Simulation of C-Component Dislocation Loops by Ions and Protons Irradiation. A Tool to Understand the Breakaway Growth of Recrystallized Zr-Based Alloys

Jean Paul MARDON¹, Nesrine GHARBI², Thomas JOURDAN³, Didier GILBON⁴,
 Fabien ONIMUS², Xavier FEAUGEAS⁵, Rosmarie HENGSTLER-EGER⁶

¹ AREVA NP, Fuel Business Unit (Lyon, France)

²CEA-DEN-DMN, Service de Recherches Métallurgiques Appliquées, SRMA (Saclay, France)

³CEA-DEN-DMN, Service de Recherches de Métallurgie Physique, SRMP (Saclay, France)

⁴CEA-DEN, Department of Nuclear Materials, DMN (Saclay, France)

⁵LaSI, University La Rochelle (La Rochelle, France)

⁶AREVA NP, GmbH (Erlangen, Germany)

During normal in-service operation, the PWR fuel assemblies exhibit a macroscopic elongation. This dimensional change is the result of three different phenomena: thermal creep, irradiation-creep and irradiation growth which occurs in the absence of any applied stress. This last phenomenon, referred as stress-free growth, is related to irradiation-induced microstructural changes taking place in the hexagonal close packed grains and corresponding to an elongation along the basal plane and a shortening along the c-axis, the volume remaining constant (Fig 1). Under neutron irradiation, recrystallized Zr-based alloys undergo stress-free growth which accelerates for high irradiation doses (Fig 2). This acceleration is correlated to specific c-component vacancy dislocation loops.

Fig. 1: Changes taking place in the hexagonal close packed grains [1]

Fig. 2: Recrystallized Zr alloys breakaway growth [1]

Since these defects are responsible for the breakaway growth, which influences the performance of the fuel assembly, it is important to know the effect of various factors on their microstructure. The industrial feedback observed on some PWR fuel assemblies suggests that a macroscopic stress applied in the early life could affect the c-loop microstructure and therefore impact the subsequent stress-free growth. In addition, some feedbacks show that in-service hydrogen pick-up could also influence the fuel assembly radiation-induced deformation. In order to have a better understanding of this phenomenon, thorough analytical studies of c-loops nucleation and growth has been undertaken using charged particles irradiation. Protons and ions irradiation experiences have been carried out on various worldwide facilities at several temperatures combined with TEM exams [1, 2].

Two complementary studies were carried out to simulate neutron irradiation: in-situ tensile tests under 1MeV Kr²⁺ ion irradiation and bending experiments under 600keV Zr⁺ ion irradiation in order to investigate the impact of a macroscopic stress applied under irradiation on the evolution of c-loop microstructure [3].

In-situ Kr^{2+} ion irradiation was performed at high dose (up to 25dpa) on RXA Zircaloy-4 at 573K with the stress applied from the beginning of the irradiation. We observe in few grains a clear effect of stress on the c-loop linear density (Fig 3).

Zr+ ion irradiations were performed at 573K on RXA Zircaloy-4 and M5® and two loading histories were considered, using a four point bending device specifically designed to apply a tensile or a compressive stress under irradiation. During the first loading experiment, the external stress was applied at an early stage of irradiation. Transmission Electron Microscopy (TEM) observations showed that the initial applied stress has no effect on the incubation dose of c-loops. For the second loading experiment, the macroscopic stress was applied when c-loops are already created. Same trend, that the one observed for in-situ experience, but with a lower effect of stress on the c-loop linear density was observed on RXA Zircaloy-4 and M5® due to dispersion from grain to grain and a much larger statistic for this experiment (Fig 4) [1]. Nevertheless, qualitative agreement between two experiments is noticed.

Fig. 3: Evolution of c-loops density for Zy4 (In-situ Kr^{2+} ion irradiation) [3]

Fig. 4: Evolution of c-loops density for Zy4 (Bending Zr+ ion irradiations) [3]

Indeed, the c-loop linear density is lower when a tensile stress is applied parallel to the c-axis, which is in good agreement with the Stress Induced Preferential Absorption (SIPA) mechanism. Based on these results under stress level below the transverse yield strength and at 573K, the quantitative statistical study conducted on a large number of Zircaloy-4 and M5® grains with different orientations of the c-axis with respect to the loading direction, shows that an externally applied stress has a slight effect on the c-loop microstructure. A trend for the evolution of their densities with the component along the c-axis of the deviatoric stress tensor was obtained and is consistent with the SIPA mechanism, but seems to be not very significant considering the dispersion from grain to grain. The average loop size is weakly affected by the applied stress and the coupling between the axial irradiation creep and the accelerated irradiation growth appears to be low at the grain scale.

