at high temperature. The first observations evidenced substantial intergranular cracking on specimens after a few months’ exposure in pure water at temperatures ranging from 300 to 350 °C. This result issued in 1959 was the starting point of a controversy that was to last for about thirty years. During the 1980s, when numerous cases of cracking occurred in plants, the corrosion scientific community acknowledged the reality of what is now named the “Coriou effect”. In fact, quite rapidly Coriou found that in pure water or in primary water at high temperatures, the susceptibility of nickel base alloys was increasing with the nickel content as shown on Fig. 7 **[1, 2]**.



*Fig. 7. Stress corrosion cracking susceptibility of austenitic alloys as function of the nickel content in pure water or in 0.1% NaCl solution at 350°C (from H. Coriou and R.W. Staehle, [2]).*

Nowadays, Alloy 600 is no more used in new PWRs or replaced equipment like steam generators. Alloy 690 is the alloy used now (Alloy 800 too) instead of Alloy 600 **[2, 14]**. It exhibits a lower amount of nickel and higher amount of chromium, as shown on Fig. 7.

In Boiling Water Reactors (BWRs), SCC affected austenitic stainless steels in the reactor coolant pipe as early as the 1950-1960s **[15]**. This cracking phenomenon was mainly due to the conjunction of the several parameters: residual stresses in welded areas, sensitization due to chromium carbide intergranular precipitation, and occurring residual oxygen contents coupled with radiolysis in the BWR environment that only consists of high-purity water.

In PWRs, first cracking cases observed on 304L and 316L stainless steels have been related to pollutants in the primary coolant circuit, mainly oxygen, chlorides, and sulphates **[16]**. Austenitic stainless steels prove to be susceptible in presence of these pollutants and even more susceptible in the case of a mixed pollution of chlorides/sulphates’ (even for moderate contents), and in presence, simultaneously, of dissolved oxygen. Diagrams like the one on Fig.8, display the SCC susceptibility ranges of stainless steels in terms of dissolved pollutants.



*Fig. 8. Ranges of susceptibility to stress corrosion cracking of stainless steel in a primary environment containing dissolved sulphates and chlorides, and in presence or absence of oxygen (reproduced from [1]).*

In the late nineties, a new case of SCC started to affect stainless steels in a primary environment with a nominal composition, but in the case of materials strongly cold-worked surfaces. It is generally admitted that a sensitivity threshold in terms of initial hardness can be observed around 300 Hv on 304L steel. This threshold also corresponds with that observed in the case of cold-worked materials exposed in BWRs **[12, 15]**.

Irradiation is also a major factor that may increase the SCC susceptibility. Such phenomena is called Irradiated Assisted Stress Corrosion (IASCC). IASCC a multi-physical phenomenon, as illustrated in Fig. 9. For not only the three major promoters of stress corrosion cracking (the environment, the material and the stress), are to be considered, but also the irradiation effects on the material (creep, swelling, intergranular segregation…) and on the environment (radiolysis) **[17]**.



*Fig. 9. Various phenomena likely to affect an irradiated material subjected to stress, and in chemical interaction with the environment (reproduced from [1])*

Following the occurrence of SCC on French PWRs in 2022 **[18]**, R&D works are needed in order to increase knowledge and understanding of SCC crack initiation conditions and SCC propagation kinetics for austenitic stainless steels in nominal primary environment. It will be a major step in the assessment of this type of risk for the whole nuclear power reactor fleet.

IV. Conclusions

Corrosion is an important issue for almost all nuclear facilities. Reactor safety and fuel cycle plants are highly dependent on this issue. NPPs economic competitiveness is also deeply concerned: current stakes are particularly high for extending the reactor lifetime to sixty years and beyond, that is twice the time initially scheduled for the previous generation’s reactors. For Generation IV reactors, the minimum exploitation time is 60 years with higher temperatures. As the ageing of these facilities is strongly governed by corrosion, the prediction of initiation and propagation of corrosion phenomena over large periods become a major issue. Research and development actions are needed to achieve the goal. Modelling and simulation of corrosion phenomena are major tools to predict the behavior of alloys aver long periods. Robust and validated quantitative models are needed and underdevelopment with a view to understanding and modelling physico-chemical phenomena at various time and space scales, from atomistic simulation to structural simulation.

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