Behavior under LOCA conditions of Enhanced Accident Tolerant Chromium Coated Zircaloy-4 Claddings

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Abstract. For enhanced accident tolerant fuels for light water reactors application, chromium coatings on zirconium based nuclear fuel claddings are developed and studied at CEA in the framework of the French CEA-EDF-AREVA collaborative program. The results obtained so far, mainly on Zircaloy-4 substrate, show very good corrosion resistance in nominal conditions and significant enhancement of the resistance of the material to oxidation in steam at high temperature (HT), up to 1300°C, with a drastic decrease of hydrogen release and/or pick-up. The present paper reports some new results obtained on chromium coated Zircaloy-4 claddings tested in loss-of-coolant accident (LOCA) conditions. In order to investigate the potential effect of the coating on the cladding mechanical behavior at HT and the capacity of the coating to sustain significant substrate deformation (*i.e.*, during ballooning until burst occurrence) without generalized cracking/peeling, a preliminary limited set of internal pressure creep and temperature ramp tests have been performed in steam environment thanks to the EDGAR facility. The thermal-mechanical tests were done for testing/burst temperatures ranging from 600°C (α_{Zr} phase domain) up to 1000°C (β_{Zr} phase domain) on 50 cm long low-tin Zircaloy-4 cladding samples with a 15µm thick outer chromium coating.

Keywords: Enhanced Accident Tolerant Fuels (EATF), chromium coating, Zircaloy-4, LOCA, internal pressure burst test, high temperature creep

INTRODUCTION

Oxidation resistant coatings are an attractive and short term solution to enhance the resistance of current zirconium based nuclear fuel cladding to accelerated high temperature (HT) steam oxidation (and potentially hydriding) upon accidental scenario such as loss-of-coolant accident (LOCA). With the aim of achieving these objectives, CEA has initiated several years ago some prospective studies and preliminary evaluation of the performances of different types of coatings, including ceramic and metallic, mono- or multi-layered coatings. During these early studies, metallic chromium coatings have shown an encouraging behavior with a very good corrosion resistance in nominal conditions and significant enhancement of the HT steam oxidation behavior in accidental conditions, when compared to reference uncoated zirconium based cladding materials. More recently, the studies conducted in the framework of the French CEA-EDF-AREVA collaborative program have confirmed the good behavior of Cr coatings in steam environment at HT, up to at least 1300°C [1, 2].

However, the HT experiments performed so far did not consider the coating behavior and its potential effect on the whole thermal-mechanical behavior of the cladding upon HT transient, when the cladding tube is subjected to internal pressure and then experiences significant deformation/ballooning until burst occurrence. Some issues are thus still pending:

- Does cracking of the Cr-coating occur at a certain level of strain at HT?
- Is the good adherence of the coating maintained at HT leading to no delamination?
- Due to higher Young's modulus and strength when compared to zirconium based alloys, may a 15 µm thick chromium coatings influence the overall thermal-mechanical response of coated cladding tubes when tested in LOCA conditions?

In an attempt to address these pending questions a set of isothermal internal pressure creep and preliminary temperature ramp tests have been recently performed at CEA on Cr coated Zircaloy-4 cladding segments, in a steam environment, using the CEA "EDGAR" facility [3]. The main results obtained so far are briefly presented and discussed here after.

MATERIALS AND EXPERIMENTAL PROCEDURE

Materials

490mm long low-tin (SRA) Zircaloy-4 cladding segments from AREVA-NP have been used. 15µm thick chromium coating was deposited using a special physical vapor deposition (PVD) method. As-received dense coatings were thus obtained without apparent porosity or cracks at the Cr-Zr interface, and with good azimuthal/axial homogeneity of thickness, except at the vicinity of the ends of the cladding tube where the coating was sometimes thinner due to "ends effect" during the coating deposition process.

Thermal-mechanical LOCA testing

The "EDGAR" facility has been extensively used at CEA during the last twenty years on various modern nuclear fuel cladding materials, some of them being pre-hydrided as a surrogate to the in-service microstructural cladding evolution [3]-[5]. The facility enables to conduct various kinds of simultaneous and independent thermal and internal pressure transients on a 490mm long specimen of fuel cladding tubes, representative of the early stages of a LOCA transient. This facility has already been described in details in reference [3]. In order to obtain rapid heating rates, up to at least 100°C/s, a direct Joule effect heating is used in a flowing steam environment. In steady-state conditions, the axial and azimuthal temperature gradients are lower than 5°C over the 300-mm useful part (gauge length) of the specimen. Temperature of the clad is followed by dedicated thermocouples and pyrometers. The circumferential deformation of the tube is measured continuously until burst occurrence (non-contact measurements using a laser device). The tests are generally performed until the cladding bursts. Postmortem measurements are systematically performed at different axial locations, including at the burst opening, for the evaluation of local hoop strain and axial extent of the balloon.

