



Thermomechanical behavior and modeling of Zircaloy cladding tubes from an unirradiated state to high burn-up

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ABSTRACT. This paper first introduces the modification due to a neutronic irradiation of the thermomechanical behavior of stress-relieved Zircaloy-4 fuel tubes that have been analysed for five different fluences ranging from a non-irradiated material to a material for which the combustion rate was very high. In the second part, we proposed a viscoplastic model able to simulate, for different isotherms, out-of-flux anisotropic mechanical behavior of the cladding tubes irradiated until high burn-up. Finally, results of numerical simulations show the ability of the model to reproduce the totality of the thermomechanical experiments.

I - INTRODUCTION

Today, over 70 % of the French electric power has a nuclear origin and is generated by 58 PWR pressurized water units. This capacity makes it necessary for reactors to adapt their production rate to the network demand by operating under load follow-up conditions. Moreover, due to economic reasons, Electricity of France wants to extend the fuel assembly life span so as to ultimately reach a 60 GWd/tU (Giga-Watt-day/ton Uranium) mass depletion rate. These new conditions of use bring about more severe mechanical loadings of the cladding than initially provided due to a greater increase of the number of power ramps.

Behavior laws currently used for predicting the mechanical behavior of fuel rod claddings do not integrate the whole loading modes inherent in these operating conditions [1]. This requires developing a behavior model capable of more sharply calculating the material anisotropic behavior and more specifically, the stress relaxation in cladding following a loading with a pellet-cladding interaction (PCI) resulting from a power ramp. Such a model should also be fit for taking into account irradiation effects upon the mechanical properties at high-rate fluences which may reach $100 \cdot 10^{24} \text{ nm}^{-2}$ ($E > 1 \text{ MeV}$) and at temperatures ranging from 350 to 400°C, which is the cladding operating temperature range. Deformations are conventionally dealt with as deformations linked to the material growth, deformations occurring under irradiation flux and deformations called "thermal" which are due to power transients. Thermal deformations are chosen for the simulation of cladding behavior resulting from PCI [1].

EDF has launched, in collaboration with some industrial partners, new testing programs on stress-relieved Zircaloy-4 cladding tubes of the same type as those used in PWR power plants. This new data base is now available and has allowed us to develop a behavior model which takes into account the anisotropy, the temperature and the irradiation effects upon the cladding

mechanical properties. The data base includes monotonic tension and uni-axial creep tests, bi-axial loading tension tests followed by a relaxation phase, and bi-axial creep tests.

II - TESTED MATERIALS AND EXPERIMENTAL RESULTS

II.1 Tested materials

Mechanical tests have been carried out on CWSR cladding tubes with nominal dimensions 9.5 mm outside diameter and 0.57 mm wall thickness. The chemical composition of these tubes is given in Table 1. The content of alloying elements is consistent with the ASTM B 353 specification.

Table 1 - Weight composition of Zircaloy-4

Alloying elements (%)					Impurities (ppm)		
Cr : 0.10	Fe : 0.21	Sn : 1.25	O : 0.11	Zr : bal.	C : 120	N : 31	H < 2

The texture analysis reveals a fairly pronounced anisotropy. The Kearns factors show the radial trend for the crystallographic texture : $f_r = 0.57$, $f_a = 0.35$ and $f_z = 0.08$, in radial, tangential and longitudinal directions respectively. By assuming that the texture axis is identical with the anisotropic axis, then the tube has an orthotropic mechanical behavior.