References

- [1] N. Gharbi, F. Onimus, D. Gilbon, J.-P. Mardon, X. Feaugas, "Impact of an applied stress on c-component loops under Zr ion irradiation in recrystallized Zircaloy-4 and M5®", JNM-D-15-00312 (to be published).
- [2] L. Tournadre, F. Onimus, T. Jourdan, J.-P. Mardon, X. Feaugas, "Impact of hydrogen pick-up and applied stress on c-component loops: toward a better understanding of the radiation induced growth of recrystallized zirconium alloys", ASTM STP 1453 (2014), pp. 853-893.
- [3] N. Gharbi, R. M. Hengstler-Eger, X. Feaugas, D. Gilbon, P. B. Hoffmann, M. A. Kirk, J.-P. Mardon, F. Onimus, "The effect of applied stress on c-component dislocation loops in Zr-Based alloys", TMS 144th Annual meeting and Exhibition, March 15-19, 2015, Orlando, USA.

Simulation of c-component dislocation loops by ions and protons irradiation. A tool to understand the breakaway growth of recrystallized Zr-based alloys

Nesrine GHARBI², Thomas JOURDAN², Didier GILBON², Fabien ONIMUS²

² CEA, DEN/DANS/DMN/SRMA/SRMP, 91190 Saclay, France

Xavier FEAUGEAS³

³ LaSI, University La Rochelle, 17000 La Rochelle, France

Jean Paul MARDON¹, Rosmarie Hengstler-Eger⁴

¹ AREVA NP, 10 rue Juliette Récamier, 69456, Lyon Cedex 06, France

⁴ AREVA GmbH, Erlangen, Germany

2nd Int. Workshop Irradiation of Nuclear Materials: Flux and Dose

November 4-6, 2015, CEA – INSTN Cadarache, France

- ▶ **Industrial feedbacks, macroscopic elongation of Zr alloys under neutron irradiation**
- ▶ **Simulation of <c>-component dislocation loops by using charged particles: Zr⁺ and Kr⁺ ions and Protons (feasibility studies)**
- ▶ **Impact of an external tensile and/or compressive stress on <c> loops**
- ▶ **Impact of in-service hydrogen pick-up on <c> loops**
- ▶ **Conclusion**

Industrial Feedbacks

Macroscopic elongation of Zr alloys under neutron irradiation

1. Axial thermal and irradiation creep

2. Irradiation free growth

Macroscopic behavior under flux

Fuel Assembly growth

J.-P. Mardon et al., 2005 Water Reactor Fuel Performance Meeting

F. Garzarolli and al., ASTM STP 1023, 1989

Microstructural evolution of Zr alloys under irradiation

$\langle a \rangle$ dislocation loops

$\langle c \rangle$ dislocations loops

Electron beam

Objectives

Prediction of the macroscopic elongation of fuel assemblies under neutron irradiation

Q1: Coupling under irradiation between 'stress-free' growth and axial irradiation creep ?

Hypothesis: c-loops are responsible for the growth breakaway

Q2: How **macroscopic stress** and **in-service hydrogen pick-up** could influence the growth acceleration?

macroscopic stress applied under irradiation

2 complementary studies

1. In-situ tensile tests under 1 MeV Kr²⁺ ion irradiation
2. Bending experiments under 600 keV Zr⁺ ion irradiation

in-service hydrogen pick-up

Protons irradiations

For the first time <c> loops have been observed in Zr alloys after proton and Zr ion irradiations

L. Tournadre ⁽¹⁾ et al.

TMS2011

► Zr⁺ Ions

RXA Zircaloy-4 at 573 K

CSNSM facility ARAMIS

5.5 dpa

7 dpa

► Protons

RXA Zircaloy-4 623 K

MIBL Tandem

11.5 dpa

➡ Nucleation starts between 4.1 dpa and 5.5 dpa

➡ Growth and nucleation continue with dose increasing

Evolution of <c> loops density vs. irradiation protons dose in Zircaloy-4 and M5® alloys

L. Tournadre ⁽¹⁾ et al.

TMS2011

RXA Zy-4

M5®

➔ Large and faulted <c> loops in consistent densities with increasing dose

➔ Fewer <c> loops in M5® than in RXA Zy-4

FUEL

AREVA

Are charged particles irradiations a good simulation of the neutron damage?

$$L_v = \frac{N \langle d \rangle}{V}$$

Protons 11.5dpa RXA Zy4

PWR 16 dpa RXA Zy-4⁽¹⁾

$\langle c \rangle$ loop linear density L_v evolution with irradiation dose for Zy-4 and M5 after 2 MeV proton irradiations at 623 K

