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CREEP BEHAVIOR AT HIGH TEMPERATURE OF THE OXYGEN STABILIZED ZIRCONIUM ALPHA PHASE OF FUEL CLADDING TUBES OXIDIZED IN LOCA CONDITIONS

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During hypothetical Loss-Of-Coolant-Accident (LOCA) scenarii, zirconium alloy fuel cladding tubes are oxidized at high temperature (HT): zirconia and oxygen-stabilized alpha, called alpha(O) are formed on the outer surface of the cladding. To model the behavior of the cladding during LOCA transients, the specific behavior of each layer must be known. This study focuses on the characterization of the viscoplastic behavior of the alpha(O) phase at HT. Model alpha(O) phase specimens enriched by 2 wt% in oxygen were produced from zirconium alloys cladding specimens. Creep tensile tests were performed in the tube axial direction under secondary vacuum between 900 and 1100°C for stresses between 2 and 31 MPa. The creep resistance of the alpha(O) phase is higher than that of the beta phase by three to four decades. The HT creep behavior of oxidized cladding was then modeled by finite element simulation.

I. INTRODUCTION

Understanding interactions between oxidation and mechanical behavior is necessary to predict the mechanical response of zirconium-based fuel claddings during a Loss-Of-Coolant-Accident (LOCA) hypothetical situation in Pressurized Water Reactors (PWR). In such a situation, fuel cladding tubes undergo a high temperature transient (potentially up to 1200°C) in an oxidizing environment and swell due to an internal pressure loading. Current safety requirements focus on cladding tube burst and swelling, which must be avoided or limited in order to guarantee fuel containment and core cooling[1].

Oxidation at high temperature induces the formation of two layers in addition to the metallic beta phase: an outer layer of zirconia and a metallic sublayer of oxygen stabilized alpha phase, called alpha(O) (Figure 1), containing 2 – 7 wt% of oxygen[2].

The usual-phenomenological behavior laws used in the modeling of fuel claddings behavior in LOCA conditions are established for materials with low oxidation levels. Experimental works on highly oxidized materials will allow us to understand and quantify the role played by the oxidation. In the long term, a physics-based simulation tool modeling cladding's behavior in LOCA conditions should be built.

The creep behavior of zirconium and of its alloys have been extensively studied in an inert environment and are well understood, even though some interrogations remain in the alpha + beta domain[3,4,5]. Several studies were conducted on the effect of oxygen on the mechanical behavior of Zircaloy-2 at high temperature. Rizkalla *et al.*[6] and Choubey *et al.*[7] performed compressive tests between 750°C and 1400°C and Choubey *et al.*[7] showed that the compressive yield stress exponentially increases with increasing oxygen concentration (up to 1.2 wt%) in alpha and beta phases. Similar exponential influence of oxygen was found for the creep resistance of the alpha phase by Burton *et al.*[8] between 700 and 800°C but they observed a negligible effect on the creep behavior of the beta phase between 1050 and 1250°C. Comparable results were obtained with Zircaloy-4 by Chow *et al.*[9] between 650 and 1400°C. These studies were restricted to oxygen content up to 1.5 wt%. Only the dislocation creep regime was observed in both alpha(O) and beta phases and it was reported that this range of oxygen concentration does not significantly affect the stress dependence of the creep rate. One study[10] investigated oxygen concentrations up to 2 wt% by performing compressive tests. The creep behavior of the alpha phase enriched in oxygen for higher contents has not been reported yet.

This study aims at modeling with physic-based tools the mechanical response at high temperature of the oxidized cladding tube seen as a stratified material. The behavior of the alpha(O) phase at high temperature was studied first for the lower oxygen content, i.e. 2 wt%, in order to define a lower bound of the strengthening effect

of the alpha(O) layer. Model alpha(O) specimens enriched by 2 wt% in oxygen were produced from zirconium alloys cladding specimens. To characterize their creep behavior at high temperature, tensile tests were performed along the axial direction under secondary vacuum. Viscoplastic flow rules have been identified. The creep behavior of the fuel cladding tube at high temperature and under internal pressure was then simulated by using finite element analysis (FEA) and the impact of the alpha(O) layer, for several thicknesses, has been estimated.