II.2 Mechanical tests realized on unirradiated cladding tubes

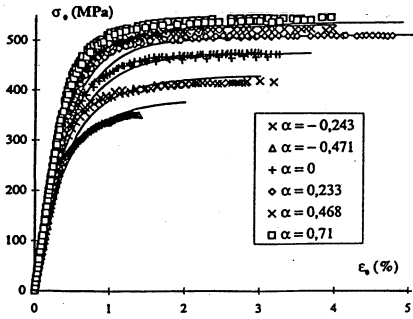
Monotonic axial tension tests have been performed at 350 and 400°C for different strain rates between $2 \cdot 10^{-6}$ and $2 \cdot 10^{-3} s^{-1}$. The experimental tests reveal the strong viscosity of the material at these temperatures. The R^P ratio of the plastic hoop strain to the axial one, $R^P = \epsilon_{\theta\theta}^P / \epsilon_{zz}^P$, is found to be, as a first approximation, independent of temperature and strain rate. To simplify the description of anisotropy in the model, the set of anisotropy coefficients will be considered as independent of temperature and strain-rate, which corroborates the results of Beauregard and al. [2] and Murty [3].

To characterize the anisotropy of these tubes, biaxial tensile tests are performed for different stress-biaxiality ratio α ($\alpha = \sigma_{\theta\theta} / \sigma_{zz}$, hoop stress divided by the longitudinal stress). These tests are carried out with a machine which allow us to impose the biaxiality ratio α , knowing the hoop stress. An example of curves $\sigma_{\theta} = f(\epsilon_{\theta})_{\alpha}$ is given in Fig. 1. Uniaxial ($140 < \sigma_{zz} < 400$ MPa) and biaxial ($100 < \sigma_{\theta\theta} < 175$ MPa) creep tests are also realized.

II.3 Mechanical tests performed out-of-flux on irradiated CWSR cladding tubes

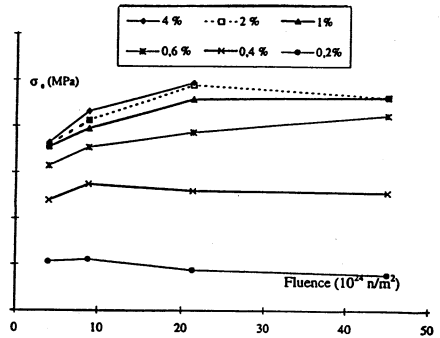
The data base consists of three programs respectively designated by A, B and C. These programs will allow us to quantify the effects of irradiation upon a fuel rod cladding throughout the life span of assemblies up to high burn-up.

- Program A [4]. Test specimens taken from tubes similar to those described earlier are introduced into capsules and irradiated in an experimental reactor. Some test specimens have been removed after 500, 1000 and 3000 hours so as to obtain claddings irradiated at different fluence levels, say about $4 \cdot 10^{24}$, $8 \cdot 10^{24}$ and $21 \cdot 10^{24}$ n/m² ($E > 1$ MeV). The upper fluence level reached is that of a cladding irradiated during one cycle in a PWR reactor. These samples have undergone no hydration or corrosion and have not been exposed to any mechanical loading.

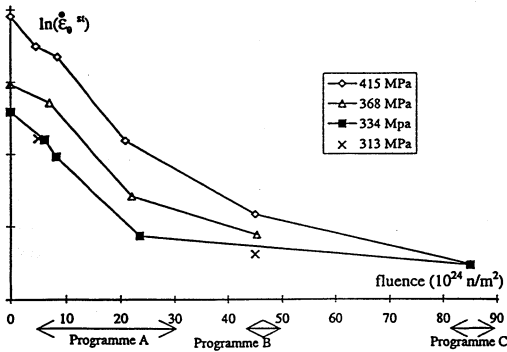


• Fig. 1 : Biaxial tensile tests performed at 350°C for different stress-biaxiality ratio $\alpha = \sigma_{\theta\theta}/\sigma_{zz}$.

