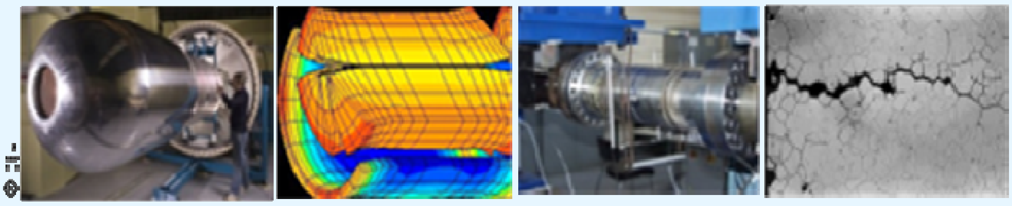
MINOS

Centre of Excellence for Nuclear Materials

Workshop

Materials Innovation for Nuclear Optimized Systems



December 5-7, 2012, CEA – INSTN Saclay, France

Benoît TANGUY et al.

CEA (France)

The Irradiation-Assisted Stress Corrosion Cracking (IASCC) issue: some Examples of Studies Carried out at CEA

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Workshop Materials Innovation for Nuclear Optimized Systems
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The Irradiation-Assisted Stress Corrosion Cracking (IASCC) Issue: some Examples of Studies Carried out at CEA

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Irradiation assisted stress corrosion cracking (IASCC) is a problem of growing importance in pressurized water reactors (PWR). An understanding of the mechanism(s) of IASCC is required in order to provide guidance for the development of mitigation strategies. One of the principal reasons why the IASCC mechanism(s) has been so difficult to understand is the inseparability of the different IASCC potential contributors (radiation induced segregation (RIS) at grain boundaries, radiation induced microstructure (dislocations loops, voids, bubbles, phases), localized deformation under loading, irradiation creep and transmutations) evolutions due to neutron irradiation. While the development of some of the contributors (RIS, microstructure) with increasing doses are at least qualitatively well understood, the role of these changes on IASCC remains unclear. Fundamental studies related to the IASCC mechanisms can be divided on two main parts: (i) How the irradiation modifies the austenitic stainless steels (ASS) microstructure (and so the ASS mechanical behavior) as a function of dose, temperature, stress, spectrum and flux and how it affects the resistance of the ASS to SCC sensitivity and (ii) How the irradiation flux modifies the corrosion process themselves (oxidation) and how it may affect the SCC kinetics. For this last topic, both a modification of water chemistry (radiolysis) and a change of oxide are probably involved.

To answer these questions, parallel to experimental characterizations at different scales, development of modeling is of growing importance in the understanding of the basic mechanisms of IASCC but also of their interactions. This lecture describes some studies carried out at CEA in order to provide further understanding in the IASCC damage modeling. First part of this lecture describes the methodology carried out at CEA in order to provide more experimental data from constant load tests dedicated to the study of initiation of SCC on neutron irradiated stainless steel. A description of the autoclave recirculation loop [1] dedicated to SCC tests on neutron irradiated materials is then given. The main steps of the interrupted SCC tests carried are described relative to their objectives.

Second part of the lecture gives some insights of the effect of irradiation on the oxidation processes. Oxidation of stainless steel in PWR primary water at 325°C has been studied by investigating the influence of defects created at the alloy subsurface by proton irradiation performed before corrosion test. Corrosion experiments were performed during two different corrosion sequences using $H_2{}^{18}O$ for the second corrosion one. The oxide layer was studied by SEM and TEM and could be divided in two parts: an external discontinuous layer composed of crystallites rich in iron and an internal continuous one richer in chromium. Tracer experiments underlined that the growth of this protective scale was due to oxygen diffusion in the grain boundary of the oxide layer. Defects created by irradiation have an effect on the two oxide layers. They are preferential nucleation site for the external layer and so increase the density of the crystallites. They also induce a slower diffusion of oxygen in the internal layer.

The third part of the lecture focuses on the plasticity of the irradiated stainless steel at the grain scale. The goal of this work is to model irradiation-induced strain localization at the grain scale, using 3D dislocation dynamics (DD) simulations. More specifically, it is attempted to predict the number of shear bands affecting (deforming) the grain boundaries, in presence of a representative irradiation defect cluster populations.

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In practice 2 types of DD simulations were used, based on their complementary capacities and limitations. Simulations (Figure 1) where irradiation-induced defect clusters are treated as planar obstacles to dislocation motion were carried out. This description has reduced computational load and is compatible with the introduction of thermally activated cross-slip for simulating multiple slip band formation. Shear band spacing and plastic strain spreading obtained using these simulations show that spacing increases with increasing dose, grain size and increasing stacking fault energy (SFE). The proposed model has been successfully extrapolated to grain sizes and defect cluster populations representative of actual fcc alloys, submitted to typical PWR neutron irradiation conditions.

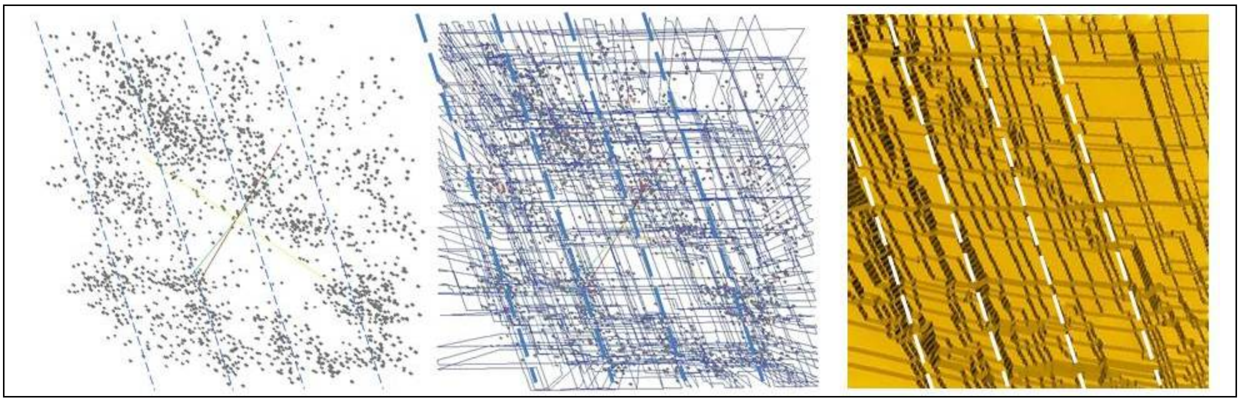

Fig. 1: Clear bands and strain localization predicted using Type-2 DD simulations. Left image: the represented loop-facets are those potentially absorbed by mobile screw dislocations. High density of absorbed loops materializes the channel position. Central image: swept loop-facets and corresponding dislocation structures. Right image: plastic strain mapping in grain boundary corresponding to left and central images.

Finally, simulation results to assess the influence of slip localization effect, which is an important feature of plasticity in irradiated ASS's, on the grain boundary stress-strain fields are presented. Slip localization is known to trigger grain boundary brittle fracture. For predicting the local stress fields, an elastoplastic slip band is assumed to be embedded at the surface of an elastic matrix. Numerous FEs computations have been carried out allowing the proposal of analytical formulae describing the grain boundary (GB) normal stress fields depending on clear band length and thickness, Schmid factor, slip band critical shear stress and remote stress mainly. Finally, finite fracture mechanics is applied, together with both critical GB fracture energy and stress criteria. This leads to analytical formulae as simple as the ones deduced from the pile-up theory, but taking into account the channel thickness.

References

[1] B. Tanguy, C. Pokor, A. Stern, P. Bossis, Initiation stress threshold Irradiation Assisted Stress Corrosion Cracking criterion assessment for core internals in PWR environment. ASME Pressure Vessels and Piping Division Conference, July 17-21, 2011, Baltimore (MD), USA.

