

## Corrosion issues in pressurized water reactors: achievements and challenges

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## CORROSION ISSUES IN PRESSURIZED WATER REACTORS: ACHIEVEMENTS & CHALLENGES

#### NUPP 2017

1<sup>st</sup> International Conference on Nuclear Power Plants: Structures, Risk & Decommissioning Croydon, London, UK / 6<sup>th</sup> – 8<sup>th</sup> February 2017

#### Damien Féron

Service de la corrosion et du comportement des matériaux dans leur environnement Commissariat à l'énergie atomique et aux énergies alternatives *French atomic energy and alternatives energies Commission* Saclay, France



## CONTENT

□ Introduction: background

□ Corrosion issues (Pressurized Water Reactor – PWR)

- □ (Fuel cladding)
- **Primary circuit**
- □ Steam generator tubbing
- □ Secondary circuits

Steam generator Instance control Pressuriser Boiling water Het colant 120 °C1 Nuclear reactor Nuclear reactor

□ Conclusion: multiscale modelling



## **NUCLEAR MATERIALS: from yesterday to tomorrow**





## MAIN ALLOYS USED IN PWRS

### **Cladding: Zirconium alloys**

## Structural alloys: steels and nickel base alloys



## Evolution of the alloys due to corrosion issues



## CORROSION ISSUES AND PWR SAFETY BARRIERS

# 1- Fuel cladding : corrosion of zirconium alloys limits the residence time of fuel materials

2- Reactor coolant boundaries : pressure vessel (irradiation damages) is limited by irradiation damages, while stress corrosion cracking of the nickel base alloys tubes of the SG limits the life time of steam generators





**3- Reactor containment**: concrete and reinforced concrete evolution (repairs possible)

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## **OPERATIONAL EXPERIENCE & CORROSION**

#### Events of Interest for Safety of French PWRs (IRSN, 2010)



Most degradation is caused by Flow Accelerated Corrosion (FAC). Stress Corrosion Cracking (SCC) is highly generic and has a large impact on safety because it appears in the second containment barrier.

Main Degradation Mechanisms leading to NPPs shutdown (R. Killian, 1995-2005)



Two main dominant mechanisms:

- Flow accelerated corrosion (FAC) and
  - Stress Corrosion Cracking (SCC)
    - Pipes and internals
      - SG tubes

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CONTROL ROD

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# PRIMARY CIRCUIT :

### **PIPES & INTERNALS**

UPPER COR PLAT

CORE SUPPORT

STRUMENTATION THIMBLE GUIDES

## PRIMARY CIRCUIT: PIPES AND INTERNALS



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### FIELD EXPERIENCE OF AUSTENITIC STAINLESS STEELS IN PWR PRIMARY WATER (SCC)

Practical experience shows that de-oxygenated, hydrogenated PWR primary water does not cause SCC. The reason is clearly related to the dissolved hydrogen in PWR primary circuits, which ensures that any radiolytic decomposition products of water are efficiently scavenged (unlike in BWRs on Normal Water Chemistry).



From llevbare & al., 2010

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Stress corrosion cracking has been experienced on unirradiated stainless steels :

- Low number of events
- 83% of the events occurred in low flow or stagnant ("occluded") zones.
- 17% occurred in nominally free-flow conditions and have been associated with cold work and excessive hardness (>300 HV)

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Objectives: to understand, to anticipate and to predict the degradation of PWR internals.

- > Preventive maintenance for the replaceable components
- > Extension of the operation time of the current PWRs (up to 60 years)
- > Understanding the degradation phenomena
- Improvement for new PWRs (materials & design)





- Cracking linked to the evolutions of the environment (radiolysis) and the materials (damages) due to irradiation
- Major influence of the dose rate





#### PWR primary water: 155 bar, 300-340°C, 30ccH<sub>2</sub>/kgH<sub>2</sub>0, 2 ppm Li, 1000ppm B

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# DE LA RECHERCHE À L'INDUSTRIE

### STRESS CORROSION CRACKING OF SG TUBES HISTORICAL PERSPECTIVES

1953: Stress Corrosion Cracking occurred in the stainless steel tubing of the steam generator of the prototype for the Nautilus. SCC failures occurred on the secondary surfaces of the tubes.

By 1957, H. Copson at INCO had demonstrated that the Stress corrosion cracking of Fe-Cr-Ni alloys, when exposed to boiling MgCl<sub>2</sub>, would stop above about 40% Ni.



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## HISTORICAL PERSPECTIVES OF ALLOY 600 SCC: 1959

## "Coriou effect"

1959: *Coriou et al.* published that Alloy 600 (75% Ni & 15% Cr) is sensitive to SCC in "pure" water at 350°C.