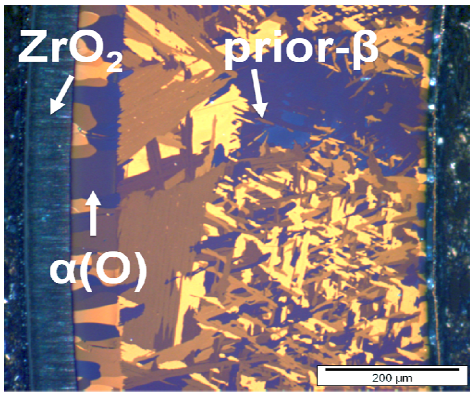

Fig. 1. Optical micrograph of a Zy-4 cladding after creep test in steam at 1100°C for 10 min. Outer surface on the left, tube axis perpendicular to the image.

II. EXPERIMENTAL PROCEDURES

II.A. Creep tests in vacuum on model alpha(O) cladding specimens

Model alpha(O) phase specimens enriched by about 2 wt% of oxygen were produced from zirconium-based alloy cladding specimens thanks to a two-step procedure: an oxidation followed by a homogenization heat treatment. First, cladding specimens were oxidized in steam at 1100°C in order to get the required oxygen content as a zirconia layer. By using optical and scanning electron microscopy, the thicknesses of the resulting layers of zirconia and alpha(O) were found to be consistent with previous oxidation data[11].

Then, oxidized specimens were put in an electrical-mechanical machine[3] under load control and under secondary vacuum (10^{-3} – 10^{-4} Pa). Two cement-pasted alumina rings delimited the 20 – 25 mm gauge region over which axial elongation was continuously measured using a laser extensometer. Temperature was monitored using S-type thermocouples spot-welded onto the specimen surface at various locations. The temperature gradient along the gauge length was 10±5°C. Specimens underwent the homogenization heat treatment at 1200°C for 3 hours under a very low tensile load in order to prevent from buckling. During this treatment, zirconia dissolved and oxygen diffused through the specimen wall[12].

Right after the homogenization heat treatment, temperature was decreased down to that of tensile creep tests and a constant load was applied (Figure 2). As already done for unoxidized specimens[3], different load jumps were applied and thus several axial stresses and corresponding strain rates were identified from each test. Loading history effects were quantified in one test by using increasing, then decreasing load levels. The difference in true strain rate versus true stress curves was lower than 30%. Investigated stresses and temperatures respectively ranged from 2 to 31 MPa and from 900 to 1100°C.

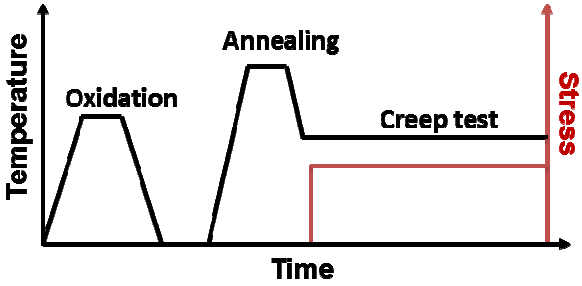

Fig. 2. Scheme of the thermal-mechanical history of the oxygen-enriched creep specimens.

II.B. Creep test in steam

One creep test in steam was performed on stress-relieved Zircaloy-4 cladding in the EDGAR test facility[13]. A sealed tubular specimen was heated by Joule effect (~20°C.s^{-1}) up to 1100°C and temperature was monitored by using an optical pyrometer. Once the test temperature stabilized (~20 s), a constant internal pressure of 5 bar was imposed. The values of temperature and applied pressure were chosen to enhance oxidation and its possible effects on the viscoplastic response of the cladding. The increase in outer diameter was continuously measured by a laser extensometer at mid-length.

II.C. Metallurgical analysis

Using in-house experimental procedures[11], optical and SEM observations were performed on polished and etched samples. For oxygen quantification, electron probe microanalysis (EPMA) was systematically used on tested enriched specimens with a ±0.1 wt% typical accuracy[11]. Texture was studied thanks to electron backscatter diffraction (EBSD) analysis, on a specimen sectioned perpendicular to the tube axis. In view of the microstructure of Figure 1, the size of the sampled region was about 2 mm^2, with a step size of 2 µm, covering about 500 grains.