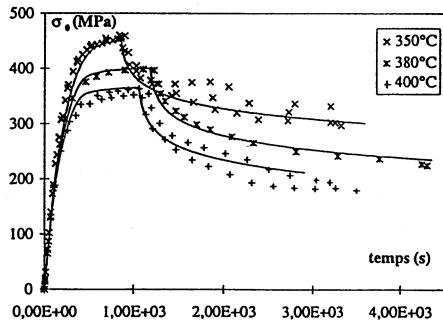
• Fig. 2 : Evolution of the flow stress, for different strains, with the fluence. $\dot{\epsilon}_{\theta} = 5.10^{-6} s^{-1}$ and $T = 350^{\circ}C$.



• Fig. 3 : Steady creep strain rates versus the fluence (Programs A,B,C) at 350°C and for different stress levels.



• Fig. 4 : Biaxial tensile test, $\alpha = 0.5$, with an imposed hoop strain rate $\dot{\epsilon}_{\theta} = 2.10^{-5} s^{-1}$ followed by a relaxation test, for $T = 350, 380$ and $400^{\circ}C$. These loadings are representative of a PCI transient.



Strain hardening tests under internal pressure ($\alpha = 0.5$) have then been conducted at two loading rates, say 2.10^{-4} and 5.10^{-6}s^{-1} at 350 and 380°C. These tests have been completed by twelve creep tests performed at 350°C and three tests at 380°C.

- Program B [5]. Claddings made of CWSR low-tin content Zircaloy-4 are irradiated in a PWR reactor. These claddings come from fuel rods taken from a single assembly following two operating cycles. Test specimens are cut from the fuel rods and a chemical decladding of the fuel is made. These operations therefore allow us to obtain specimens that have been irradiated up to a fluence of 45.10^{24} n/m^2 ($E > 1 \text{ MeV}$). It is worth noting that claddings are corroded and hydrided as they have been in contact with the primary medium. Irradiated test specimens are subject to bi-axial tensile tests ($\alpha = 0.5$) at three required deformation rates : 2.10^{-4} , 2.10^{-5} and 5.10^{-6}s^{-1} , and to creep tests for five stress levels ranging from 300 to 520 MPa at 350°C. Other creep tests have also been performed at 380°C.

- Program C [6]. Tests are conducted on test specimens cut from a fuel rod irradiated during four cycles in a PWR reactor. These claddings are consequently corroded and hydrided too. Bi-axial tensile tests ($\alpha = 0.5$) have been conducted at 350, 380 and 400°C given deformation rates ranging from $1.4 \cdot 10^{-4}$ to $1.4 \cdot 10^{-7}\text{s}^{-1}$. Creep tests have also been realized at these temperatures given tangential stress levels ranging from 300 to 500 MPa.

II.4 Analysis of the results obtained on irradiated claddings

The elastic behavior of the material does not seem to be significantly modified by the neutronic flux. However, the inelastic behavior of the claddings is strongly affected by the irradiation. We have plotted in Fig. 2 the variations of the flow stress required for obtaining a given deformation versus the fluence, at 350°C and at loading rate of 5.10^{-6}s^{-1} . This graph proves that the irradiation makes the material harden at a high rate at fluences ranging from 10. to 20.10^{24} n/m^2 and that it then tends to become stable. The saturation in the evolution of mechanical properties during tensile tests relative to the fluence confirms the conclusions obtained by several authors [7-8]. During the secondary creep phase, strain hardening could be regarded as being constant and it is therefore possible to analyse the recovery phenomenon by analysing the curves of the diametrical deformation steady rate versus the constant stress for different fluences. From this representation it is possible to obtain the Fig. 3 which shows how the secondary creep rate varies at 350°C as a function of the fluence. We plot on this figure the points derived from tests performed within the framework of programs A, B and C. We see that the creep rate becomes much lower before stabilized for high fluences according to the results given in references [9-11]. Note that the whole results seem therefore consistent (Fig. 2 and 3).