ISOTHERMAL CREEP TESTS RESULTS

Before conducting more LOCA representative thermal ramp tests, a set of isothermal EDGAR creep tests has been done, covering the main burst temperature ranges of interest for DBA (design basis accident) LOCA conditions, *i.e.* between 600°C (α_{Zr} phase domain) and 1000°C (β_{Zr} phase domain).

Overall creep behavior

Figure 1 shows the evolution of the time to rupture (until burst) as a function of the initial hoop stress of both uncoated and 15µm thick Cr-coated Zircaloy-4 clad segments, for creep tests performed at different temperatures.

Additionally, Figure 2 shows two typical creep curves obtained on uncoated and 15µm thick Cr-coated Zircaloy-4 clad segments tested at 600°C for an internal pressure applied of 120 bar. From these two figures it is observed that, for a given internal pressure, typical creep rupture times of 15µm thick coated materials are two to three times higher than those of the uncoated material. This means that the 15µm thick chromium coating induces a strengthening effect at HT.

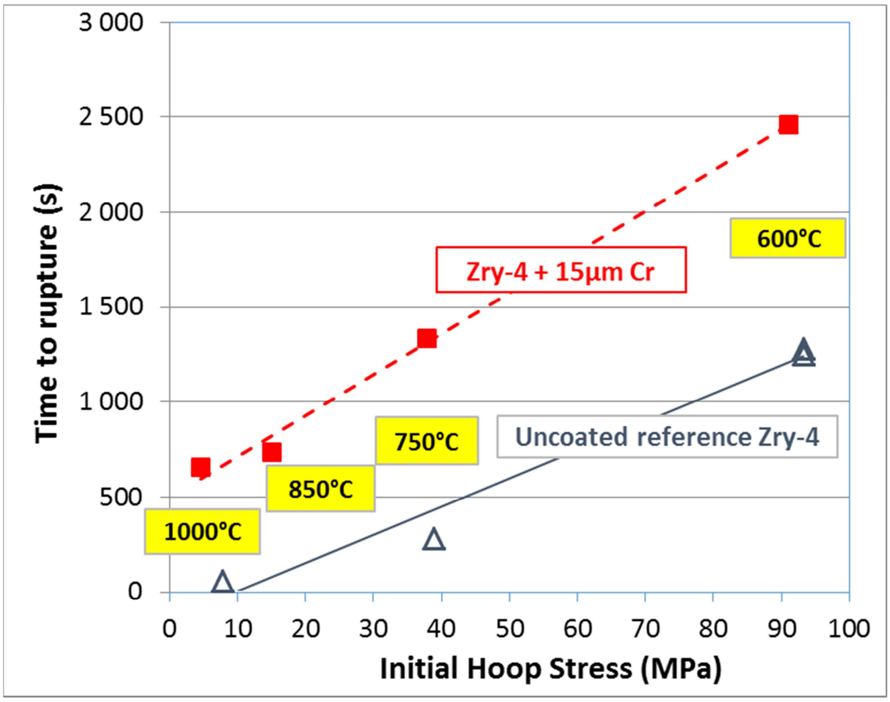


Figure 1 – Time to rupture as a function of the initial hoop stress of uncoated and 15µm thick Cr-coated Zry-4 clad segments for EDGAR (internal pressure) isothermal creep tests performed at different temperatures