Intergranular SCC of Alloy 600 (350°C, pure deoxygenated water) – 3 months







Intergranular oxidation on Alloy 600 (350℃, pure deoxygenated water, 3 months)



Stress corrosion cracking susceptibility by CORIOU



SCC susceptibility as function of the nickel content (Coriou's work – summary 1967)

Work done with a series of laboratory alloys Fe-18%Cr-Ni with various Ni content

Pure water and solution with 0.1% NaCl / 350°C / 6 months / 1.2 and 1.7  $\rm E_{0.2}$ 

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#### HISTORICAL PERSPECTIVES OF ALLOY 600 SCC 1960-1985





### **TODAY SCC PHASES & INVESTIGATIONS**



Propagation: Internal oxidation & Hydrogen

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## **INITIATION – GRAIN BOUDARY OXIDATION**

# 

Oxygen transport in the crack tip (nano-SIMS and tracers)

Solution-annealed material (no intergranular carbide) 1340 hr in nominal primary water + primary water containing tracers (deuterium and <sup>18</sup>O) for **67 hr** 

-> <sup>18</sup>O is located at the tip of the IG penetration and in the outer part of the surface oxide layer, where the oxide growth occurs.

-> 18O diffusion occurs during the second stage of the test in 67 hr

-> estimate of the oxygen transport coefficient : > 10<sup>-13</sup> cm<sup>2</sup>.s<sup>-1</sup>

Oxygen transport up to the crack tip is not a rate limiting step.

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M. Sennour & al, JNL, 393 (2009) 254-266 P. Laghoutaris & al. , JNM 393 (2009) 254–266 C. Guerre & al., Environmental Degradation, TMS, 2011, 1477-1488



Two series of experiments were done, in which the specimens are exposed to:

– a PWR primary medium in which the water is replaced by deuterated water  $D_2O$  (D content 97.90%), pressurized by classical hydrogen gas (H<sub>2</sub>)

– a classical primary medium (with  $H_2O$ ), pressurized by deuterium gas ( $D_2$ ) (D content 99.05%).



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## **STRESS CORROSION CRACKING OF NI ALLOYS**

## Large progress on mechanisms



Internal oxidation model with an eventual influence of the cathodic hydrogen in grain bound

Steam generator tubes are no longer made of Alloy 600 MA or alloy 600TT

Good behavior of Alloy 690 and Alloy 800 linked to the higher chromium content

Artist's view of the Coriou's crack, by Juliette Plisson



### **PWR SECONDARY CIRCUITS**

# FLOW ACCELERATED CORROSION (FAC) & CARBON STEELS



### **BRIEF HISTORICAL BACKGROUND OF FAC**

#### INTERNATIONAL



12/02/1995 Pleasant Prairie (Fossil Plant 600 MW, USA)

2 persons died

09/08/2004 - MIHAMA 3 (Japon) Tube leakage downstream diaphragm



09/05/2007 – IATAN (Fossil Plant, USA)



2 persons died





#### Since 2001

FRANCE (No death)

BRT-CICERO<sup>™</sup> is used on all units in France

2002 Flamanville

Tube support plates degradation on AHP 500 heaters



## FLOW ACCELERATED CORROSION - FAC "BASIC UNDERSTANDING"

Electrochemical based phenomena accelerated by flow rate (and not mechanically – erosion / abrasion)

- Metallic materials in contact with liquid water at high flow rates
- Carbon steels (CS) or low Alloyed Steels (LAS) covered with a protective layer magnetite / hematite (passive layer)
- Stationary conditions: V<sub>film formation</sub> = V<sub>oxide dissolution</sub>
  High flow rate

Oxide dissolution is fasterOxide film thickness is decreasingCorrosion rate is increasing





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Inside tubes



## FAC: IMPORTANCE OF THE CHEMICAL CONDITIONNING

The flow accelerated corrosion rate decreases when pH increases (iron solubility decreases when pH increases) Importance of the chemical agent (linked to the formation of complexes)



With only 0,5% of chromium in the steel, the FAC rates are decreased by a factor of 10 at least



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## **MAJOR COUNTERMEASURES TO FAC IN POWER PLANTS**

Plants	equipment	material	water chemistry	improvement	remarks
	where the feed	improvement	рH	[O <sub>2</sub> ]	
	water flows in				
PWR	steam generator	increasing [Cr]	increasing pH	AVT(H&O)	minimizing [O <sub>2</sub> ]
					at SG inlet
BWR	reactor core	increasing [Cr]	- (neutral)	oxygen	[O <sub>2</sub> ] generated
				injection	due to radiolysis
Fossil	boiler	increasing [Cr]	increasing pH	AVT(O)	
				OT From S	5. Uchida & al., FAC2013

Importance of the design of the components, modeling and prediction of degradations (COMSY, BRT-CICERO, WATHEC, CHECWORKS...) coupled with non-destructive examinations.





### CONCLUSION

#### Corrosion issues lead to evolutions of materials

- SG tubes: Alloy 600 to Alloy 690
- FAC: from carbon steels to low alloyed steels (LAS – less than 1%Cr)

## **MODELLING & SIMULATION**

## **MULTISCALE MODELING**



- $\Rightarrow$  Needs to replace semi-empirical models by more phenomenological models
- ⇒ Multiscale modeling is then needed
- ⇒ Strategy for homogenization & harmonization



#### SCC: Influence of hydrogen on crack propagation



P. Laghoutaris & al. , JNM 393 (2009) 254–266

- Molecular dynamic simulation: LAMMPS
- Interatomic potential: EAM (type)
- 750 000 atoms (Ni) Ductile crack without H

Brittle crack with H

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Atomistic simulation of oxidation & experimental observations in agreement



Atoms & molecules by Juliette Plisson