III. RESULTS AND DISCUSSION

III.A. Creep tests in vacuum on model alpha(O) cladding specimens

III.A.1. The oxygen-enriched cladding microstructure

EPMA analysis confirmed that the oxygen concentration was uniform through the wall, ranging from 2 to 2.5 wt%. Figure 3 shows the outer part of the cladding, where there is no remaining zirconia. The material microstructure is composed of large alpha(O) grains, their size ranges from 30 to 200 µm. The alpha(O) phase predominates with a phase fraction of 85±5%. However, a 15±5% residual fraction of prior-beta phase remains close to the inner surface after the annealing treatment, whatever the subsequent creep test temperature. This suggests that this amount of beta phase was already present at the annealing temperature of 1200°C. This point is in agreement with previous studies on oxygen-enriched Zircaloy-4[9,14,15] (Figure 4). Based the curves of Figures 4, one expects a 10 – 25% beta phase fraction at 1200°C for a global 2 – 2.5 wt% oxygen concentration. On the other hand, diffusion might not have been sufficiently rapid to dissolve this beta phase during the creep tests performed at lower temperature (900 - 1100°C). The corresponding fraction of beta phase was thus assumed to be present at high temperature during the entire creep test. It had to be taken into account in the interpretation of the mechanical results.

III.A.2. Creep ductility of the oxygen-enriched material in vacuum

From the creep tests, the 2 wt% oxygen-enriched cladding can sustain significant strain levels when tested in axial creep between 900 and 1100°C. Moreover, it was not possible to obtain the ductility at fracture for each specimen due to the limited elongation allowed by the extensometry device. A creep test at 900°C was stopped without failure after a 15% axial strain. Another test was performed at 1000°C up to failure. Laser profilometry of the broken specimen indicated a 22% diametral strain in the necking region while SEM observation revealed a local reduction in thickness of almost 100% at fracture, together with some dimples (Figure 5). On the one hand, such ductile behavior contrasts with the brittle behavior reported for zirconium-based alloys with similar and higher oxygen contents at lower temperatures, 25 and 135°C[12]. On the other hand, Tseng *et al.*[10] reported strain values higher than 8% during compression tests performed between 850 and 1100°C on 1.8 wt% oxygen-enriched Zircaloy-4, so that high-temperature ductility of such specimens is in agreement with scarce available literature data.

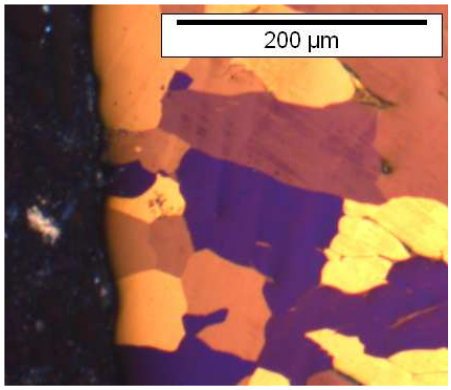

Fig. 3. Optical micrograph of an oxidized, then homogenized sample containing 2 wt% of oxygen. Same orientation as in Figure 1, outer surface on the left.

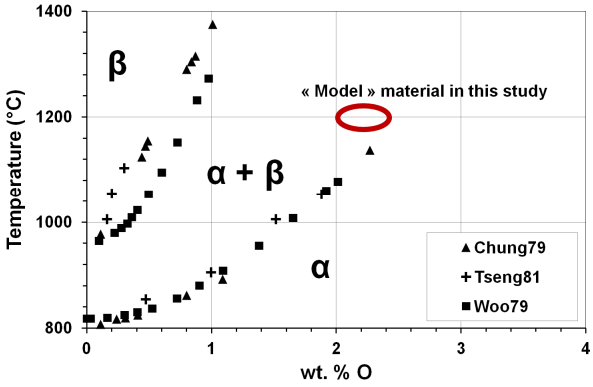

Fig. 4. Zircaloy-4 – oxygen isopleth from the equilibrium phase diagram[9,14,15]. The oxygen content and homogenization temperature are indicated with a red ellipse.

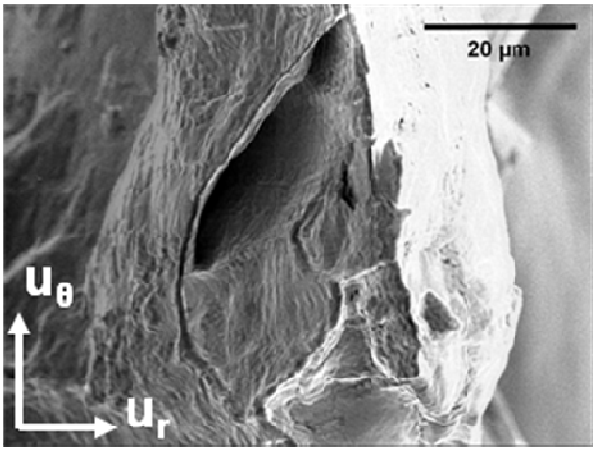

Fig. 5. SEM micrograph of the fracture surface of the 2 – 2.5 wt% oxygen enriched-material broken at 1000°C.