Acknowledgements

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DE LA RECHERCHE À L'INDUSTRIE

The Irradiation-Assisted Stress Corrosion Cracking (IASCC) issue: some examples of studies carried out at CEA

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[2]: Section for Research on Applied Metallurgy
[3]: Service de la Corrosion et du Comportement des Matériaux dans leur Environnement

DEN, CEA Saclay

MINOS Workshop, Materials Innovation for Nuclear Optimized Systems
December 5-7, 2012, CEA – INSTN Saclay, France

Background : Structural integrity of Core Internals

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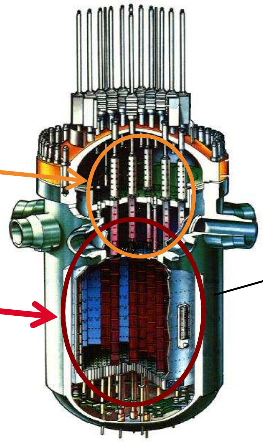
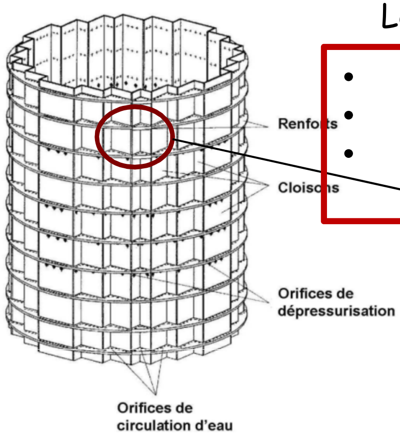
Severe irradiation conditions in PWR core (RPV+internals) → Material ageing and potential degradation → Limit of reactor operational life

Upper Internals

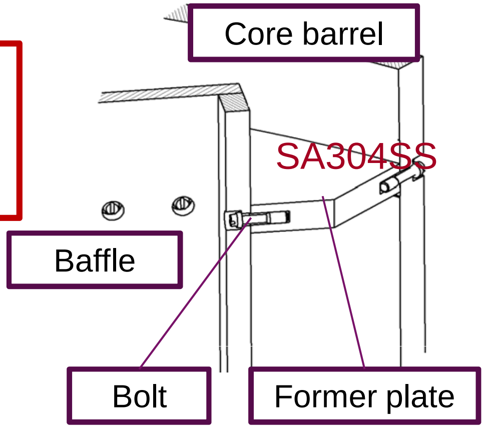
Lower Internals





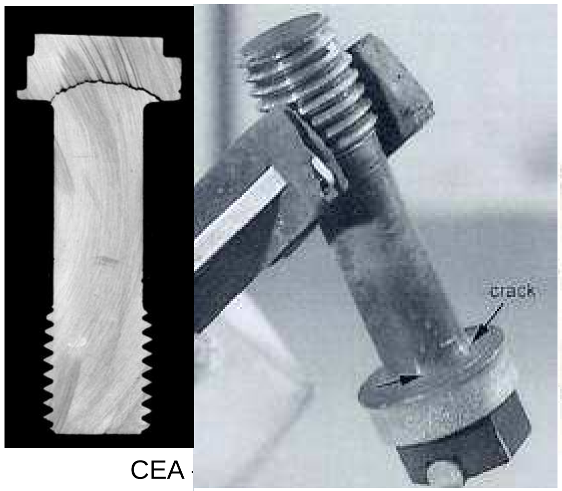
Lower Internals

- $10^{-7} – 10^{-8}$ dpa/s
- 290 – 380 °C
- 80 -100 dpa after 40 years



CW316SS 900-1000 bolts / reactor vessel

•**Field of experience:** IASCC of the baffle/former bolts



[Thomas, Fontevraud 2002]

->IG Crack at the head to shank transition region (8.5 dpa – 300°C)

-> Focus on IASCC initiation for PWR's bolts

The internals lifetime has an important impact on the nuclear power plant lifetime because the cost and difficulty of their replacement

IASCC Initiation: Mechanistic issues

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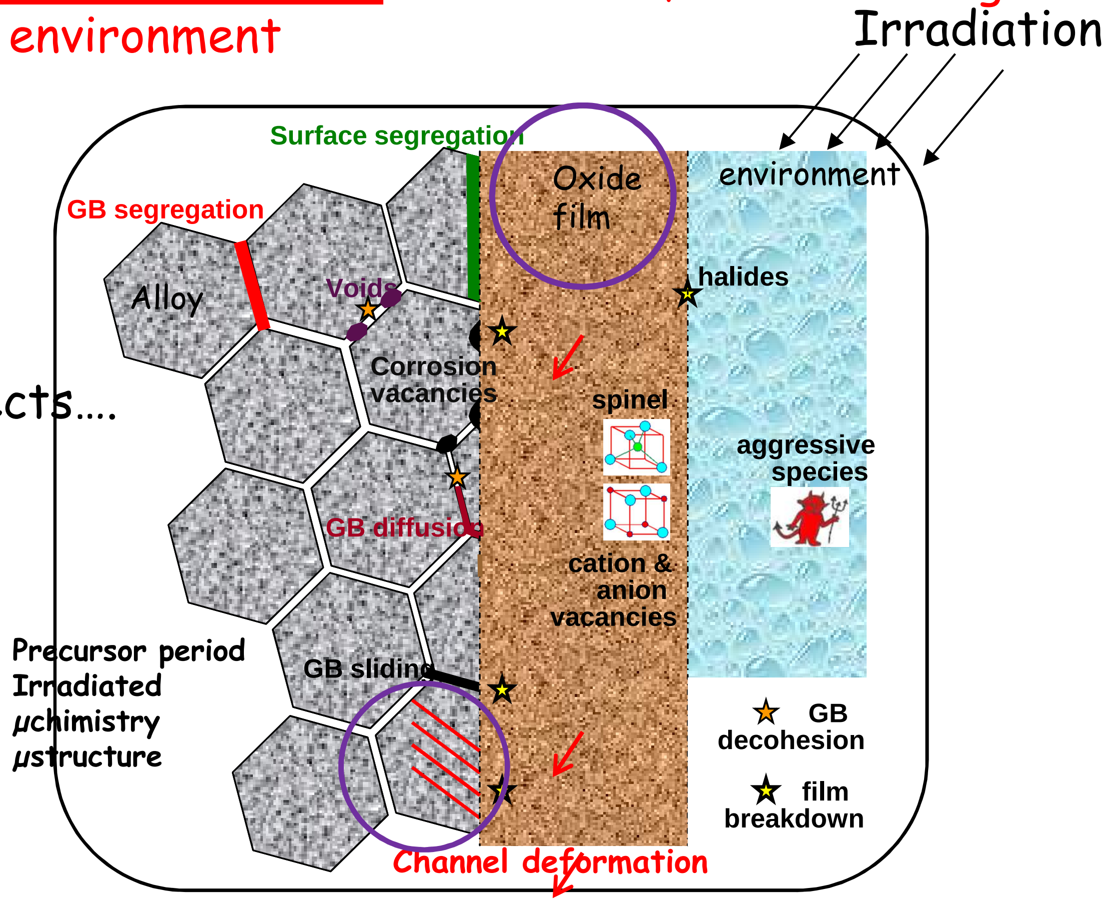
➢ Simultaneous effects of radiation, stress and high temperature water environment

+synergistic effects….



[from M. Vankeerberghen, SOTERIA]

-> mechanisms understanding of IASCC initiation and its modelling are still under investigation

CONTENT

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- Background
- Initiation Stress Threshold IASCC Criterion Assessment
- Influence of irradiation on the oxide layer growth mechanism and on diffusion process in the oxide layer
- Effect of localization on Grain boundary microcracks nucleation
- Investigation of clear bands formation mechanisms

INITIATION STRESS THRESHOLD IASCC CRITERION ASSESSMENT

Section for Research on Irradiated Materials

Objectives and methodology

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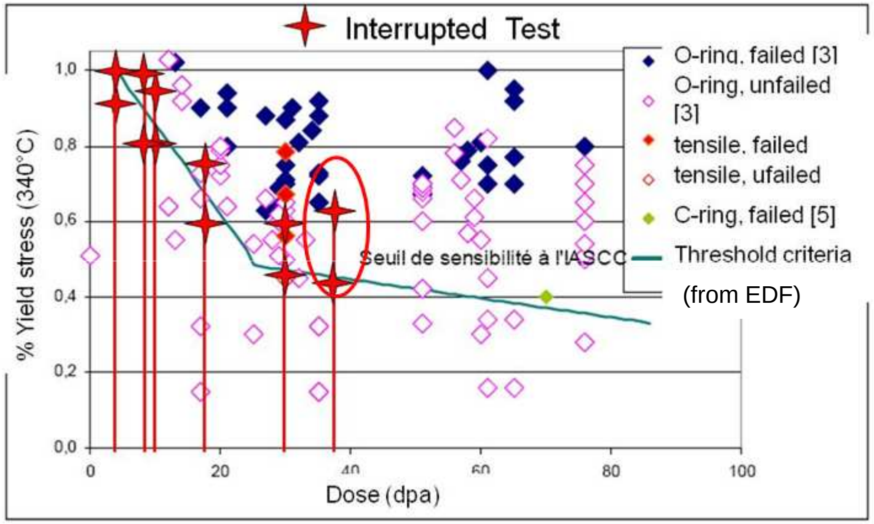
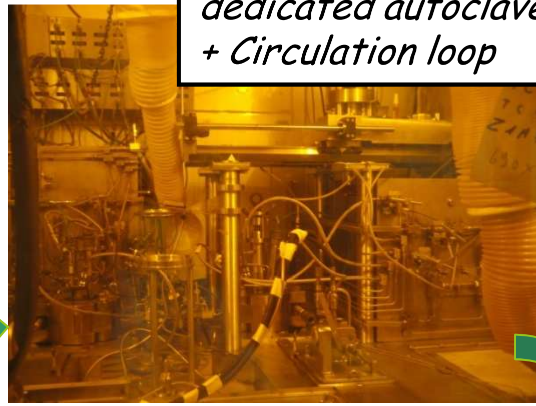
Motivation : Industrial interest to establish an IASCC fracture criteria to understand and predict life duration of the bolt assembly. Sensitivity to first order parameters (Temperature, stress, deformation mode, irradiation, chemical composition)