III - MODELING AT DIFFERENT TEMPERATURES OF THE BEHAVIOR OF IRRADIATED ZIRCALOY-4

We worked with a behavior model developed on other types of materials and adapted to the Zircaloy case [12-13]. The equations of the model are given in Table II. By admitting the case of small deformations, the total strain can be splitted into two terms, an elastic strain [ϵ^e] considered isotropic and a viscoplastic strain [ϵ] which is anisotropic. Experimental studies including cyclic tests clearly indicated the kinematical nature of the hardening. So, the stress is the sum of two components, a viscous stress $\sigma_v = \sigma - \alpha$ and an internal stress [α]. Three kinematical variables are necessary to simulate the strong linearity of the mechanical behavior of Zircaloy. The evolution laws of the kinematical hardening variables [α], [$\alpha^{(1)}$] and [$\alpha^{(2)}$] define the mechanical behavior completely. The first term of these equations is the linear part

of the hardening, the second term represents the dynamic recovery and the last one represents the time dependent static recovery. The introduction of the anisotropy in this model is made via the four fourth rank tensors affecting the flow directions $[\underline{M}]$, the linear part of kinematical hardening $[\underline{N}]$, as well the dynamic and static recoveries, $[\underline{Q}]$ and $[\underline{R}]$, of these same hardening variable. Y is a scalar variable which describe the hardening of the material during cyclic loadings.

Irradiation causes a marked hardening while reducing the creepability of the material. To take into account the saturation observed in the evolution of the mechanical properties, we do not use the fluence ϕ as a damage variable but a state variable denoted D in table II. A number of model parameters will depend upon this variable. Note that such a formulation of the variable D corresponds to an isotropic representation.

Table II : Equations of the model

$$[\dot{\epsilon}^T] = [\dot{\epsilon}^e] + [\dot{\epsilon}]$$

$$[\dot{\epsilon}^e] = \frac{1+\nu}{E(T)} [\dot{\sigma}] - \frac{\nu}{E(T)} [\Delta]^t [\dot{\sigma}] [\Delta]$$

$$[\dot{\epsilon}] = \frac{3}{2} \bar{\epsilon} \frac{[\underline{M}][\sigma' - \alpha']}{\sigma - \alpha} \text{ with } \bar{\epsilon} = \dot{\epsilon}_0(T, D) \left[\sinh \left(\frac{\sigma - \alpha}{N(T, D)} \right) \right]^n$$

In these equations : $[\sigma'] = [\sigma] - \frac{1}{3} [\Delta]^t [\sigma] [\Delta]$, $[\alpha'] = [\alpha] - \frac{1}{3} [\Delta]^t [\alpha] [\Delta]$,

$$\overline{\sigma - \alpha} = \left\{ \frac{3}{2} [\sigma' - \alpha']^t [\underline{M}] [\sigma' - \alpha'] \right\}^{1/2}$$

- Equations of the kinematical hardening variables $[\alpha]$, $[\alpha^{(1)}]$ and $[\alpha^{(2)}]$

$$[\dot{\alpha}] = p \left(\frac{2}{3} Y^*(T) [\underline{N}][\dot{\epsilon}] - [\underline{Q}][\alpha - \alpha^{(1)}] \bar{\epsilon} \right) - r_m(T, D) \sinh \left(\frac{\bar{\alpha}}{\alpha_0(T, D)} \right)^{m_0} [\underline{N}][\underline{R}] \frac{[\alpha]}{\bar{\alpha}}$$

$$[\dot{\alpha}^{(1)}] = p_1 \left(\frac{2}{3} Y^*(T) [\underline{N}][\dot{\epsilon}] - [\underline{Q}][\alpha^{(1)} - \alpha^{(2)}] \bar{\epsilon} \right)$$

$$[\dot{\alpha}^{(2)}] = p_2 \left(\frac{2}{3} Y^*(T) [\underline{N}][\dot{\epsilon}] - [\underline{Q}][\alpha^{(2)}] \bar{\epsilon} \right) \text{ with } [\dot{\alpha}^{(k)}]_{(0)} = 0 \text{ and } \bar{\alpha} = \left\{ \frac{3}{2} [\alpha]^t [\underline{R}] [\alpha] \right\}^{1/2}$$

