Centre of Excellence for Nuclear Materials

Workshop

Materials Innovation for Nuclear Optimized Systems

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

Fabien ONIMUS et al.

CEA (France)

Impact of Fuel Assembly Transportation on Zirconium Alloys: toward a Mechanistic Understanding

Workshop organized by:

Christophe GALLÉ, CEA/MINOS, Saclay – christophe.galle@cea.fr

Constantin MEIS, CEA/INSTN, Saclay – constantin.meis@cea.fr

Impact of Fuel Assembly Transportation on Zirconium Alloys: toward a Mechanistic Understanding

Fabien ONIMUS¹, Joël RIBIS¹, Brice BOURDILIAU², Chantal CAPPELAERE²

¹ CEA-DEN-DMN, Service de Recherches Métallurgiques Appliquées, SRMA (Saclay, France)

² CEA-DEN-DMN, Service d'Etudes des Matériaux Irradiés, SEMI (Saclay, France)

Zirconium alloys are commonly used in pressurized water reactor as fuel rod cladding tubes. After irradiation and cooling in pool, the spent nuclear fuel assemblies are transported for wet storage to a devoted site. During dry transportation, at temperatures around 400°C, the cladding experiences a creep deformation under the hoop stress induced by the internal pressure of the fuel rod. A recovery of the radiation damage can occur during transportation that can affect the subsequent mechanical properties [1].

The recovery of the radiation damage during heat treatments has been investigated using microhardness tests at room temperature on neutron irradiated cladding materials made of fully recrystallized Zr-1%Nb alloy. Transmission electron microscopy (TEM) observations performed on irradiated thin foils have also shown that, simultaneously with the recovery of the hardness, the dislocation loop density, induced by irradiation, falls while the loop size increases (Fig. 1). Moreover, the TEM analysis has revealed that only vacancy loops are present in the material after long-term annealing, the interstitial loops having entirely disappeared. A numerical cluster dynamic modeling [2] has been used in order to reproduce the material recovery for various annealing conditions (Fig. 1).

Figure 1: Evolution of : (a) the loop density and (b) the loop diameter during a thermal annealing at 350°C, 400°C and 450°C. Experiment (Exp) and modeling (Sim).

Furthermore, the mechanical behavior of the cladding after post-irradiation creep test has been investigated. Creep tests under internal pressure were conducted at 400 and 420°C on the neutron irradiated recrystallized Zr-1%Nb alloy. After depressurization and cooling, ring tensile tests were carried out at room temperature. In addition, transmission electron microscopy observations have been performed after testing [3]. The post-creep mechanical response exhibited a decrease of the strength compared to the as-irradiated material. This decrease is associated with a significant recovery of the ductility, which becomes close to the ductility of the unirradiated material (Fig. 2).

The transmission electron microscopy examinations, conducted on ring samples, revealed that the radiation defects have been annealed (Fig. 3). It was also observed that as for the unirradiated material, the deformation occurred homogeneously throughout the grains. No dislocation channeling was observed contrary to the as-irradiated material [4]. These observations explain the recovery of the strength and of the ductility after post-irradiation creep that may also occur during dry transportation.

Figure 2: Stress-strain curve obtained during ring tensile test on Zr-1%Nb alloy at room temperature.

Figure 3: Dislocation microstructure after ring tensile test subsequent to a post-irradiation creep test at 420°C.

References

- [1] C. Ferry, C. Poinssot, C. Cappelaere, L. Desgranges, C. Jegou, F. Miserque, J.P. Piron, D. Roudil, J.M. Gras. *Journal of Nuclear Materials*, 2006, Volume: 352, Pages: 246–253.
- [2] J. Ribis, F. Onimus, J.-L. Béchade, S. Doriot, A. Barbu, C. Cappelaere, C. Lemaignan. *Journal of Nuclear Materials*, 2010, Volume: 403, Pages: 135–146.
- [3] B. Bourdilliau, F. Onimus, C. Cappelaere, V. Pivetaud, P. Bouffieux, V. Chabretou, A. Miquet. *Journal of ASTM International*, 2011, Vol. 7, No. 9 (16th International Symposium on Zirconium in the Nuclear Industry, ASTM STP 1529, M. Limbäck, P. Barberis Eds, West Conshohocken, PA, 2011, pp. 929-953).
- [4] F. Onimus, I. Monnet, J.-L. Bechade, et al. *Journal of Nuclear Materials*, 2004, Volume: 328, Issue: 2-3, Pages: 165-179.

DE LA RECHERCHE À L'INDUSTRIE

Impact of fuel assembly transportation on zirconium alloys: toward a mechanistic understanding

F. Onimus, J. Ribis, B. Bourdilliau, C. Cappelaere

INDUSTRIAL BACKGROUND : END OF LIFE OF THE FUEL ASSEMBLY

MINOS

Spent Nuclear Fuel Assembly

In-reactor use

Temperature from
400°C up to 450°C

Dry transportation

In-pool storage

Fuel assembly

Neutron irradiation of the
Zr alloy cladding

Post-irradiation creep of the
Zr alloy cladding and
radiation damage recovery

Room temperature mechanical
properties at retrieval ?

Recrystallized Zr-1%Nb alloy

Chemical composition (% wt)

%wt	O	Sn	Fe	Cr	Nb	Zr
RXA Zr-1%Nb	0.12	0	0.02	0	1	Bal.