➔ Close to neutron irradiated microstructure

➔ Densities L_v consistent with neutron results

Charged particles irradiations (mainly Protons) are representative of PWR operating conditions

Studied materials

◆ Recrystallized Zircaloy-4 and M5®*

Element (wt%)	Sn	Nb	Fe	Cr	O
RXA Zy-4	~1.30	-	0.22	0.11	0.13
M5®	-	1.00	0.035 – 0.040	-	0.14

◆ Samples taken in the (TD, RD) plane of an intermediate product: TREX

○ Tube of ~11 mm thick

○ Transverse texture

(0002) Pole figure

Equiaxed and recrystallized grains

FUEL * M5® is a trademark of AREVA NP registered in the USA and in other countries

► Impact of an external stress on the c-loops

Specific and appropriate samples

- Sampling for both experiments allowed a large variability of c-axis orientation with respect to the direction of the applied loading

In-situ tests

Bending experiments

In-situ tensile tests under ion irradiation

- ◆ 1 MeV Kr²⁺ ion irradiation at 573 K
- ◆ The entire thickness of the sample is irradiated

Experimental procedure

- ▶ Heating to 300°C
 - ▶ Stress applied at the beginning of the irradiation
- Stretching of the sample until dislocation glide occurs

→ yield strength reached

- ▶ Irradiation with in-situ TEM

Straining sample

Heating /Straining stage

Impact of tensile stress on <c>-loops: sample at YIELD STRENGTH

- ◆ Zircaloy-4 at yield strength, 4 adjacent grains, different orientations
- ◆ Stress level: 256 MPa
- ◆ Final dose: 25-27dpa (SRIM calculation with “full damage cascades” option)

Numerous <c>-loops in grains with <c>-axis far from the tensile direction

FUEL

Few <c>-loops in grains with <c>-axis close to the tensile direction

Impact of tensile stress on <c>-loop linear density and diameter

Zy-4

<c>-loop linear density

<c>-loop average diameter

- ❑ Tensile stress || <c> : lower density
- ❑ Tensile stress ⊥ <c> : higher density
- ❑ Within each sample but for few grains, the <c>-loop density shows a significant decrease with increasing deviatoric stress tensor component in the c-direction
- ❑ Combining the data from all samples, the slope remains the same but dispersion is observed
- ❑ The <c> loop average diameter is not dependent on the deviatoric stress tensor component in the c-direction

BENDING EXPERIMENTS under ion irradiation

- ◆ 600 keV Zr ion irradiation at 573 K
- ◆ Damage created on a thin layer of 300 nm depth
- ◆ Irradiation campaign in two steps

ARAMIS facility at CSNSM Orsay

1

Pre-irradiation $\sigma = 0$ up to 4.1 dpa
(Full Cascades)

Stress applied when c-loops are already created in the microstructure

2

Control samples

- ◆ Two mechanical loading

FUEL

Bending

$$\sigma_{max} = -6 \frac{PB}{bh^2} u$$

$$\sigma_{max} = -6 \frac{PB}{(L-l)(L+2l)}$$

AREVA

Impact of tensile stress on $\langle c \rangle$ -loop MICROSTRUCTURE

Zircaloy-4 @ 7 dpa

TENSILE LOADING

➤ Numerous $\langle c \rangle$ -loops in a grain with c-axis far from the tensile direction

➤ Few $\langle c \rangle$ -loops in a grain with c-axis close to the tensile direction

A tensile stress applied along the c-axis reduces the $\langle c \rangle$ -loop formation

FUEL

Impact of compressive stress on <c>-loop MICROSTRUCTURE

Zircaloy-4 @ 7 dpa

Chargement en compression

●	$\sigma_0 = 138 \text{ MPa}$
■	$\sigma_0 = 83 \text{ MPa}$
▲	$\sigma_0 = 76 \text{ MPa}$
◆	$\sigma_0 = -67 \pm 14 \text{ MPa}$

A compressive stress applied perpendicular to the c-axis **reduces** the <c>-loop formation

➤ Numerous <c>-loops in a grain with c-axis close to the compressive direction

➤ Few <c>-loops in a grain with c-axis far from the compressive direction

FUEL

Quantification of applied stress on c-loop microstructure

Not significant effect of stress on <c> loops microstructure and density

❑ <c>-loop size distribution quite homogeneous in each grain

❑ Large statistical study → dispersion from grain to grain

- Tensile stress $\parallel$ <c> : low density
- Tensile stress $\perp$ <c> : slightly higher density
- Compressive stress $\perp$ <c> : low density
- Compressive stress $\parallel$ <c> : slightly higher density

➤ In agreement with SIPA mechanism

➤ Not significant effect of stress on <c> loops microstructure and density

$$\text{Zy-4: } L_v = -2,37 \times 10^{10} \sigma_D^{<c>} + 8,11 \times 10^{12} \text{ (m}^{-2}\text{)}$$

$$\text{M5®: } L_v = -2,38 \times 10^{10} \sigma_D^{<c>} + 9,88 \times 10^{12} \text{ (m}^{-2}\text{)}$$

Conclusions on stress impact

Questions

Q1: Effect of stress on <c>-loops density ?