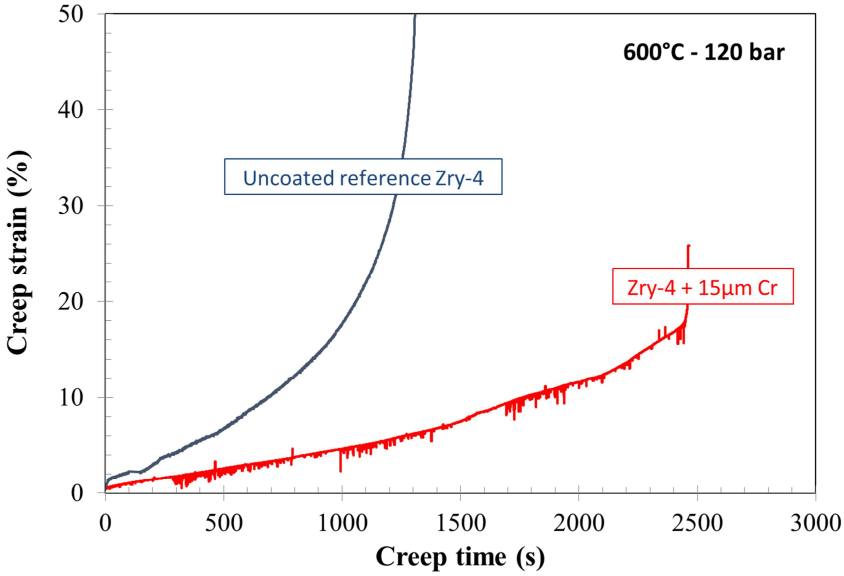


Figure 2 – Typical creep curves obtained on uncoated and 15µm thick Cr-coated Zry-4 clad segments at 600°C, for an applied internal pressure of 120 bar

Discussion

Given the rather thin thickness of the chromium coating studied here, such a strengthening effect could be regarded as astonishing. At that point, it can be mentioned that H. G. Kim et al. [6] have recently conducted some internal pressure thermal ramp tests on ~80µm thick chromium coated Zircaloy-4 cladding tubes prepared by 3D laser coating. From these results, the authors stated that *"…the cladding burst was observed in the non-coated region in the partially Cr-coated samples, it is believed that the burst resistance of the Cr-coated cladding was improved over the non-coated cladding. Of course, there is possibility to increase the burst strength in the Cr-coated area since the cladding thickness in the Cr-coated region was increased about 80 µm..."* To analyze more in depth Kim et al.'s data and to compare them with the present data obtained with a much thinner coating, let us recall first that the actual hoop stress ($\sigma_{\theta\theta}$, in MPa) is related to the difference between the internal and the external pressure (P, in MPa) and the actual external clad diameter (D) and thickness (e) according to $\sigma\theta\theta = P(D - e)/(2e)$ - **Equation 1**:

$$\sigma_{\theta\theta} = P(D - e)/(2e) \text{ - } \textbf{Equation 1}$$

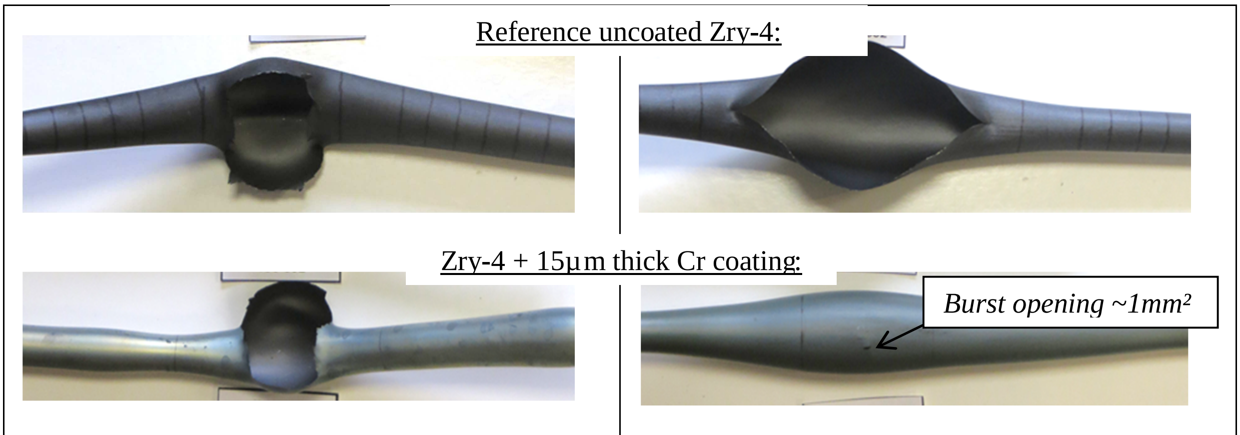
Then, at a given creep testing temperature, the actual creep rate is assumed to follow a Norton-type power law:

$$d\varepsilon_{\theta\theta}/dt = K\sigma_{\theta\theta}{}^{n} \text{ - } \textbf{Equation 2}$$