In-cell tensile test dedicated autoclave + Circulation loop



LECI nuclear facilities at CEA

In-cell SEM

- <u>Environment</u>: Simulated PWR: 1200 ppm B, 2 ppm Li, 25<H2<35 (cc/kg)
- T(test)=340°C
- Interrupted test (1000h+1000h) with two loading levels
- <u>Loadings</u> : **constant load** (α_i irradiated yield stress at considered dose)

Methodology & results examples

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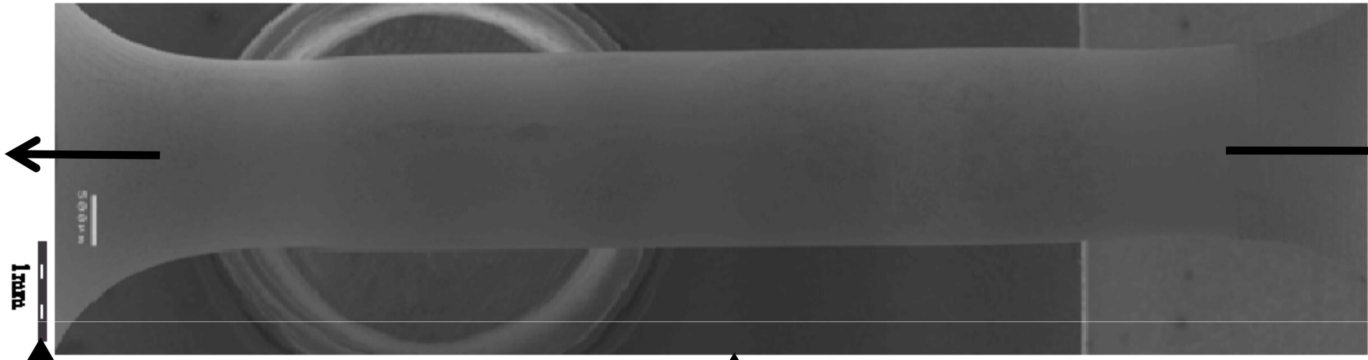
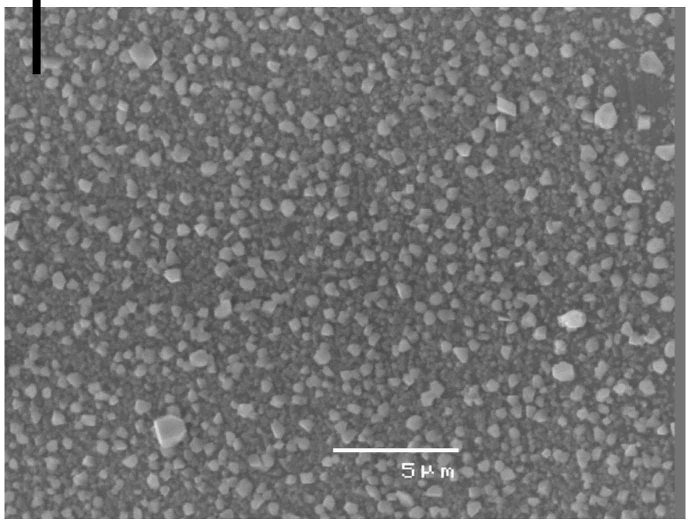
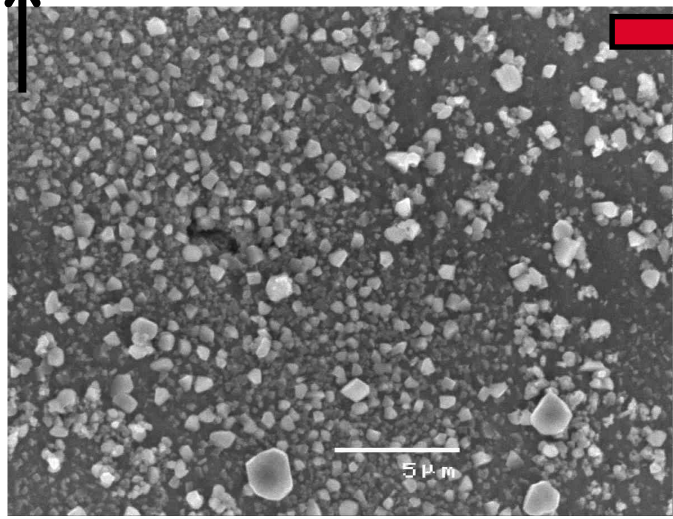
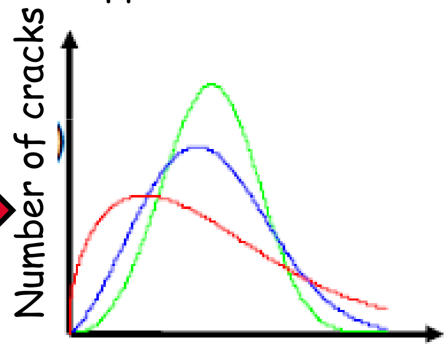
Surface characterization: surface oxidation features of irradiated stainless steels in primary PWR environment + microcracks distribution







Microcracks size distribution as a function of dose and applied stress



->On-going program

INFLUENCE OF IRRADIATION ON THE OXIDE LAYER GROWTH MECHANISM

Service de la Corrosion et du Comportement des Matériaux dans leur Environnement

Materials and methodology

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- CW316L , 3MeV proton-irradiated (~40µm depth, surface^{irr} Φ~1cm), 2.10^{12} protons/cm^2/s
- Sample preparation : SiC paper (→ 4000) + diamond + OPS
- Methodology -> Tracer experiments :
  - 1st corrosion sequence with $H_2\,^{16}O$ media
  - 2nd corrosion sequence with $H_2\,^{16}O$/ $H_2\,^{18}O$
  - Localisation of ^{18}O and ^{16}O in the oxide layer

1st corrosion sequence: 620 hours

- 1000ppm [B], 2ppm [Li], [H2]dissolved = 1.3*10^{-3} mol.l^{-1}
- $pH_{325°C}$ = 7.2

Corrosion loop at LECA, T = 325 °C, P = 155 bar

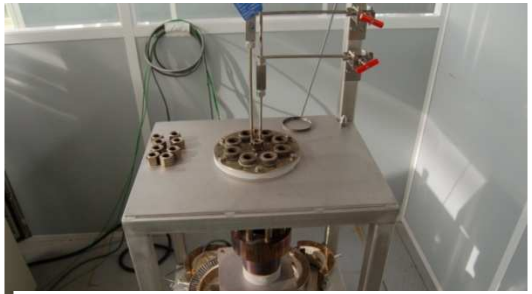
->

2nd corrosion sequence:

- 1000ppm [B], 2ppm [Li], [H2]dissolved = 1.3*10^{-3} mol.l^{-1}
- $pH_{325°C}$ = 7.2