- Equations of the scalar variable Y^*

$$Y^* = Y_0(T) + Y$$

$$\dot{Y} = b(Y^{\text{sat}} - Y) \bar{\epsilon}$$

- Equation of the irradiation damage variable D

$$\dot{D} = D_1(T^*)(D_0(T^*) - D)\phi \quad , \quad D(0) = 0$$

The integrated form is given by : $D = D_o(T^*)(1 \exp - D_1(T^*)\phi)$.

In these equations, ϕ is the neutronic flux, ϕ the fluence and T^* the irradiation temperature which can be different from the temperature T of the test. The evolution of $D_o(T^*)$ and $D_1(T^*)$ is not specified, these two parameters being considered as constant in the present study.

- Temperature dependence of the parameters

$$E(T) = E_o + E_1 T$$

$$\alpha_o(T) = \alpha_1 + \alpha_2 T$$

$$\dot{\epsilon}_o(T) = \dot{\epsilon}_1 \left(\exp - \frac{\Delta H}{kT} \right) (N(T))^n$$

$$r_m(T) = r_{m0} \left(\exp - \frac{\Delta H^*}{kT} \right) (\alpha_o(T))^{m_0}$$

$$N(T) = N_o + N_1 T$$

$$Y_o(T) = Y_1 + Y_2 T$$

- Irradiation dependence of the parameters

$$\dot{\epsilon}_o(T, D) = \dot{\epsilon}_o(T) (N(T, D))^n \exp(-\xi_1 D^{\xi_2})$$

$$r_m(T, D) = r_m(T) \exp(-\chi_1 D^{\chi_2})$$

$$N(T, D) = N(T) + N_2(1 - \exp - N_3 D)$$

$$\alpha_o(T, D) = \alpha_o(T) \exp(-\alpha_3 D^{\alpha_4})$$

In this formulation the Voigt matrix notation is adopted :

$$[\sigma]^t = [\sigma_{11} = \sigma_1, \sigma_{22} = \sigma_2, \sigma_{33} = \sigma_3, \sigma_{12} = \sigma_4, \sigma_{13} = \sigma_5, \sigma_{23} = \sigma_6]$$

$$[\epsilon]^t = [\epsilon_{11} = \epsilon_1, \epsilon_{22} = \epsilon_2, \epsilon_{33} = \epsilon_3, 2\epsilon_{12} = \epsilon_4, 2\epsilon_{13} = \epsilon_5, 2\epsilon_{23} = \epsilon_6]$$

$$[\Delta]^t = [1, 1, 1, 0, 0, 0]$$

$[M]$, $[N]$, $[Q]$, $[R]$ have an orthotropic symmetry. For example for $[M]$:

$$[M] = \begin{bmatrix} M_{11} & M_{12} & M_{13} & & & \\ M_{13} & M_{22} & M_{23} & & & \\ M_{13} & M_{23} & M_{33} & & & \\ & & & M_{44} & & \\ & & & & M_{55} & \\ & & & & & M_{66} \end{bmatrix}$$

with the incompressibility relations :