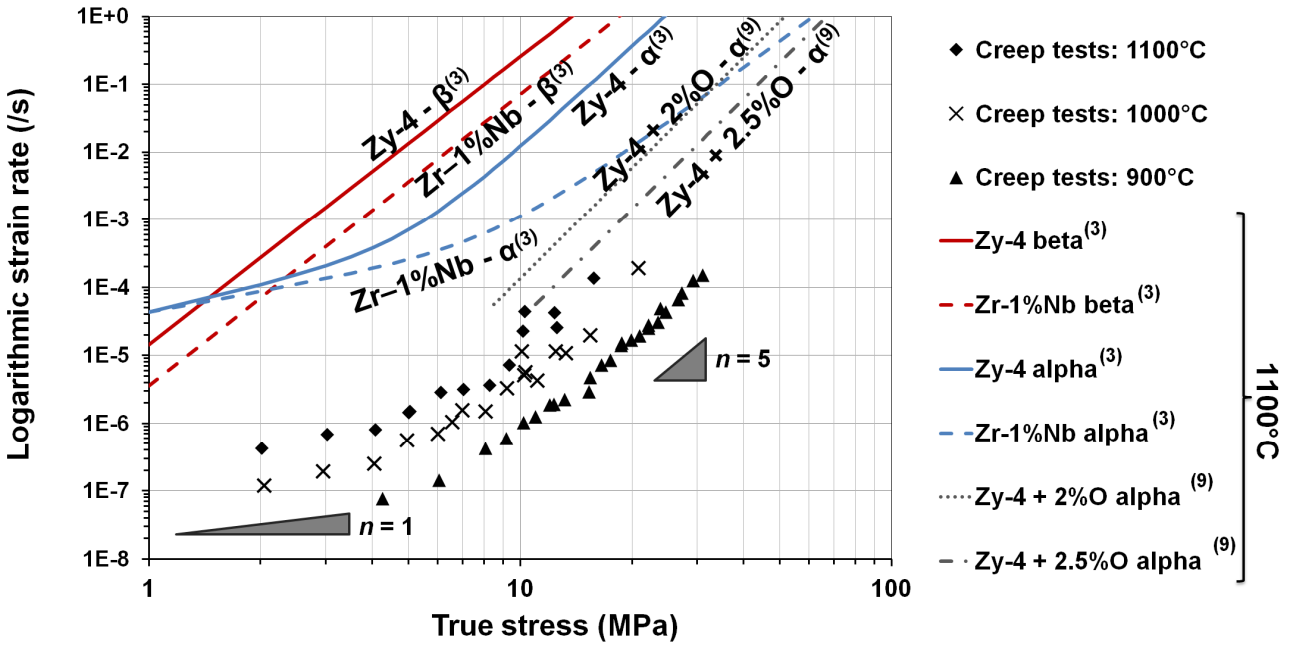

Fig. 6. Stress dependence of the strain-rate: present experimental data (symbols) for oxygen-enriched material, predictions at 1100°C for zirconium alloys which are non-enriched[3] (color lines) or enriched with oxygen[9] (grey lines).

III.A.3. Creep flow behavior of the oxygen-enriched material in vacuum

For every considered load level, only steady-state creep was observed. Creep results in the axial direction in vacuum are summarized in Figure 6. Two creep regimes are observed. For higher stresses ($\sigma > 10$ MPa), the stress exponent n is close to 5 and, thus, suggests a "dislocation creep" regime[16]. For lower stresses ($\sigma < 5$ MPa), data are consistent with a stress exponent n close to 1. These two macroscopic creep regimes are similar to the ones reported at lower temperatures in the alpha phase that is non-enriched in oxygen[3]. However the grain size is rather coarse in the oxygen-enriched material and thus the linear regime observed here might result from another physical mechanism than a diffusional flow[16]: a Harper-Dorn mechanism for example In order to distinguish between these two possible mechanisms, materials with various grain sizes should be tested, which is out of the scope of the present study. Based on values of n respectively equal to 1 and 5, experimental results were found to be well described by the following power-law equation:

$$\dot{\varepsilon} = \frac{A}{T}\sigma^n \exp\left(-\frac{Q}{RT}\right) \qquad (1)$$

The apparent values of activation energy Q are both in agreement with those previously reported in non-enriched alpha phase in Zircaloy-4 and Zr–1%Nb[3].

