

**Behavior of chromium coated M5 claddings upon
thermal ramp tests under internal pressure
(loss-of-coolant accident conditions)**

M. Dumerval, Q. Houmaire, J.-C. Brachet, H. Palancher, J. Bischoff, E.
Pouillier

► **To cite this version:**

M. Dumerval, Q. Houmaire, J.-C. Brachet, H. Palancher, J. Bischoff, et al.. Behavior of chromium coated M5 claddings upon thermal ramp tests under internal pressure (loss-of-coolant accident conditions). Topfuel 2018 - Light Water Reactor (LWR) Fuel Performance Meeting 2018, Sep 2018, Prague, Czech Republic. cea-02328967

HAL Id: cea-02328967

<https://hal-cea.archives-ouvertes.fr/cea-02328967>

Submitted on 25 Feb 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

BEHAVIOR OF CHROMIUM COATED M5 CLADDINGS UPON THERMAL RAMP TESTS UNDER INTERNAL PRESSURE (LOSS-OF-COOLANT ACCIDENT CONDITIONS) (FOR POSTER SESSION)

M. Dumerval, Q. Houmaire, J.C. Brachet,
DEN, CEA, Université Paris-Saclay, Service de Rech. Métall. Appliquées
F-91191, Gif-sur-Yvette, France

H. Palancher
DEN, DEC, CEA, F-13108 Saint-Paul-lez-Durance, France

J. Bischoff
Framatome, 10 rue Juliette Récamier, 69456 Lyon, Cedex 06, France

E. Pouillier
EDF R&D, MMC Department, avenue des Renardières, 77818 Moret-Sur-Loing, France

Corresponding author: 33(0)169085616, jean-christophe.brachet@cea.fr

ABSTRACT

It has been previously shown that Cr-coatings, even with a thickness limited to 10-15 μ m, can have a positive impact on the high temperature isothermal creep and burst behavior of the cladding under internal pressure, typical of hypothetical Loss-of-Coolant Accident (LOCA) situations. Then, in view of being more representative of LOCA conditions, thermal ramp tests were recently performed at CEA thanks to the "EDGAR" facilities on 50 cm long M5 clad segments with 12-15 μ m thick Cr-coating. Heating rates ranging from 0.1 up to 25°C/s and internal pressures from 10 to 100 bars have been applied up to the rupture temperature. It was observed that, compared to the uncoated reference material, failure of the Cr-coated cladding segments occurred at comparable or higher temperatures with significantly smaller balloon and/or rupture opening. Very limited oxidation and excellent adhesion of the Cr-coating were also confirmed, even at the balloon location where the cladding was highly deformed.

M5 is a trademark or a registered trademark of Framatome or its affiliates in the USA or other countries.

1. INTRODUCTION AND EXPERIMENTAL PROCEDURE ("EDGAR" TESTS)

Chromium coated zirconium based fuel claddings are developed and studied within the CEA-Framatome-EDF joint research program as an "Enhanced Accident Tolerant Fuel" (EATF) cladding short-term concept for nuclear light water reactors. In addition to their already demonstrated improved resistance to High Temperature (HT) steam oxidation [1], it has been previously shown, for both Zircaloy-4 and M5 substrates, that Cr-coatings, even with a thickness limited to 10-15 μ m, can have a positive impact on the HT isothermal creep and burst behavior of the cladding under internal pressure [2]-[3]. The results obtained so far showed that in some conditions the Cr coating had a HT strengthening effect associated with significantly smaller clad ballooning and/or rupture opening. Such a behavior, if confirmed for more realistic LOCA dynamic thermal ramp conditions, should be of interest to mitigate nuclear fuel sub-assembly flow blockage, localized heat incursion issues and potential fuel relocation/dispersal (already observed after LOCA integral tests performed on high burn-up materials).

Then, in view of being more representative of LOCA anisothermal dynamic conditions, thermal ramp tests were recently performed at CEA on 50 cm long M5 clad segments with 12-15 μm thick Cr-coating, with heating rates ranging from 0.1 up to 25°C/s and internal pressures from 10 to 100 bars. The reference HT thermomechanical testing “EDGAR” CEA facilities [4] have been used. Their main characteristics are recalled here after.