➢ 17 h: determination of **diffusion coefficient**

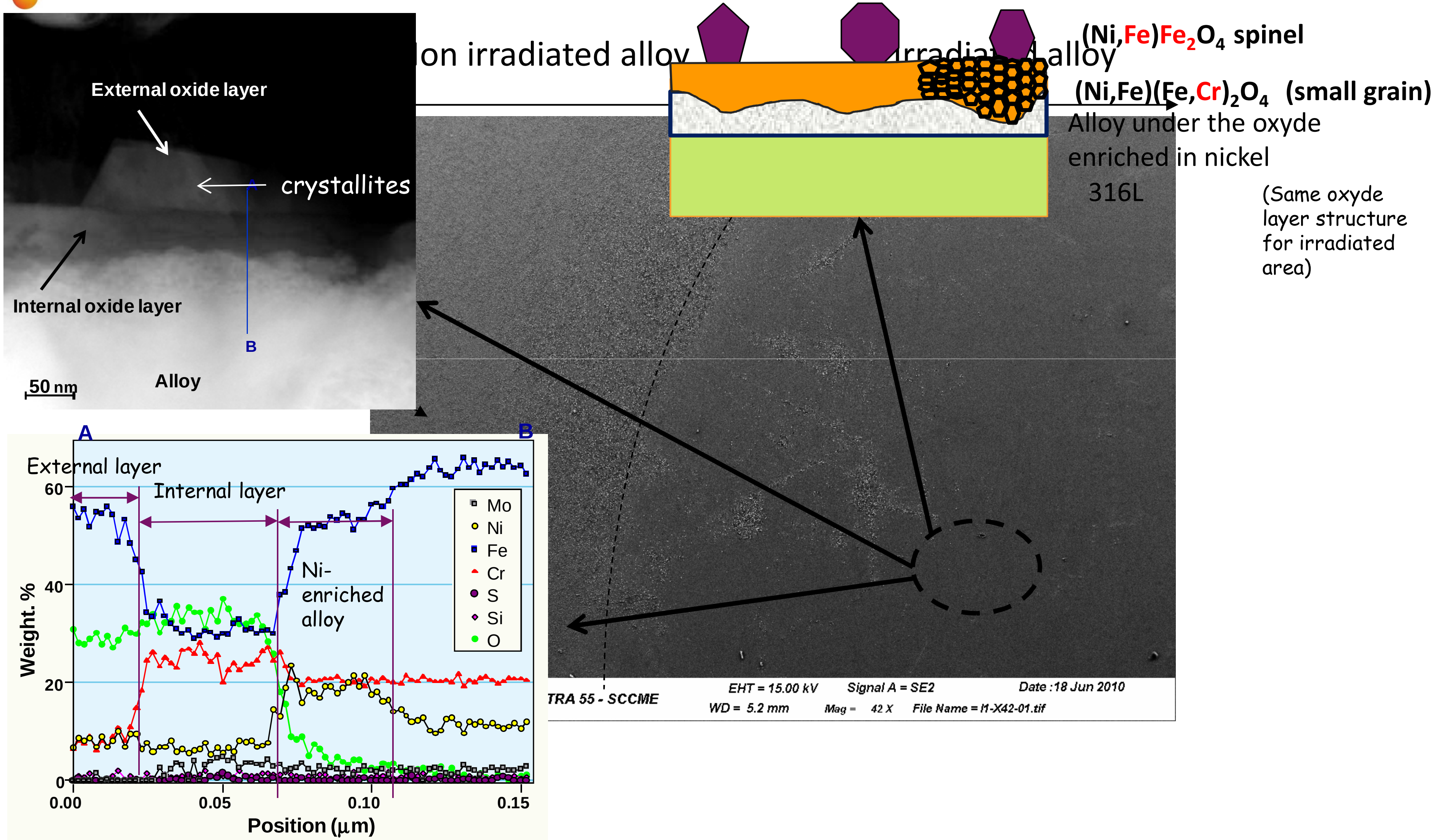
➢ 404h: determination of **corrosion mechanism**



Titanium autoclave (300ml), T = 325 °C, P = 128 bar

Effect of irradiation on the external layer (after 1st sequence)

TEM Characterization after 1024 hours (coop. with M.Sennour and C. Duhamel (ENSMP))



EDX analysis

Effect of irradiation on the external layer (after 1st sequence)

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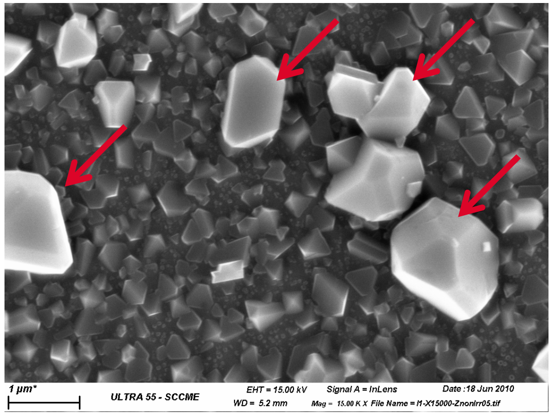
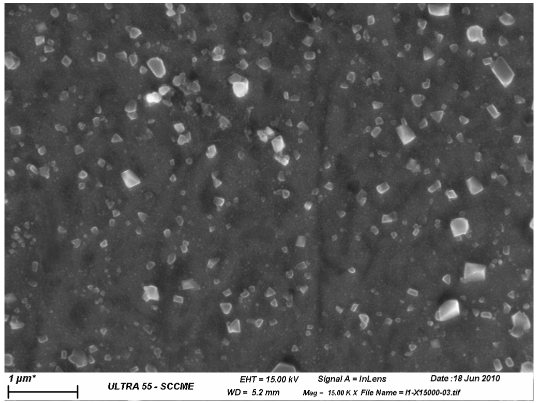
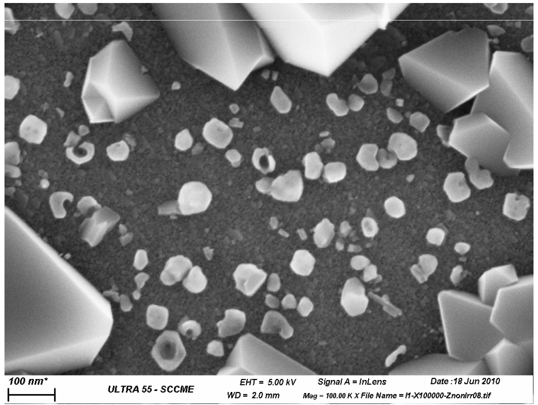
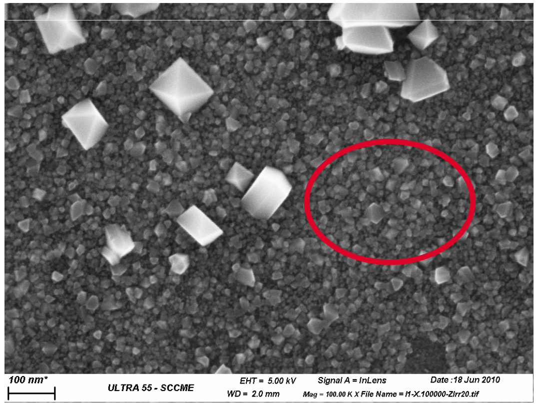
Non irradiated alloy		Irradiated alloy
	x15000	
	x100000	

➡ Bigger cristallites (external layer) on non irradiated sample

➡ Lot of very small cristallites (external layer) on irradiated sample

→ influence of irradiation on nucleation/growth of external layer

Effect of irradiation on the internal layer (after 2nd sequence)

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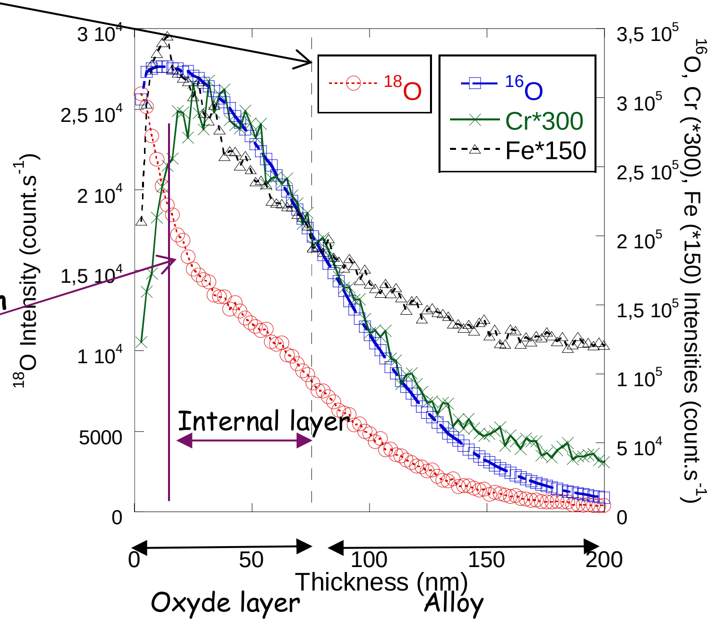
Short second sequence (no significant oxyde layer growth) -> determination of diffusion coefficient: Dgb

SIMS analysis (collaboration F. Jomard (CNRS))

Non irradiated alloy

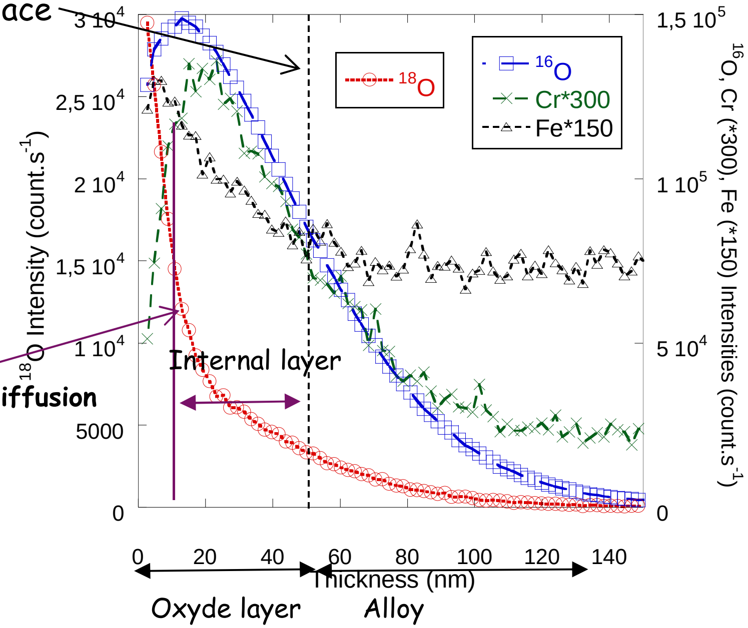
alloy/oxyde interface



^{18}O diffusion

Irradiated alloy

alloy/oxyde interface



^{18}O diffusion

Diffusion at oxyde gbs

^{18}O diffusion profile → $C(x) = K\,\mathrm{erfc}\left(\dfrac{x}{2\sqrt{D_{GB}t}}\right)$ →

Av. concentration

	Non irradiated	Irradiated
Dgb ($cm^2.s^{-1}$)	~3.10^{-16}	~5.10^{-17}

->diffusion is faster for non irradiated samples.