Onimus F, Siloé, 553 K, $0.6 \times 10^{25} \text{ n m}^{-2}$

Griffiths M. EBR II, 700 K, $1.1 \times 10^{25} \text{ n m}^{-2}$

→ creation of a high density of small loops

Heat treatment → to simulate transportation

(a)

As-irradiated material

$$\langle \rho_b \rangle = 1,2 \cdot 10^{22} \text{ m}^{-3}$$

$$\langle d \rangle = 14 \text{ nm}$$

(d)

After heat treatment
at 400°C during 250 h

$$\langle \rho_b \rangle = 2,5 \cdot 10^{21} \text{ m}^{-3}$$

$$\langle d \rangle = 18 \text{ nm}$$

(f)

After heat treatment
at 450°C during 960 h

$$\langle \rho_b \rangle = 1,2 \cdot 10^{20} \text{ m}^{-3}$$

$$\langle d \rangle = 176 \text{ nm}$$

→ The loop size increases while the density decreases

Cl → Radiation damage recovery

Loop nature using inside / outside contrast method

V as Inside contrast

(a)

V as Outside contrast

(b)

Annealing	Vacancy loops
As-irradiated	50%
350°C 250 h	50%
350°C 500 h	50%
400°C 250 h	65%
400°C 500 h	71%
450°C 960 h	100%

- Only vacancy loops at the end of the recovery
- Interstitial loops recover faster than vacancy loops

What is the loop recovery mechanism ?

LOOP RECOVERY : MECHANISM

As-irradiated microstructure

Shrinking of the vacancy and interstitial loops

Shrinking of the smallest loop at the expense of the biggest loop

Annealed microstructure

Predictive modelling approach : cluster dynamic

Single-vacancy evolution :

$$\frac{dC_v(1)}{dt} = \text{production} - \text{annihilation}$$

No creation of point defects during annealing

Cluster evolution :

$$\frac{dC(n)}{dt} = a_{n-1}C(n+1) - b_nC(n) + c_{n-1}C(n-1)$$

Mean-field modeling

Sinks : clusters, dislocation lines, grain boundaries / free surfaces

Cluster dynamic modelling → computation of loop size and density

Loop density

Loop diameter

→ Correct agreement between experiment and modeling

Evolution of vacancy and interstitial loop size distribution at 400°C

Evolution of the proportion of vacancy loops during heat treatment

Annealing temperature (°C)	Annealing time (h)	Experiment Number of analysed loops	Percentage of vacancy loops (%)	Percentage of vacancy loops (%)
			Modelling	
As-irradiated	As-irradiated	–	50 [23]	50
350	250	19	50	45
350	500	24	50	43
400	250	37	65	100
400	500	38	71	100
450	960	13	100	100

→ Correct agreement between experiment and modeling

High density of small loops
→ pinning of dislocations
→ radiation hardening

$$\Delta\tau_c = \alpha\mu b\sqrt{Nd}$$

Hardness as a function of the loop density & size

→ Radiation hardening recovery during heat treatment
→ Correct prediction of radiation hardening recovery

cea LOOP RECOVERY DURING POST-IRRADIATION CREEP

Post-irradiation creep → to simulate transportation

As-irradiated

Creep test at 400°C during
240 h under 130 MPa

After post-irradiation creep :

- decrease of the loop density
(+ increase of the loop size)
- Radiation damage recovery during creep
- Effect on the tensile mechanical behavior ?

- Irradiation induced hardening (increase of the yield stress and ultimate tensile strength)
- Loss of macroscopic ductility (decrease of the uniform elongation), but the failure remains ductile (strong necking)

→ What are the deformation mechanisms ?

RT tensile test on non irradiated material

→ homogeneous glide of dislocations in the prismatic planes mainly

Ring tensile test at room temperature after irradiation

Channels in the basal plane

Channels in the prismatic planes

- Heterogeneous deformation inside the grains (dislocation channeling)
- both basal and prismatic slip can be activated (depending on the grain orientation)
- > easier basal slip than before irradiation

High density of small loops
 -> pinning of dislocations
 -> radiation hardening

Clearing of loops by gliding dislocations

- > microscopic strain softening
- > early localization of the deformation at the specimen scale
- > decrease of the Uniform Elongation observed during ring tensile tests

-> Why is the basal slip more easily activated after irradiation than before irradiation ?

Loop Burgers vector :

$$\underline{b} = \langle a \rangle = \frac{1}{3} \langle 11\bar{2}0 \rangle$$

Dislocation Burgers vector : $\underline{b} = \langle a \rangle = \frac{1}{3} \langle 11\bar{2}0 \rangle$

B dislocation - loop

⇒ Glissile junction

P dislocation - loop

⇒ Sessile junction

→ Easy Basal channeling / difficult Prismatic channeling

- Recovery of the radiation induced hardening after creep test
- Recovery of the macroscopic ductility (uniform elongation)

→ What are the deformation mechanisms ?

Testing after creep at 400°C

Testing after creep at 420°C

Prismatic glide mainly

After ring tensile test following post-irradiation creep :

- > very few remaining loops → lower radiation hardening
- > homogeneous prismatic glide mainly, no channel
- recovery of the uniform elongation

- Radiation damage recovery :
 - Occurs by exchange of vacancies between loops
 - Shrinking of Int loops and of small Vac loops at the expense to bigger Vac loops
- Cluster dynamic modeling :
 - Correct prediction radiation damage recovery & radiation hardening recovery
- Mechanical behavior after neutron irradiation :
 - decrease of the uniform elongation due to the dislocation channeling
 - failure occurs after a strong necking → ductile failure
 - evolution of activated slip systems explained by junction between dislocations and loops
- Impact of transportation (post-irradiation creep) :
 - radiation hardening recovery due to the loop annealing during creep
 - recovery of the uniform elongation due to homogeneous glide of dislocations
 - better knowledge and understanding of the effects of transportation on the mechanical properties of the fuel assembly at retrieval

Thank you !