The EDGAR-1 and EDGAR-2 facilities were built in 1974 and 1994 respectively at CEA and present similar characteristics. These devices are devoted to characterize the thermomechanical behavior of cladding tubes during the first step of LOCA transients. Since the eighties, more than two thousand EDGAR tests have been performed at CEA on several types of cladding materials, including pre-hydrided materials as a surrogate to high Burn-Up claddings [5]. The tests are performed on cladding tube measuring 490 mm in length and allow to apply many kinds of thermal and pressure transients, in steam environment. The specimens are heated by direct Joule effect (with a low-voltage alternative current) in order to obtain rapid increase of temperature i.e. up to 200°C/s, and internally pressurized by argon gas between 5 and 150 bars. This heating mode enables to obtain low clad thermal azimuthal gradients (less than 5°C) because it is well known that in LOCA conditions, azimuthal thermal gradients may strongly influence the circumferential elongations of the cladding up to rupture [6]. Thus, the primary objective of EDGAR testing was to be able to perform precise thermally controlled experiments to derive cladding (clad material dependent) HT creep constitutive laws and burst criteria, without parasitic effects. To preserve the biaxial stress state of the tube, the top grip of specimen is free of axial displacement and its weight is mechanically compensated. The temperature is measured by two methods:

- a K-type thermocouple centered inside of the clad,
- through quartz windows optical pyrometer measurements, operating up to 1300°C.

During the test, temperature is controlled most of the time by the pyrometer but could be back-controlled by the internal thermocouple. Additionally, some welded multi-thermocouples and, more recently, an infrared camera have been sometimes used for instrumented testing, to check the axial and/or azimuthal thermal gradients evolution upon some EDGAR test sequences.

Most of time, the experiments are made with a constant pressure measured by a differential pressure transducer. However, the EDGAR-1 facility allows increasing or decreasing the pressure over time in the range of -10 to +10 bars/s. The EDGAR-2 facility enables to increase or decrease the pressure as a function of the recorded deformation and/or temperature, or according to any given pressure versus time transient.

The cladding diameter is measured in the middle of the useful clad length during the test by a laser sheet device through the quartz window, to follow continuously the circumferential deformation of the tube and then to be able to plot and analyze the creep versus time resultant curves. The ballooning area is generally located just below the actual location of the laser clad diameter measurement. This is why, for most of the tests performed, the deformation measured continuously does not represent the maximum deformation (total elongation at rupture) but is more representative of a “uniform” clad deformation (before the deformation localization and plastic instability occurrence leading to the final rupture). All data are recorded in real time with an adjustable frequency between 1 and 10 Hz by a computer: i.e., temperature, pressure and clad circumferential deformation.

As illustrated in Figure 1, the typical post-mortem measurements are the:

- i) maximum circumferential deformation (A_t),
- ii) circumferential deformation at +20 mm and -20 mm of burst (A_r)¹,
- iii) average circumferential deformation ($\epsilon_{\theta\theta,h}$) in the homogeneous part of the clad.