Effect of irradiation on the internal layer (after 2nd sequence)

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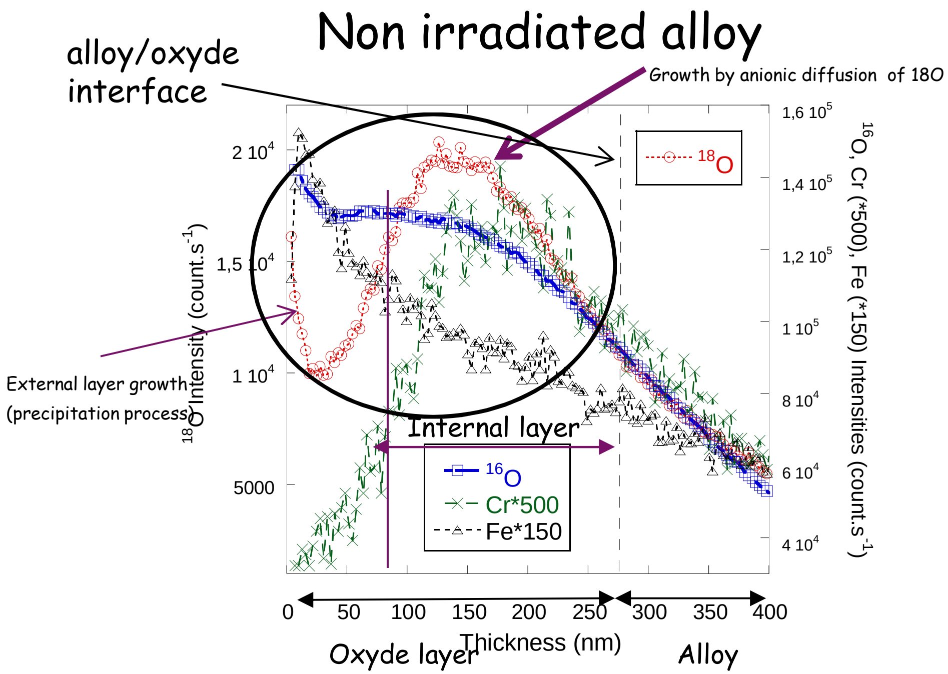
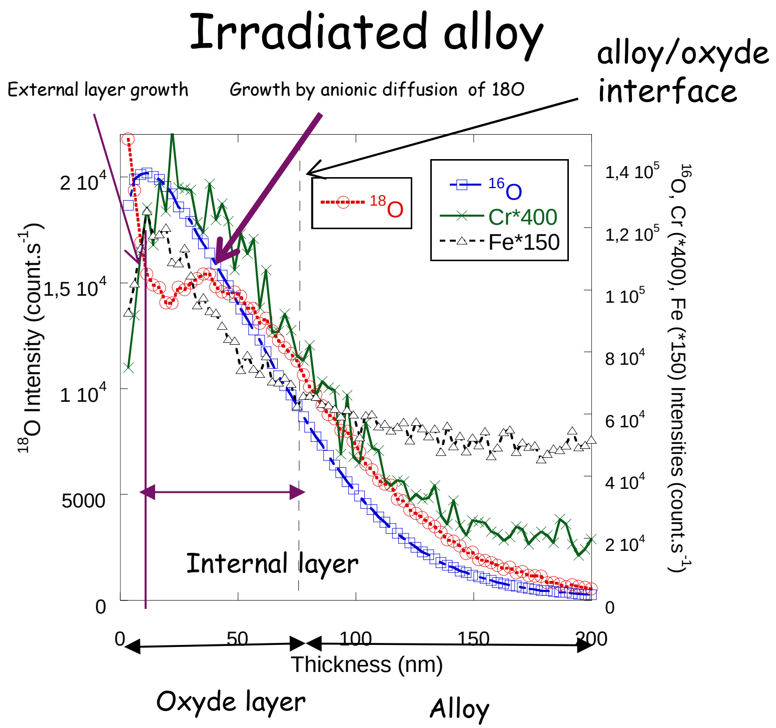
Long second sequence (growth of oxyde layer) -> determination of corrosion mechanisms

SIMS analysis (collaboration F. Jomard (CNRS))





➡ confirms that this diffusion is faster for non irradiated samples.

➡ Accumulation of ^{18}O close to the alloy-oxyde interface: anionic diffusion along the grain boundaries of the oxide as the mechanism of internal layer growth

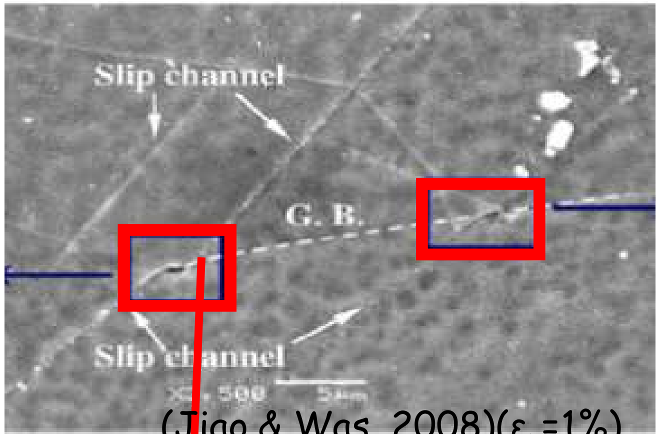
EFFECT OF LOCALIZATION ON GRAIN BOUNDARY MICROCRACKS NUCLEATION
(DYNAMIC STRAINING)

Section for Research on Applied Metallurgy

Methodology: cristalline constitutive laws + FE simulations

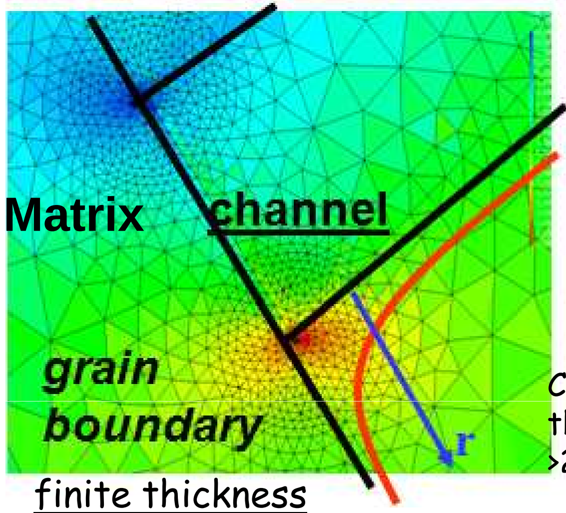
MINOS

SEM observations: microcrack nucleation observed at the intersections of channels & grain boundaries



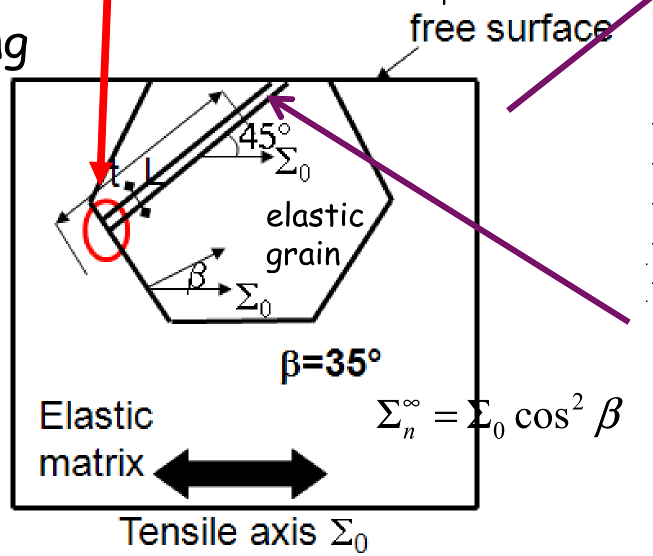
(Jiao & Was, 2008)(ε_p=1%)