For the typical internal pressures applied here, the exponent (n) value must range from ~4 to ~6 ("dislocation creep" mechanism) [7]-[8]. Considering as an average a stress exponent value of 5 and by combining Equations 1 and 2, it is possible to evaluate, for any given applied internal pressure, the relative decrease of the creep rate due to the increase of the clad wall thickness only. From these simple calculations it can be observed that an increase of 80µm of the clad thickness may induce a relative decrease of ~50% of the actual creep rate while, for the present study, increasing by 15µm the clad thickness may only induce a 10% relative decrease of the actual creep rate. Thus, this evaluation shows that for the rather thin Cr-coated materials tested here, the increase in time to rupture and the associated decrease of the steady-state creep rate observed cannot be explained by the moderate wall clad thickness increase and thus might be due to an "intrinsic" strengthening effect of the chromium coating.

Post-testing balloon and failure appearances

Figure 3 shows some typical post-test visual aspect of uncoated and 15µm thick Cr-coated Zircaloy-4 clad segments that have experienced EDGAR creep tests until burst at 750°C and at 850°C.



(a) After creep tests at **750°C**	(b) After creep tests at **850°C**

Figure 3 - Burst appearance of uncoated and 15µm thick Cr-coated Zircaloy-4 clad segments that have experienced isothermal EDGAR creep tests at 750°C and at 850°C

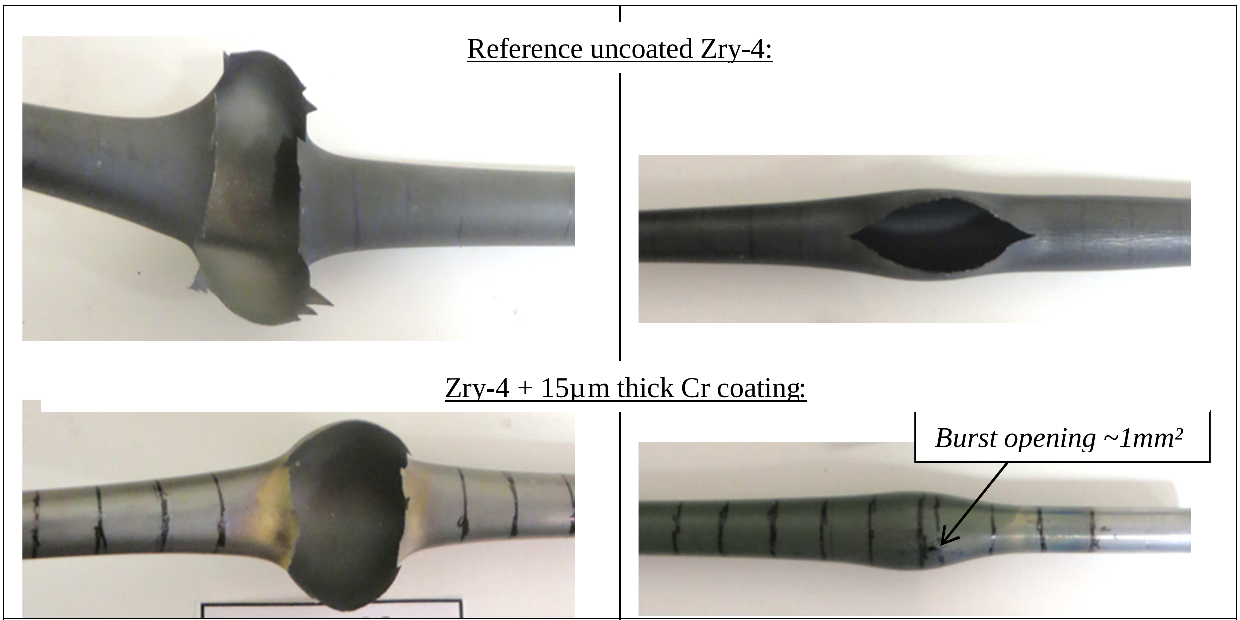
- *Remark: On Figure 3 and Figure 4, circular black pen marks are visible; they correspond to locations where diametric deformation of the tube at various elevations are performed after the test (i.e., post-mortem circumferential strain measurements).*

However, it can be observed that:

- whatever the applied temperature/pressure values, the chromium coating is still fully adherent after having experienced ballooning and burst, including at the vicinity of the burst where the Zircaloy-4 clad substrate is highly deformed;
- the post-test surface of the coated cladding shows mainly negligible oxidation (with a typical “metallic” aspect), while the uncoated reference materials, tested in the same conditions, show a black surface due to formation of a relatively thick outer zirconium oxide; this confirms the good resistance of Cr coatings to HT steam oxidation;
- for creep temperatures below ~800°C (i.e., full α_{Zr} temperature range), circumferential “uniform” and maximum strains of Cr-coated claddings are generally lower than those of uncoated reference materials;
- for creep temperatures above ~800°C (i.e., {α_{Zr}+ β_{Zr}} and full β_{Zr} temperature ranges), even if significant ballooning occurs before burst, the burst opening is very small, of the order or less than 1mm²; this apparent intrinsic Cr-coating effect on the burst opening may have positive consequences by mitigating the potential fuel relocation/dispersal as observed after semi-integral or integral tests performed on high burnup (BU) irradiated claddings (cf. integral LOCA “IFA” tests performed at Halden and semi-integral LOCA tests at Studsvik…).

PRELIMINARY THERMAL RAMP TESTS RESULTS

With the aim of being more representative of LOCA type transients, two preliminary temperature ramp tests under internal pressure have been performed at 1°C/s. For the higher internal pressure applied (100 bar), rupture occurs in the α_{Zr} temperature range, *i.e.* in the 700-800°C temperature range, while for the lower internal pressure applied (10 bar) burst occurs above 1000°C within the full β_{Zr} temperature range. Keeping in mind the limited number of temperature ramp tests performed so far on coated materials, temperature to rupture of coated cladding tubes seems slightly higher than those of the uncoated materials. Nevertheless, more ramp tests are required before drawing definitive conclusions. Figure 4 shows the uncoated/coated clad segments appearance after the internal pressure ramp tests. It can be observed that the preliminary ramp tests performed at 1°C/s seem to confirm the isothermal creep and burst behavior of Cr-coated claddings when compared to reference uncoated materials: *i.e.*, good coating adherence, negligible oxidation, smaller balloon when burst occurs in the α_{Zr} temperature range (high internal pressure applied) and very small burst opening after ballooning when burst occurs in the β_{Zr} temperature range (lower internal pressure applied).



(a) **1°C/s - 100 bar**	(b) **1°C/s - 10 bar**
⇨ ***Burst occurrence in the α_{Zr} temperature range***	⇨ ***Burst occurrence in the β_{Zr} temperature range***

Figure 4 - Clad segment appearance after internal pressure ramp tests at 1°C/s

CONCLUSIONS AND FURTHER WORK:

The present paper reports some new results obtained on chromium coated Zircaloy-4 nuclear fuel claddings tested in LOCA conditions showing some improvements of the material accidental behavior. In order to investigate the potential effect of the coating on the cladding mechanical behavior at HT and its capacity to sustain significant substrate deformation, a set of internal pressure creep and temperature ramp tests have been performed at HT in steam environment. The thermal-mechanical tests were done on 490 mm long (SRA) low-tin Zircaloy-4 cladding samples with a 15µm thick outer chromium coating and for testing/burst temperatures ranging from 600°C (α_{Zr} phase domain) up to at least 1000°C (β_{Zr} phase domain). It is shown that:

- whatever the applied temperature/pressure values, the chromium coating is still fully adherent after having experienced ballooning and burst, including at the vicinity of the burst opening where the Zircaloy-4clad substrate is highly deformed;
- a HT strengthening effect of the coating on the overall creep clad behavior is evidenced when compared to uncoated Zircaloy-4 cladding materials tested in the same conditions;
- as a consequence, it is observed that, in the 600-750°C temperature range (α_{Zr} phase domain) and after burst occurrence, the balloon sizes (*i.e.*, “uniform “and maximum hoop strains) are generally reduced when compared to uncoated materials;
- regarding the burst mechanism in the β_{Zr} phase temperature range (~1000°C), it is interesting to observe that, even if some ballooning occurred prior to the cladding failure, the actual burst openings are generally very small (in the order of 1 mm^2 or less), reducing the risk of fuel fragments dispersal in the coolant.

On-going and further work – A detailed study of the coating ductility as a function of both the local clad circumferential strain and the creep/burst temperature is on-going. More thermal ramp tests under internal pressure on coated materials, including M5™ substrate, are planned in the near future to be able to acquire a good knowledge and to model the thermal-mechanical behavior of chromium coated nuclear fuel cladding tubes subjected to different types of LOCA transients.

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