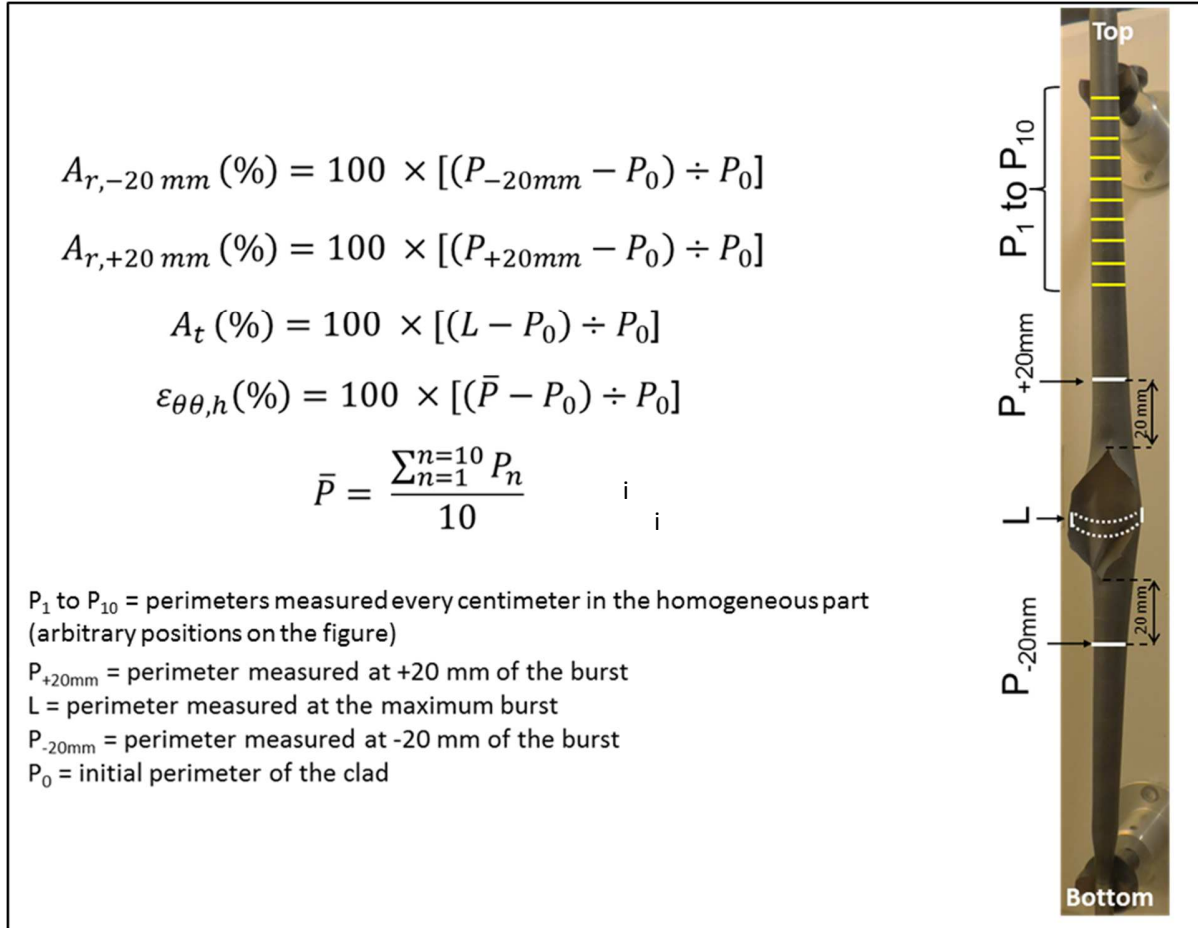


Fig 1. Postmortem measurements on an EDGAR tested clad segment of 490mm in length

2. BRIEF OVERVIEW OF THERMAL RAMP (EDGAR) TESTS RESULTS

In the following, only as-received coated and uncoated M5 materials are considered. From the different ramp tests performed on 12-15 μm thick Cr coated M5 clad segments, it can be observed that:

- Depending on the internal pressure applied and of the heating rate, the rupture time/temperature measured are in most cases higher than the uncoated reference material ones tested at similar ramp rates. As an illustration, Figure 2 shows an example where, for the particular heating rate and internal pressure conditions applied, the time to rupture of the Cr-coated-M5 is significantly higher than the simulated² uncoated one. For another EDGAR test performed under

¹ +/-20mm is an arbitrary axial location which has been chosen to be more or less representative of an average value between the circumferential elongation just at the burst opening ends and the one corresponding to the homogenous part of the overall (ballooned) cladding tube length.

² The reason why the comparison is done with a simulated curve and not with an experimental one for the uncoated reference materials is to be able to take accurately into account the actual thermal-pressure ramp applied on the Cr-

low internal pressure and more dynamic conditions, the ramp test was stopped when the Cr-coated clad reached 1133°C without burst occurrence and limited creep deformation due to a security temperature threshold of the EDGAR facility. In comparison, two tests already performed in similar conditions on uncoated reference M5 clad segments induced rupture temperatures ranging between 1030 and 1090°C (β_{Zr} temperature range) so the Cr-coating seems to increase the rupture temperature by at least $\sim 50^\circ\text{C}$ in these conditions. These particular tests seem to confirm the potential HT strengthening effect of the Cr coating on the actual creep properties of Cr-coated claddings, as already observed upon isothermal creep test, especially within the 600-850°C temperature range (α_{Zr} domain).