Grain boundary stress fields induced by the impingement of pre-existing channels

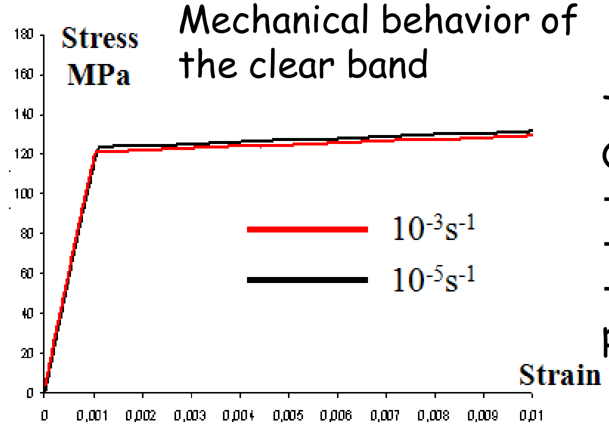


Channel thickness~50nm
>200 atomic planes

Modelling



Mechanical behavior of the clear band



Time-independent behavior
Quasi perfect plasticity
→Critical shear stress: τ_0
→Hardening slope: H
→Latent hardening parameter: q

Comparison with the predictions of the pile-up theory: 'close-field' –'far-field'

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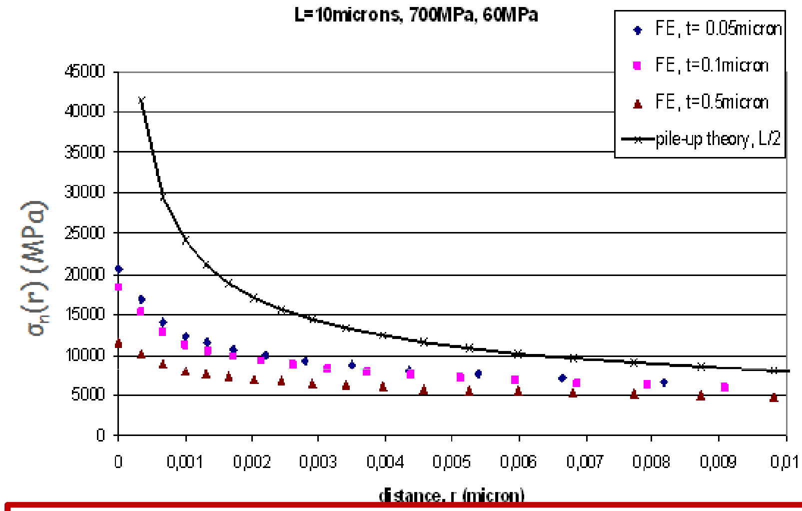
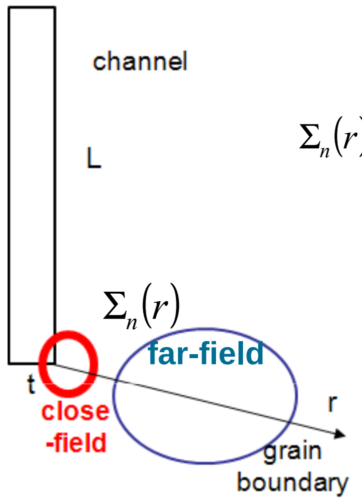
$t<r<<L$

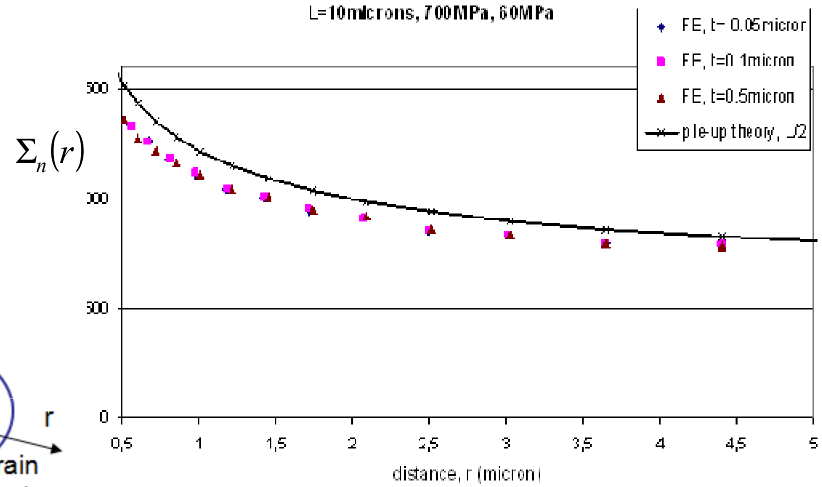
$r<<t<L$

close-field





far-field



->Large overestimation by the pile-up theory (stress exponent: 0.5) using a length equal to one-half of the grain size

->Very close to the intersection of the channel and grain boundary: weaker singularity due to

- the notch effect → $\sigma_n(r) \sim 1/r^{\alpha}$, $\alpha=0.32<0.5$ ($t<<r$)
- slip band plasticity
- single slip

-Whatever the grain size L and the aspect ratio, L/t, the discrepancy between the pile-up theory prediction and the curves computed by the FE method is lower 10%

-In fact, if $r>t$, the stress singularity is very close to the pile-up or crack one as expected (Leguillon et al., 2007)

Effect of localization on grain boundary crack nucleation

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Prediction of remote stress to GB microcrack nucleation based on:

- An energy balance criterion (fracture energy: $\gamma_{fracture}$)
- A critical stress criterion using: $\sigma_c = \sqrt{\dfrac{Y\gamma_{fract}}{d_0}}$

$$\Sigma_c = (1/f)\left(\tau_0 + \left(\frac{2\lambda}{YC}\right)^{1-\lambda} A^{1-2\lambda}\left(\frac{t}{L}\right)^{1/2}\left(\frac{d_0}{t}\right)^{1-\lambda}\sqrt{\frac{Y\gamma_{fract}}{d_0}}\right)$$

channel lengths, t, L [microstructure, (dose)]

the Schmid factor, f, channel critical shear stress, τ_0 [dose]

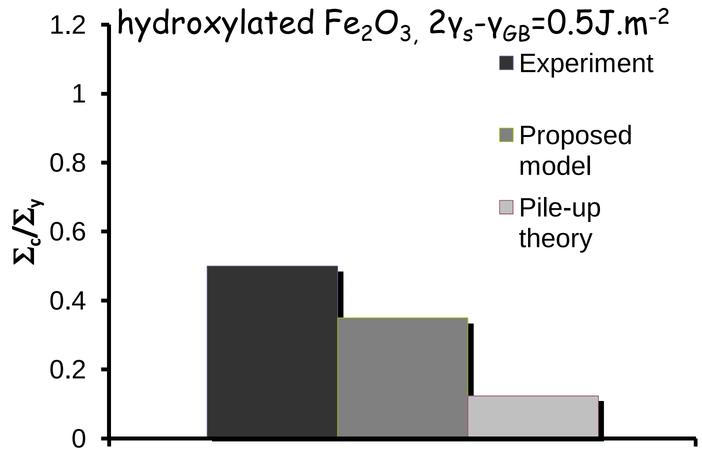
fracture energy, ($2\gamma_s - \gamma_{GB}$) [oxydation, dose]

(Sauzay & Vor, Eng. Fract. Mech., in press)

without oxidation:

Exp. data: (Nishioka, 08)

PWR water tensile tests, 320°C



Exp. data: (Nishioka, 08) 35dpa

t=50n… J/m²)

-> Developped criteria leads to an improvment of experiments description

INVESTIGATION OF CLEAR BANDS FORMATION MECHANISMS

Section for Research on Applied Metallurgy

Methodology: 3D-DD simulations

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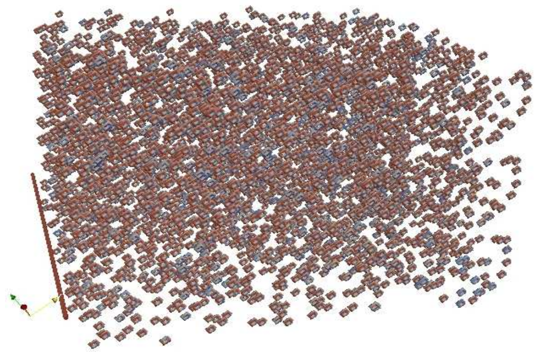
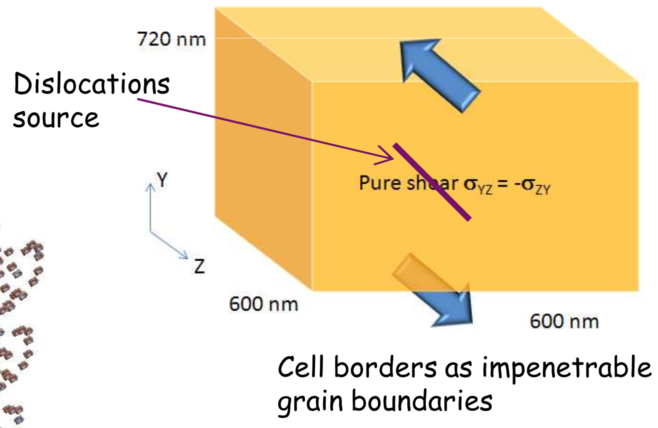
Type 1: boundary conditions simulation volume
-> investigation of single clear band formation mechanisms
•Random prismatic loop positions (D = 10 nm, L = 50 nm), $3.7*10^{22}$ loops/m^3(<1dpa)
•Loops resistance is intrinsic for srew dislocation