For comparison purposes, creep flow results for alpha and beta single-phase of typical zirconium alloys non-enriched in oxygen, also tested along the axis of the cladding tubes are also plotted in Figure 6. The alpha phase behavior was extrapolated to higher temperatures from reported data[3]. Indeed, in this temperature range alpha single phase does not exist without oxygen addition. The creep resistance of the 2 – 2.5 wt% oxygen enriched material is much higher than that of zirconium alloys non-enriched in oxygen (i.e. containing 0.12 - 0.14 wt% of oxygen).

Creep properties established by Chow *et al.*[9] for alpha phase of Zircaloy-4 containing 0.14 – 1.5 wt% of oxygen are also displayed in Figure 6. They are based on experimental creep results ranging from 650 to 1000°C and for strain rates between 5.10^{-5} and 7.10^{-2} s^{-1}. Therefore, the transition to a nearly linear creep regime could not be expected from these existing results. Nevertheless, the expected strengthening effect of the oxygen weight concentration c in the dislocation creep regime, modeled by Chow with an exponential term (Eq. 2 and 3) overestimates the strain rate by a factor of about 3 with respect to the present experimental values obtained on a biphasic material (85% alpha(O)) containing 2 – 2.5 wt% of oxygen.

$$\dot{\varepsilon}(c) = \dot{\varepsilon}_0 . \exp(-Bc) \qquad (2)$$

$$\dot{\varepsilon}_0 = 2.03\times10^5 \times \sigma^{5.43} \exp\left(-\frac{320000}{RT}\right); B = 2.8 \qquad (3)$$

III.A.4. Estimate of the creep behavior of the alpha(O) phase layer alone

Our purpose is to describe the behavior of the alpha(O) phase layer during a LOCA transient – internal pressure at high temperature – in order to model the three-layered oxidized cladding. This approach will reduce the uncertainties of the modeling in high oxidation ranges. Performing tensile creep tests in vacuum allowed us to establish viscoplastic flow rules of the 2 wt% oxygen-enriched material. These equations can be used to describe the alpha(O) layer of an oxidized cladding only after discussing the three following points:

- The mechanical influence of the small amount (15±5%) of beta phase present in the model material but not in the alpha(O) layer of claddings oxidized in steam.
- The possible difference in texture (arising from difference in phase transformation history) and in mechanical loading (axial tension vs. internal pressure).
- The investigated 2 wt% oxygen concentration lower than the 4 wt% mean oxygen content in the alpha(O) layer[2] only gives a lower bound of the strengthening effect of the alpha(O) layer on the three-layered structure.

Mechanical influence of the 15±5% beta phase

A homogenization approach was used to evaluate the impact of the 15±5% remaining fraction of beta phase on the viscoplastic flow behavior of oxygen enriched specimens. The fact that the experimental creep rates are strongly lower than the ones for as-received beta and alpha phases (up to 4 orders of magnitude) indicates that the major alpha(O) phase is stronger than the beta phase (which cannot dissolve high amount of oxygen, Figure 4) and probably controls the global mechanical response of oxygen-enriched specimens.

With such a contrast in viscoplastic behavior between alpha(O) and beta phases, and by considering the high fraction of alpha(O) phase, an inverse approach based on a two-phase mean field model with a Taylor assumption[17] – homogeneous strain rate – can give a relevant estimate of the viscoplastic flow behavior of the alpha(O) phase alone. The viscoplastic flow rule of the beta phase of Zircaloy-4 was taken from tensile creep results on cladding tubes reported by Kaddour *et al.*[3] as in Figure 6. As a starting point, flow rule coefficients of the alpha(O) phase were taken from Table 1. Then, assuming that the strain rate was homogeneous, i.e. equal in the two phases (Eq. 4), stresses in the alpha and beta phases (respectively, σ_α and σ_β) and the macroscopic stress, Σ, were numerically calculated (from above flow rules and by using Eq. 5) using a least squares iterative method.

$$\dot{\mathrm{E}} = \dot{\varepsilon}_\alpha = \dot{\varepsilon}_\beta \qquad (4)$$

$$\Sigma = (1 - f_\beta)\sigma_\alpha + f_\beta \sigma_\beta \qquad (5)$$

where f_β is the fraction of beta phase.