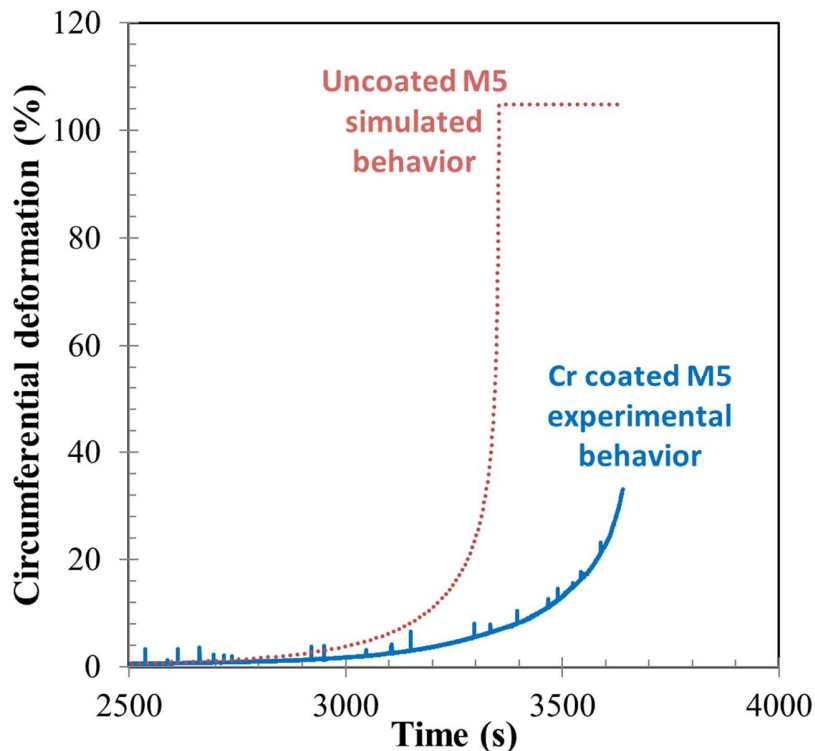


Fig 2. – Typical evolution of Cr-coated M5 clad circumferential elongation measured upon a low heating rate thermal ramp test for an intermediate internal pressure applied; comparison with the simulated uncoated M5 reference cladding behavior

- In Figures 3 to 5, the post-mortem circumferential deformation parameters measured on Cr-coated M5 are compared to the uncoated reference M5 ones tested in the same conditions. It must be mentioned that for a few cases, the data obtained on one coated clad segment is compared to two data points relying to uncoated reference M5 (already tested in the same conditions), thus increasing slightly the apparent number of data points. Additionally, some typical clad balloon and burst opening aspects are illustrated in Figure 3. It is obvious from these figures that the balloon sizes and circumferential elongations of the Cr-coated M5 are significantly lower than the uncoated M5 ones, thus confirming the tendency already observed upon isothermal creep tests [2]-[3]. It can be also observed that, for heating rates higher than or equal to 1°C/s (which are quite prototypical of most of LOCA transients), the post-mortem clad “uniform” circumferential

coated clad segment which may slightly differ from the previous ramp tests performed on uncoated reference materials

elongations³ are systematically lower than ~30%. While the maximum (total) elongation measured at burst location is lower than 70%. Taking into account that there is almost no significant azimuthal thermal gradient produced upon the EDGAR tests, these values can be considered as an upper (conservative) limit for the clad behavior upon LOCA, as compared to the LOCA cladding behavior in a more representative nuclear fuel sub-assembly environment (bundle effects)⁴.