Type 2: boundary conditions simulation volume
-> investigation of clear band multiplication
•Random positions, loops=planar internal obstacles (facets, D = 10 nm), $1*10^{22}$ loops/m^3(<1dpa), ε_p=1.4*10-3
•Facets resistance=f(incoming dislocation type)

Random loops position



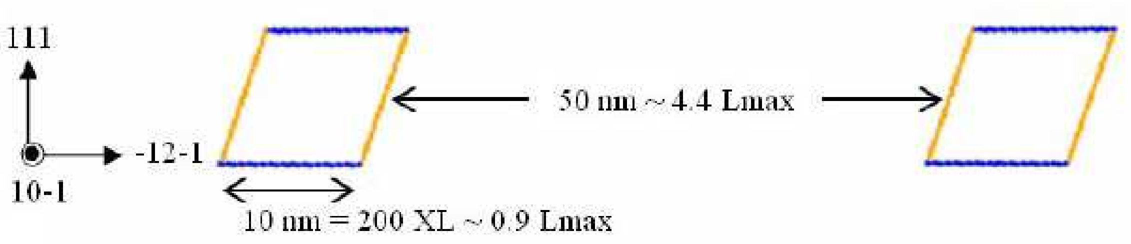
Thermally activated cross-slip is switch off



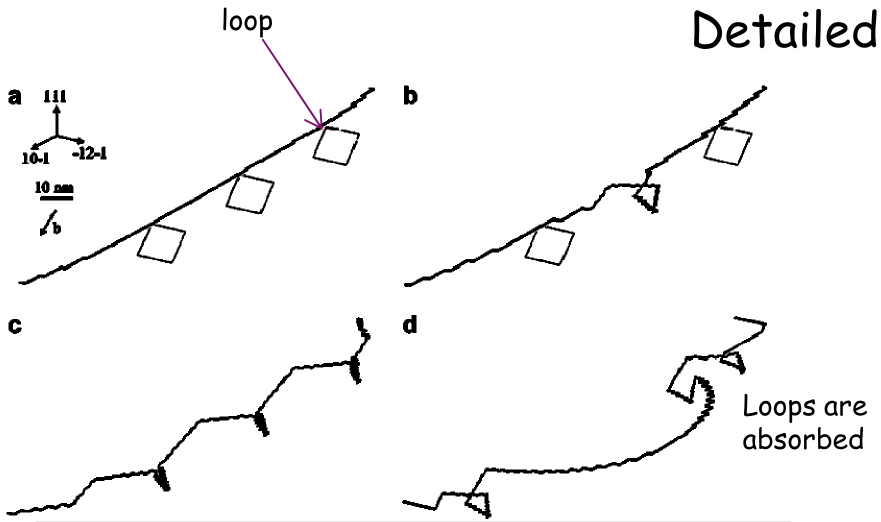
Single loop = 1 soft Facet

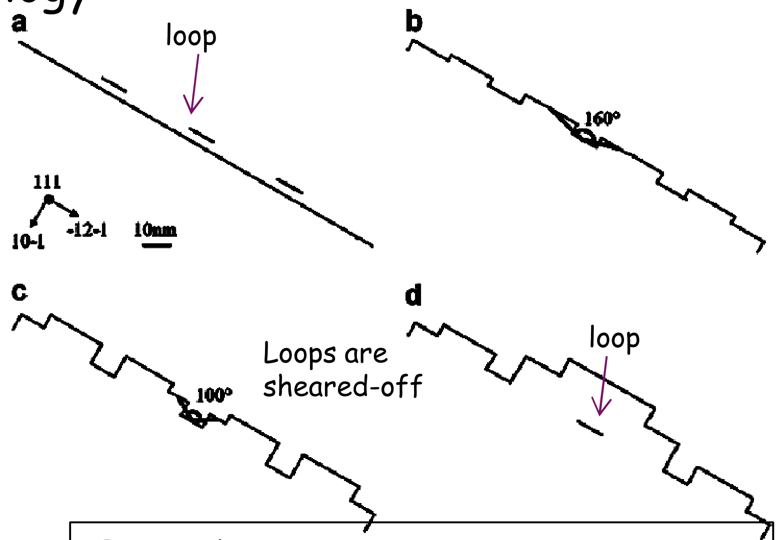
Dislocation-loop interactions based on MD simulations (type 1)

Goal: introduction of loop-induced evolutions in DD simulations



Detailed loop topology





Screw/Loop interactions:
Strong pinning force
Elevator *climb* effect (unpinning mechanism:re-emission of the dislocation in upper plane)

Edge/Loop interactions :
Weak pinning force
Planar dislocation motion

Details of single clear band formation (type 1 simulations)

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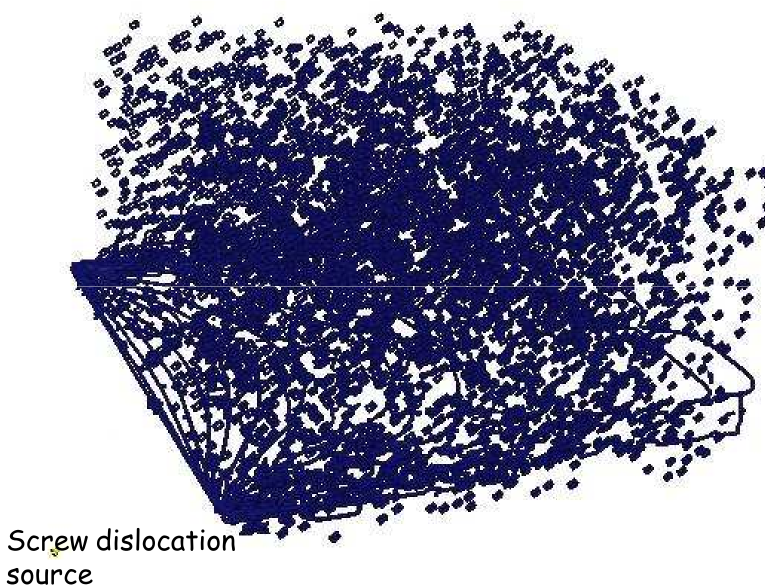
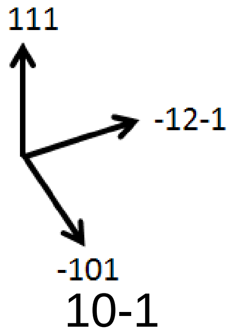
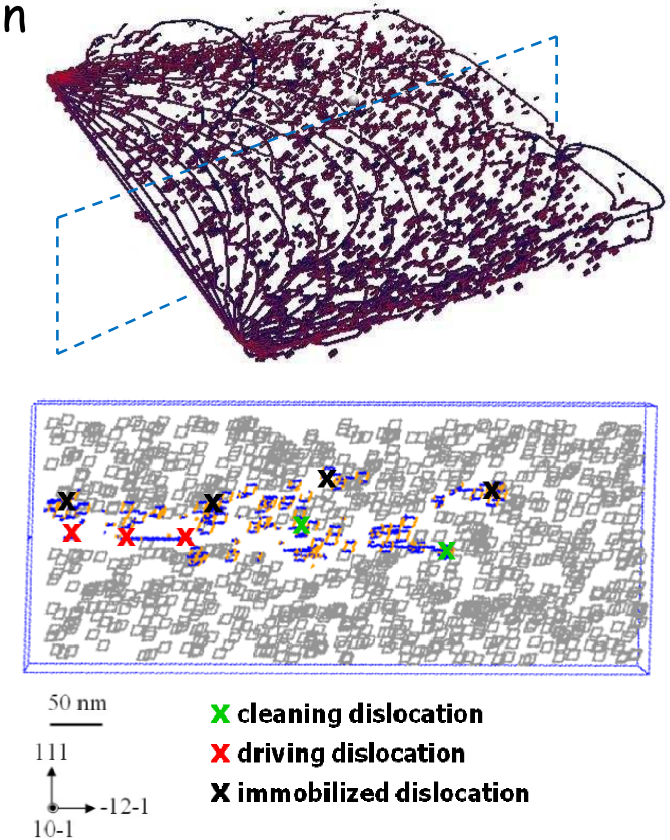
View of simulation cell after the clear band formation