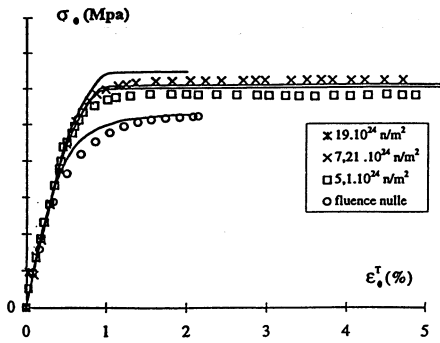
$$M_{11} + M_{12} + M_{13} = 0$$

$$M_{13} + M_{22} + M_{23} = 0$$

$$M_{13} + M_{23} + M_{33} = 0$$

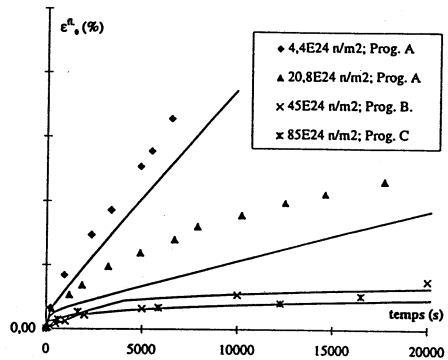
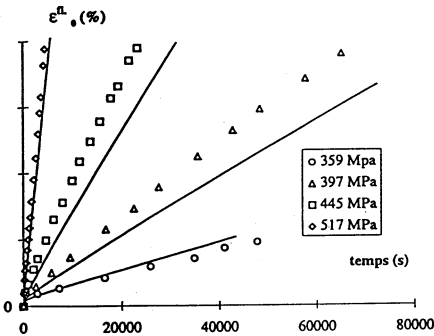
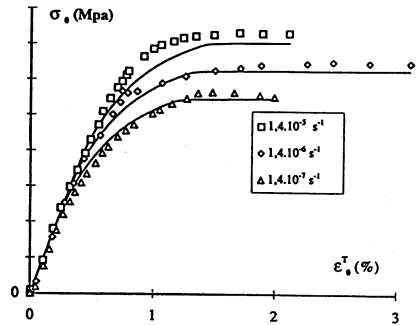
IV - IDENTIFICATION OF THE MODEL AND SIMULATIONS

The parameters of the model are obtained by numerical simulations on uni and biaxial tensile and creep tests at 350 and 400°C. The evolutions of parameters with the temperature are verified by comparing simulations with tests results at 380°C. On Fig. 1 the model is in good agreement with biaxial tensile tests performed at 350°C with different ratios α . Thus the description of the anisotropy of the tube is correct. The same adequation between tests and predictions has been obtained for three complex loadings realized at 350, 380 and 400°C, Fig. 4, which are representative of a PCI transient : biaxial tensile test, $\alpha = 0.5$, with an imposed hoop strain rate of $2 \cdot 10^{-5} s^{-1}$ followed by a relaxation test. The Fig. 5 illustrates the capability of the model to simulate biaxial tensile tests at 350°C for different fluences. The results are fairly good. In the same way, the Fig. 6 and 7 show that the model is able to predict the mechanical behavior of irradiated cladding tubes at 350 and 380°C.



• Fig. 5 : Biaxial tensile tests performed at 350°C for an imposed hoop strain rate of $2.10^{-4} s^{-1}$ for different fluences. Experimental results and simulations .

• Fig. 6 : Modeling of a tensile-internal pressure test for different strain rates at 380°C, performed on irradiated cladding tubes until 4 cycles in PWR (Program C) .



• Fig. 7 : Simulation of tensile-internal pressure tests performed at 350°C on irradiated cladding tubes :
 - a - irradiated 1000 H, for different stress levels
 - b - $\sigma_{00} = 415$ MPa, for different fluences (Programs A,B,C) .

V - CONCLUSIONS

The analysis of the out-of-flux experimental data basis composed of biaxial tensile, creep and relaxation tests carried out on irradiated CWSR cladding tubes shows the hardening and a decrease in creep deformation due to neutron irradiation. These irradiation effects are introduced into this model through an internal state variable of damage which is a function of the fluence. Finally, we show the ability of the modeling to calculate the anisotropic behavior of cladding tubes during tensile, high hoop stress levels creep and relaxation tests in the temperature range 350-400°C and fluences range $0-85.10^{24}$ n/m² ($E > 1$ MeV). This model is already introduced in the EDF finite element Code-Aster and will be implemented in the cladding tube calculation code of EDF, Cyrano 3 to allow the calculation of PCI transients.

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