By these means, the influence of the beta phase on the viscoplastic flow rule was estimated to a factor two in the higher stress regime, and was negligible in the lower stress regime. This factor two was thus applied in the higher stress range only by dividing by two creep coefficient *A* of Eq. 1 – identified from the axial creep tests – leading to the curves of Figure 7. From this point, it appears that the model by Chow *et al.* overestimates the strain rate by a factor 6.

Influence of texture and mechanical loading mode

By considering the anisotropy in mechanical properties of the hexagonal alpha phase, transfer of the obtained viscoplastic flow rules to another mechanical loading mode and material texture has to be done carefully. EBSD analysis revealed that the textures of both our oxygen-enriched specimens and the alpha(O) layer of cladding pressurized in steam are comparable: in both, <c> axes were perpendicular to the cladding axis. However axial tension differs from biaxial loading resulting from prescribed internal pressure. Nevertheless, stress and strain anisotropy of oxygen-enriched alpha(O) at 900 – 1100°C cannot be inferred from that of more conventional alpha phase, for which no data are available at such high temperature. Consequently, this effect could not be numerically taken into account in the present study and the alpha(O) layer was considered as isotropic.

Oxygen concentration effect

The oxygen concentration varies between 2 and 7 wt% within the alpha(O) layer of an oxidized cladding[2]. Therefore, the identified flow equations for 2 wt% give a lower bound of the strengthening effect of oxygen. Further studies are under way to assess the experimental creep behavior of model materials with higher oxygen content. An evaluation of the upper bound strengthening effect for 7 wt% oxygen content based on the extrapolated strengthening law of Chow *et al.*[9] is shown in part IV.

III.B. Creep test in steam

In a steam environment, at the beginning of the creep test, the observed strain rate is similar to the one observed under vacuum. After a short time (~40 s) the increase in diameter is strongly slowed down (Figure 8) – significant oxidation having already started during heating and temperature stabilizing stage. The predicted weight gain in oxygen according to the data from Brachet *et al.*[11] is plotted in the same figure. From the tests carried out in vacuum[3,4], no "primary creep stage" is expected, nor observed, in this range of stress level and test

temperature. This is a clear demonstration that the combined formation of zirconia and alpha(O) reinforces the oxidized cladding with respect to creep. Such strengthening was also estimated by modeling in the following part.

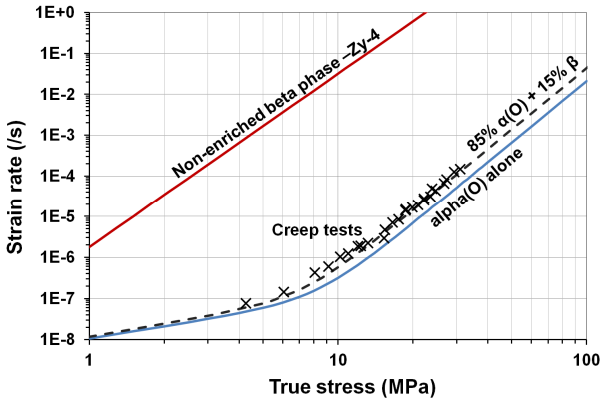

Fig. 7. Creep test results (symbols) and viscoplastic flow rules (lines), all determined in uniaxial tension along the tube axis at 900°C.

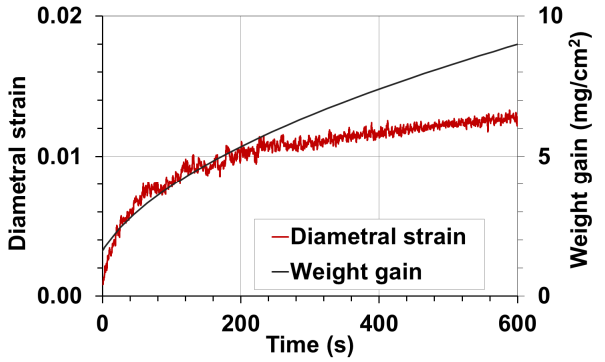

Fig. 8. Predicted weight gain based on data from Brachet *et al.*[11] versus time, together with creep strain of Zy-4 at 1100°C and 5 bar of internal pressure in steam.