-> Dislocation glide only possible through a collective mechanism : **leading dislocations clear the band**. **Trailing dislocations** concentrate the stress on leading dislocations by piling-up effect





-> clear band with finite thickness is due to collective dislocation effects: pile-up formation and arm exchange

Investigation of clear band multiplication (type 2 simulations)

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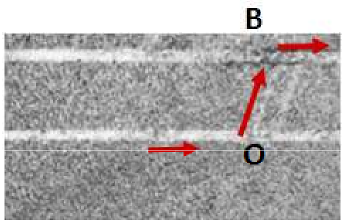
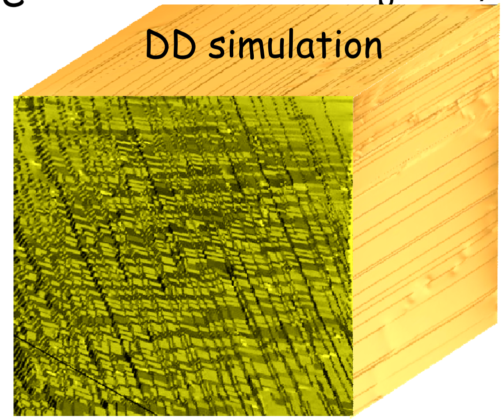
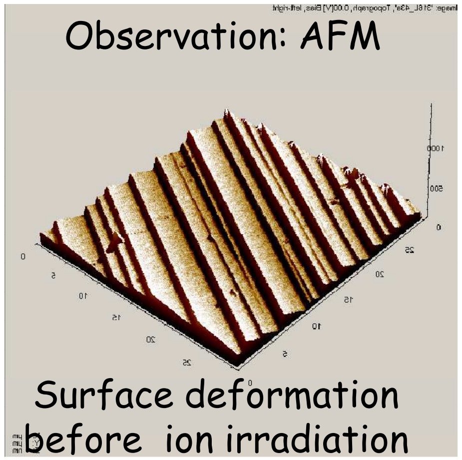
Acute cross-slip (long range mechanism): clear band multiplication mechanism



No loops

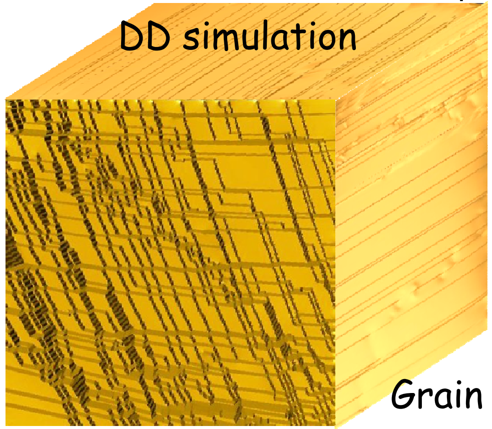
$\varepsilon_p = 1,4\times10^{-3}$



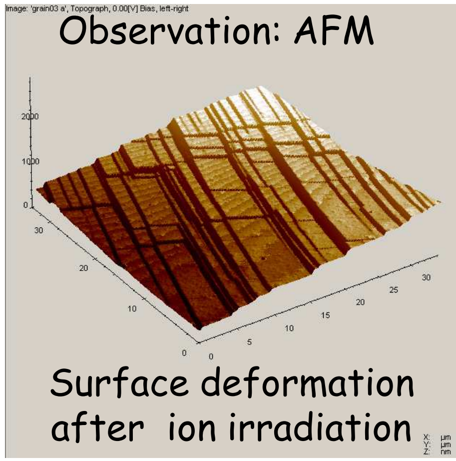


10^{22} loops/m^3 (~ 0.5 dpa)

$\varepsilon_p = 1,4\times10^{-3}$



Grain size~1 micron



The number of slip bands per unit volume: trends are correctly described
-> radiation-induced strain localization is well captured by DD model

Micro-mechanical model predictions (type 2 simulations)

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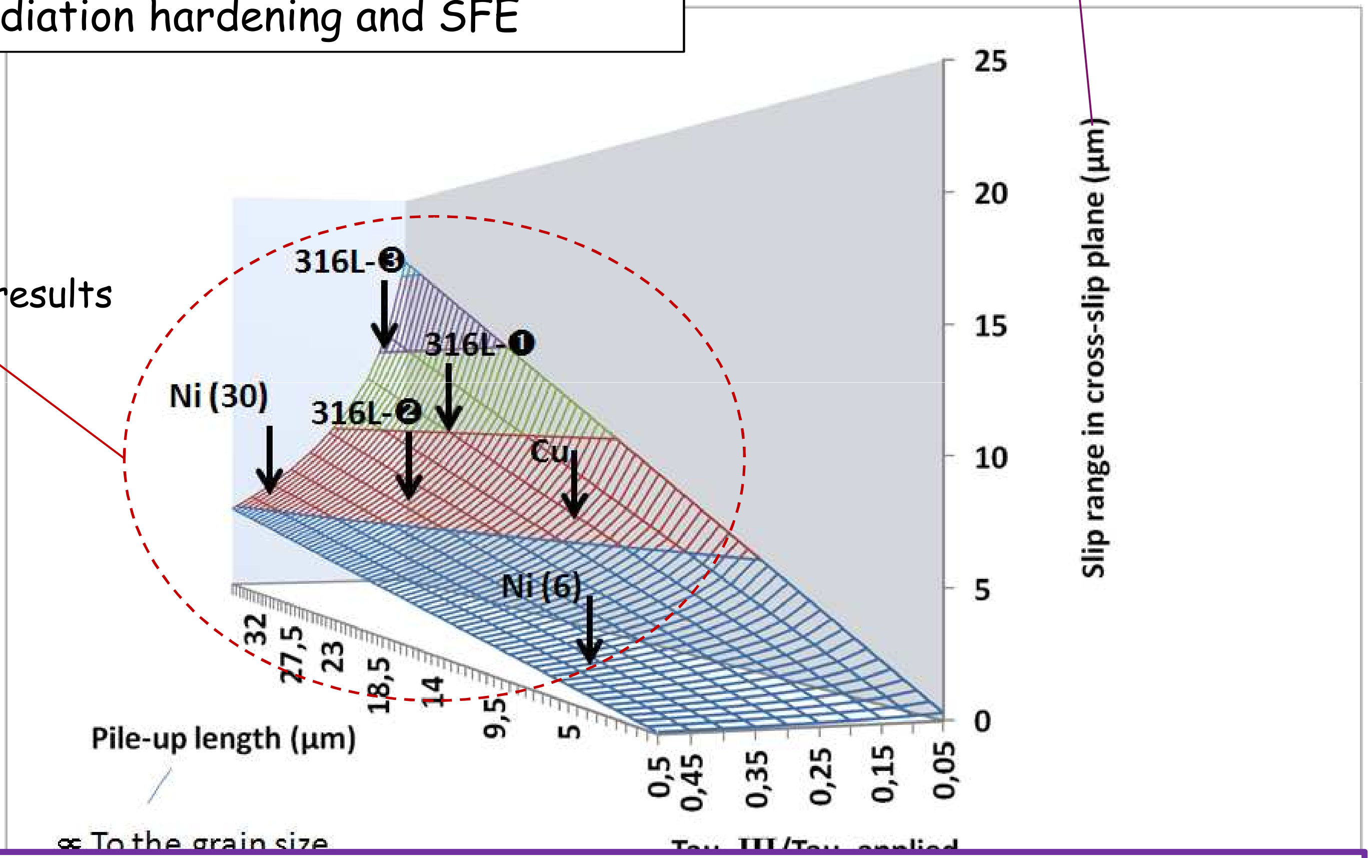
-> 3 influential parameters: grain size, irradiation hardening and SFE



-> Quantitative predictions of clear bands nucleation and multiplication under dynamic straining

Conclusions: Overview of the studies related to Internals at CEA

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	Experimental (Neutron irradiated materials)	
Microstructure and radiation hardening	Mechanical tests TEM	
Radiation Induced segregation	TEM-EDX, TAP	
Localization of the deformation	Mechanical tests, TEM	↔ Simulation tool : JANNUS CEA (irradiation with particles)
Swelling	Irradiation at high doses, Swelling measurement, TEM	↔ Simulation tool : JANNUS CEA (irradiation with particles)
Irradiation Creep	In-reactor creep tests, TEM	
SCC of irradiated material	SCC tests on recirculation water loop	↔ Simulation tool : JANNUS CEA (irradiation with particles)
In-reactor IASCC	Swelling mandrel	

Simulation tool : JANNUS CEA (irradiation with particles)

Modelling

Mutiscales modelling (rate theory, molecular dynamics, dislocations dynamics, crystal plasticity, mesoscopic mechanical behavior)

Thank you for your attention

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