Q2: Coupling between irradiation free growth and axial creep ?

1: Low impact of tensile and compressive stress <c> loops microstructure

2: Low coupling between growth breakaway and axial creep at grain scale

☐ Evolution of c-loops density with stress in agreement with SIPA

► Impact in-service hydrogen pick-up on the c-loops

Toward a better understanding of the hydrogen impact on the radiation induced growth of zirconium alloys

How in-service hydrogen pick-up could influence the growth acceleration?

What is the impact of a pre-hydriding on the <c> loop microstructures?

L. Tournadre et al.

TMS2012
141st Annual Meeting & Exhibition

Neutron irradiation simulated by Protons

Michigan Ion Beam Laboratory
FOR SURFACE MODIFICATION AND ANALYSIS

Punching

Hydrogen in the matrix increases the <c> loop density

M5® : effect of Hydrogen in the “matrix” (far from precipitated hydrides)

At 19 dpa – 350°C

In pre-hydrided samples :

- ➡ C-loop microstructures more homogeneous
- ➡ Density slightly higher
- ↻ Also observed at 8.1 dpa and 12.5 dpa

TMS2012
141st Annual Meeting & Exhibition

L. Tournadre et al.

FUEL

Locally high $\langle c \rangle$ loop densities at the vicinity of precipitated hydrides

350wppm H M5[®] – 19 dpa – 2 MeV proton irradiations – 623 K

L. Tournadre et al.

- ➔ Locally high $\langle c \rangle$ loop densities
 - ➔ In some specific areas, $\langle c \rangle$ loop gathered as “bundles”
- ↻ Also observed at 8.1 and 12.5dpa

Hydrogen atoms in solid solution enhances the nucleation / growth of c-loops

M5[®] – 19 dpa – 350°C

Thickness measured by EELS

FUEL

How hydrogen could influence <c> loop microstructure?

- ▶ Hydrogen atoms in solid solution (~100wppm at 623 K in non-irradiated materials) enhances the nucleation and growth of c-loops

↪ Homogeneous distribution and density slightly higher in the “matrix”

- ▶ Locally high <c> loop densities at the vicinity of precipitated hydrides

➡ Hydride dissolution: a source of hydrogen in solid solution

➡ <c+a> remaining dislocations after dissolution: a nucleation site for <c> loops

↪ <c> loop “bundles”

Two main mechanisms ?:

- trapping of hydrogen atoms in solid solution on <c> loop nuclei
- <c+a> remaining dislocations after dissolution

Future prospects deformation modeling under flux

- ▶ **Zr+ or Kr+ Ions and Protons irradiations :**
 - ◆ produce relevant data in much shorter time than neutron (hours/days or weeks / years)
 - ◆ create c-component dislocation loops
 - representative of neutron damage (mainly protons)

- ▶ **An excellent tool :**
 - ◆ to understand the breakaway growth of recrystallized Zr-based alloys
 - ◆ to model FA irradiation growth
 - ◆ Other applications (corrosion, ..)?

Thank you for your attention

Back-up

FUEL

Stress Induced Preferential Absorption Mechanism (SIPA)

- ◆ Influence of the applied stress on the elastic interaction energy between the point defects and the dislocation loops
- ◆ Modification of the capture efficiency of Self-Interstitial Atom (SIA) by a dislocation loop

$$Z_{SIA}^{loop}(\sigma) = Z_0 + C \sum_{i,j} n_i \sigma_{ij}^s n_j$$

W.G. Wolfer, *Journal of Nuclear Materials*, 90 (1980) 175

- Tensile stress applied perpendicular to the c-axis

- ◆ For a vacancy <c>-loop located in the basal plane

$$Z_i^l(\sigma) = Z_0 - \frac{1}{3} C \sigma$$

- Tensile stress applied along the c-axis

$$Z_i^l(\sigma) = Z_0 + \frac{2}{3} C \sigma_0$$

“

Any reproduction, alteration, transmission to any third party or publication in whole or in part of this document and/or its content is prohibited unless Company Name has provided its prior and written consent.

This document and any information it contains shall not be used for any other purpose than the one for which they were provided. Legal action may be taken against any infringer and/or any person breaching the aforementioned obligations.

”