As already observed on both Cr-coated Zircaloy-4 and M5 after isothermal creep testing up to rupture at different temperatures [2,3], and whatever the applied heating rate/ internal pressure values:

- the chromium coating is still fully adherent with no coating spallation, after having experienced ballooning and burst, including at the vicinity of the burst where the M5 clad substrate is highly deformed. This would thus preserve the residual Cr-coating resistance capacity upon subsequent HT steam oxidation (until the final core reflooding by the Emergency Cooling Systems);
- 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 outer zirconium oxide; this confirms the good resistance of Cr coatings to HT steam oxidation (see macrographs on Figure 3);
- when the rupture occurs at temperatures higher than ~850°C (i.e., corresponding to $\{\alpha_{Zr} + \beta_{Zr}\}$ or fully β_{Zr} temperature ranges), burst opening is generally very small, of the order or less than 1mm². This apparent intrinsic Cr-coating effect on reduction of the burst opening may have positive consequences by mitigating the potential fuel relocation and dispersal as sometimes observed after semi-integral or integral tests performed on high burnup (BU) irradiated fuel rods. Additionally, it is believed that such a small burst opening may significantly limit the steam access to the inner surface of the clad, thus delaying the potential clad embrittlement risk due to secondary hydriding phenomena.

³ Values measured at +/-20 mm from the burst opening and averaging values for the homogenous part of the cladding, which correspond to Figures 4 and 5 respectively

⁴ This last situation inducing likely some azimuthal thermal gradients due to the thermal influence of the neighboring nuclear fuel rods, leading to a decrease of the cladding total elongation at rupture, as discussed previously