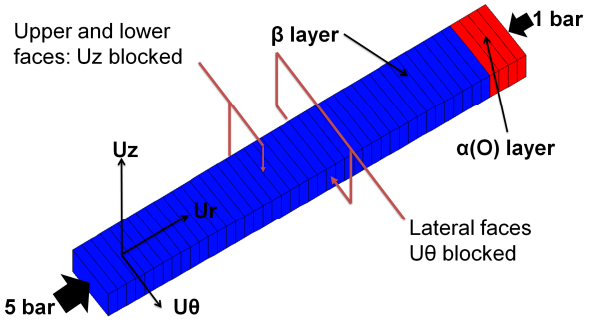

Fig. 9. Mesh, loading and boundary conditions used in FE simulations of internal pressure loading at 1100°C.

IV. MODELING OF THE CREEP BEHAVIOR OF THE OXIDIZED CLADDING SEEN AS A STRATIFIED MATERIAL

IV.A Constitutive equations

The Zircaloy-4 one-side oxidized cladding can be considered as a three-layer-stratified material at high temperature (1100°C): the inner beta layer, an intermediate alpha(O) layer and an outer zirconia layer. In the present study, the mechanical contribution of zirconia could not be estimated, its mechanical strength being unknown at the studied temperatures. This contribution can possibly be evaluated by inverse analysis once the creep strength of the alpha(O) layer with its gradient in oxygen content will be fully elucidated. In the present calculations that aim at providing a lower bound to environmentally-induced creep strengthening, only two layers were considered in the model, namely, an inner beta layer and an outer alpha(O) layer. Their creep flow rules are respectively given by Kaddour *et al.*[3] for the beta phase of Zircaloy-4 and by corrected creep flow equations based on our experimental results for the 2 wt% alpha(O) or by the extrapolated Chow creep laws for the 2 – 4 – 7 wt% alpha(O) layer. Moreover the effect of oxygen atoms having diffused into the beta phase on its creep behavior was neglected, in agreement with the results of Burton *et al.*[8] and Chow *et al.*[9]. In absence of experimental data about creep anisotropy of this layer, an isotropic von Mises equivalent stress was used in the viscoplastic potential. No work-hardening and threshold stress were introduced, as experimentally observed in this range of temperature and stress levels. The test temperature of 1100°C was chosen for creep simulations according to experimental conditions.

IV.B Finite element model and numerical procedures

Using the finite element code Cast3M 2012[18], the mechanical behavior of the oxidized cladding was modeled for creep tests under internal pressure in an inert atmosphere. Therefore, no growth of the alpha(O) layer was considered during the whole simulation. A small portion (1° sector) of the cladding was meshed with quadratic brick elements. The number of elements was constant for the beta layer regardless of it thickness. On the other hand, the number of elements for the alpha(O) layer was adapted to its thickness. Boundary conditions were chosen according to the cylindrical symmetry of the problem (Figure 9). The mesh geometry was updated at each time step. A 1-second time step was used. It was checked that a smaller mesh size and a smaller time step did not affect the macroscopic response of the structure.

IV.C Simulation results

The simulated diametral strain as a function of time for a creep test simulation under 5 bar internal pressure and at 1100°C in inert atmosphere is shown in Figure 10 for various alpha(O) phase layer thicknesses. At 1100°C, replacing an outer layer of 5, 10 and 50 µm of beta phase by alpha(O) phase divide the initial strain rate in the cladding by a factor 1.7, 2.9 and 11 respectively. The thin layer of strong alpha(O) phase thus strongly enhances the resistance to viscoplastic hoop deformation under internal pressure. For the range of alpha(O) layer thicknesses expected from oxidation in steam at 1100°C during the creep test, the experimental strain rate of the cladding is still significantly lower than the ones predicted by these simulations. As expected, the predicted strengthening effect due to a 2 wt% alpha(O) layer is a lower bound of the strengthening due to oxidation in steam.

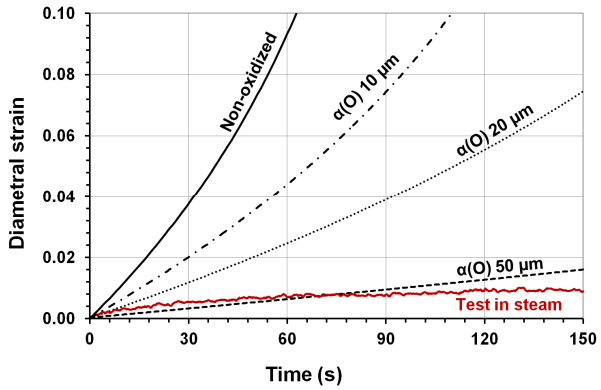

Fig. 10. Creep test in steam (red line) and creep simulations in inert atmosphere for various alpha(O) layer thicknesses (grey lines) – Zy-4 at 1100°C and 5 bar.