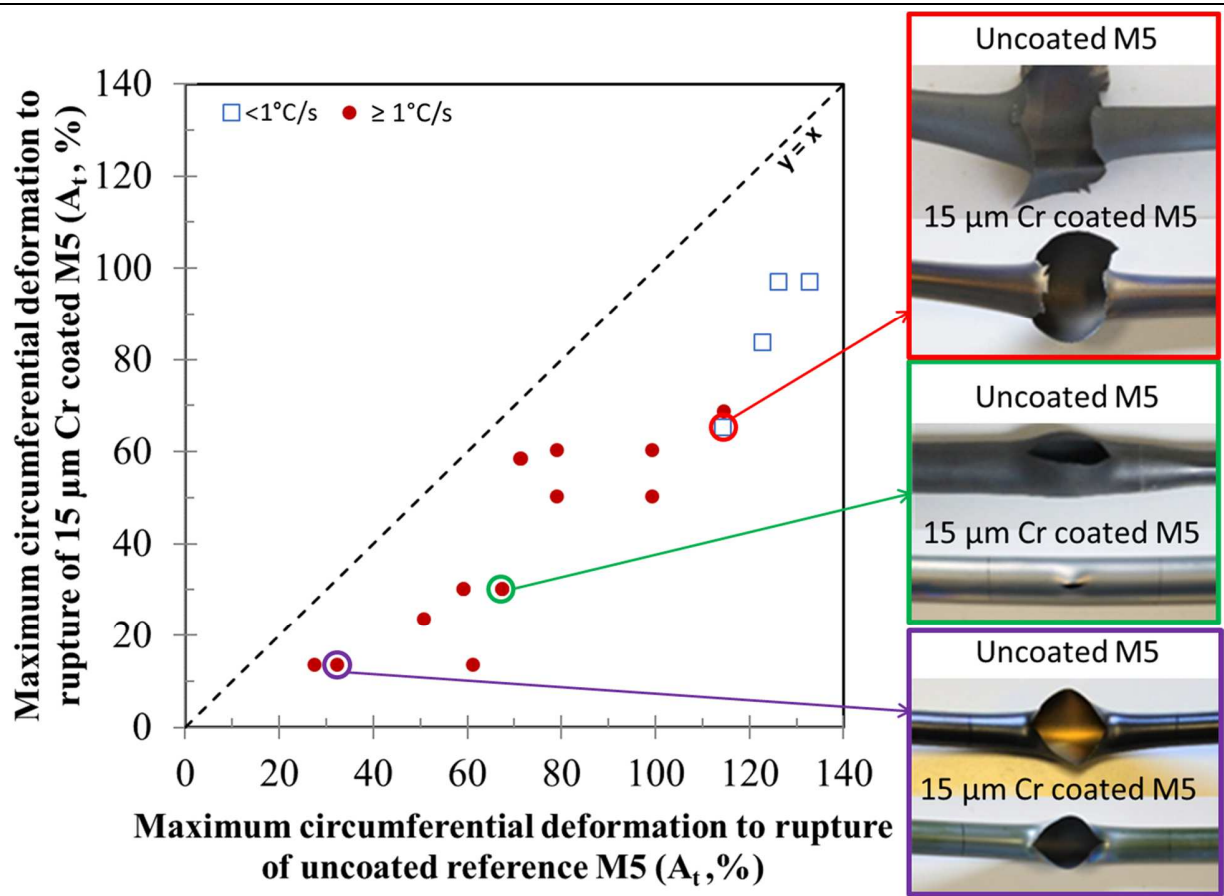


Fig 3. Maximum circumferential deformations (A_t) for Cr coated M5 compared to those measured for uncoated references, tested in the same conditions (Temperature range: 0.1°C/s to 25°C/s , Pressure range: 10 to 100 bars)

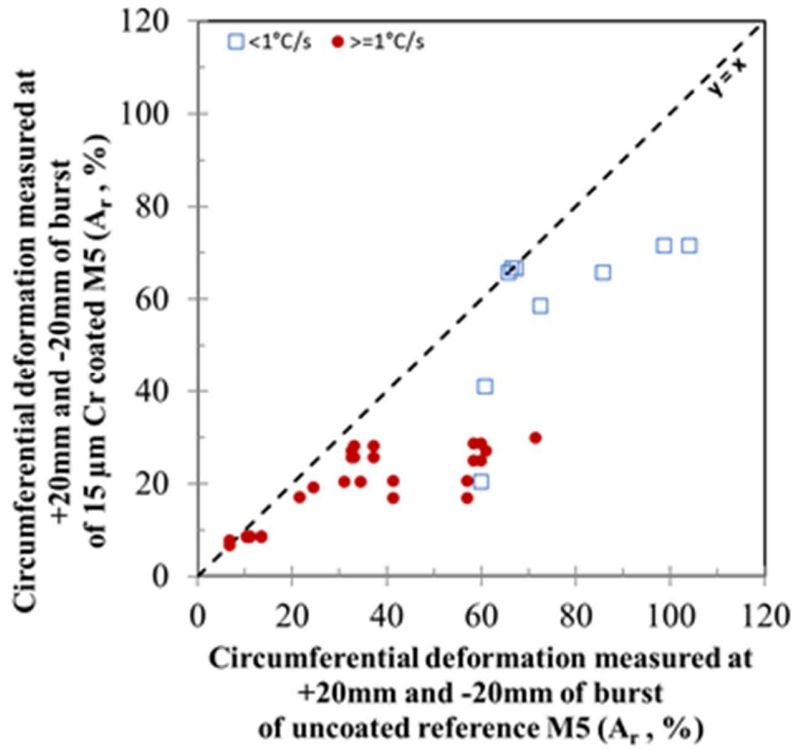


Fig 4. Circumferential deformations measured at +/- 20 mm from the burst (A_r), for Cr-coated M5 compared to those measured for uncoated M5 ones, tested in the same conditions