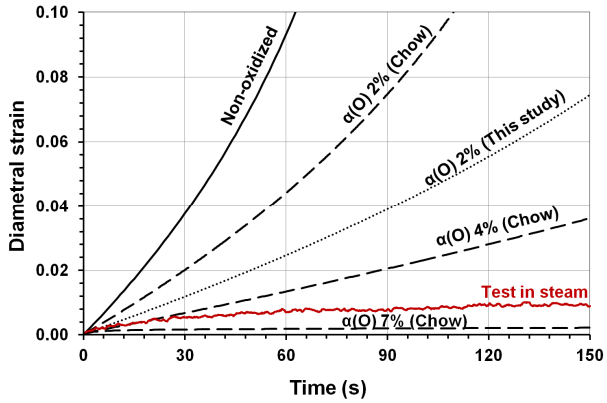

Fig. 11. Creep test in steam (red line) and creep simulations in an inert atmosphere for a 20-µm-thick alpha(O) layer with various oxygen contents (grey lines) – Zy-4 at 1100°C under 5 bar.

Extrapolating the law established by Chow *et al.*[9] for the highest oxygen content, i.e. 7 wt%, gives an another estimate closer to an upper bound of the effect of the alpha(O) layer (Figure 11). However extrapolating the same law[9] for 4 wt%, which is the mean oxygen content in the alpha(O) layer, leads to an insufficient prediction of reinforcement of the cladding. To improve this result, two assumptions can be made:

- On the one hand the increase in creep resistance of the alpha(O) phase for 4 wt% of oxygen can be underestimated by extrapolating the strengthening equation proposed by Chow *et al.*[9] based on experimental results until 1.5 wt%. The difference between our creep results on 2 wt% oxygen-enriched material and the prediction based on Eq. 2 supports this solution (Figure 6).
- On the other hand, if the behavior of the alpha(O) layer is correctly described, the mechanical contribution of zirconia has also to be taken into account in order to predict the total reinforcement of the cladding.

Creep tests on 4 wt% oxygen-enriched materials will help answering this question in a near future. Despite its limitations, the model presented here is thus a first, promising step toward improvement of prediction of the high temperature mechanical behavior of cladding tubes.

V. CONCLUSIONS

This study aimed at modeling the high temperature behavior of zirconium-based cladding in LOCA conditions. In order to model the oxidized cladding as a layered material, the constitutive behavior of the beta layer was taken from literature, so that the experimental part of the study focused on characterizing the alpha(O) layer, in particular under low stress levels.

Zirconium-based specimens enriched in oxygen by up to about 2 wt% were obtained and microstructurally characterized. Then, tensile axial creep tests were performed in a vacuum atmosphere. It was observed that this material exhibits high ductility in the temperature range from 900 to 1100°C, a 15% axial strain being reached under tension without fracture. In addition, oxygen-enriched specimens showed a high creep resistance, in agreement with the known mechanical strengthening effect of the oxygen. For the first time to the authors' knowledge, two creep regimes (creep exponents of ~1 and ~5, respectively, for lower and higer stresses) were observed, similarly to the two creep regimes already reported in the alpha phase of as-received zirconium alloys.

Homogenization calculations showed the remaining 15±5% fraction of beta phase in the oxygen enriched sample has a limited, but non-negligible impact (a factor two) on the viscoplastic strain rate.

As a consequence, the 2 wt% oxygen enriched power laws were used to model the alpha(O) behavior in FE computations of a two-layered tube.

These calculations with a 2 wt% oxygen enriched alpha(O) phase highlight a significant strengthening effect, up to a decade in hoop strain rate under internal pressure, which is a lower bound. On the other hand calculations based on extrapolated strengthening effect for 7 wt% of oxygen overestimate strengthening as expected.

Additional experimental tests are planned on 4 wt% and 7 wt% oxygen enriched samples – if technically feasible– to estimate the impact of the alpha(O) layer, and thus, of the oxide layer on the behavior of the layered cladding in LOCA conditions more precisely.

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