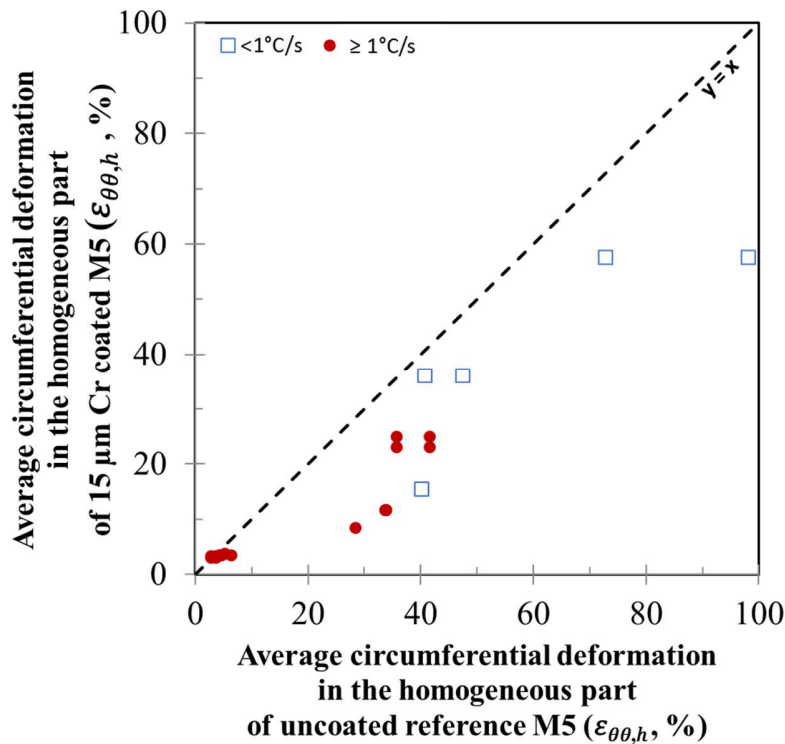


Fig 5. Average circumferential deformation in the homogeneous cladding part ($\epsilon_{\theta\theta,h}$), for Cr-coated M5 compared to those measured for uncoated M5 ones, tested in the same conditions

3. CONCLUSIONS

Chromium coated zirconium based fuel claddings are developed and studied within the CEA-Framatome-EDF joint research program as an “Enhanced Accident Tolerant Fuel” (EATF) cladding short-term concept for nuclear light water reactors. In addition to their already demonstrated improved resistance to High Temperature (HT) steam oxidation, the present study focused on their thermomechanical behavior upon thermal ramps under internal pressure, typical of LOCA transients. When compared to uncoated reference M5 claddings tested in the same conditions, the “EDGAR” thermal ramp tests performed on 10-15 μm Cr-coated M5 confirm potential LOCA behavior improvements, preliminarily observed upon isothermal creep tests:

- comparable or higher rupture temperatures (HT Cr-coating strengthening effect);
- significantly lower balloon sizes and, in some conditions, extremely small burst opening sizes;
- negligible HT steam oxidation and no coating spallation, even at the vicinity of the burst where the clad substrate is highly deformed, thus preserving the Cr-coating resistance capacity to subsequent HT steam oxidation.

On-going and further work – Additional LOCA ramp tests are on-going to continue to build a larger database and improve the statistics in the LOCA creep and burst behavior, to potentially derive more precise behavior models if necessary. In the future, it would be also interesting to extend internal pressure tests at HT on irradiated materials to confirm the LOCA behavior improvement observed on as-received coated materials, after achieving significant Burn-Up.

4. REFERENCES

1. J.C. Brachet et al., "Behavior of Cr-coated M5TM claddings during and after high temperature steam oxidation from 800°C up to 1500°C (Loss-of-Coolant Accident & Design Extension Conditions)", this conference
2. J.C. Brachet et al., "Behavior under LOCA conditions of Enhanced Accident Tolerant Cr-Coated Zircaloy-4 Claddings", proceedings of TopFuel 2016, Boise, Idaho, USA (Sept. 11-16, 2016)
3. J.C. Brachet et al., « Behavior of Cr-coated M5TM claddings under LOCA conditions », Proceedings of 2017 Water Reactor Fuel Performance Meeting, (Sept. 10-14, 2017), Jeju Island, Korea
4. T. Forgeron, et al., "Experiment and modelling of advanced fuel rod behavior under LOCA conditions : $\alpha \leftrightarrow \beta$ phase transformation kinetics and EDGAR methodology", in proceedings of the 12th International Symposium on Zirconium in the Nuclear Industry, ASTM STP 1354, (2000), pp. 256-278
5. L. Portier, et al. "Influence of Long Service Exposures on the Thermal-Mechanical Behavior of Zy-4 and M5 Alloys in LOCA Conditions," J. ASTM Int., Vol. 2, No. 2, (2005), JAI12468
6. F.J. Erbacher, "LWR fuel cladding deformation in a LOCA and its interaction with the emergency core cooling", ANS/ENS Topical Meeting on Reactor Safety Aspects of Fuel Behaviour, (1981), Sun Valley, USA