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Effect of chromium grain size and morphology on the HT oxidation behavior of chromium coated Zr based alloys

Nihed Chaabane-Jebali, Jean-Christophe Brachet, Matthieu Le Saux, Didier Hamon, Elodie Rouesne, Stéphane Urvoy, Thomas Guilbert, Michel Schlegel, Michel Tabarant, Fernando Lomello

► **To cite this version:**

Nihed Chaabane-Jebali, Jean-Christophe Brachet, Matthieu Le Saux, Didier Hamon, Elodie Rouesne, et al.. Effect of chromium grain size and morphology on the HT oxidation behavior of chromium coated Zr based alloys. Materials Science and Engineering - session Structural Materials and Metallurgy, Feb 2019, San Francisco, United States. cea-02394049

HAL Id: cea-02394049

<https://cea.hal.science/cea-02394049v1>

Submitted on 21 Feb 2020

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Nihed Chaâbane^a, Jean-Christophe Brachet^b, Matthieu Le Saux^b, Didier Hamon^b, Elodie Rouesne^b, Stéphane Urvoy^b, Thomas Guilbert^b, Michel Schlegel^c, Michel Tabarant^c, Fernando Lomello^c

^a INSTN, CEA, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France

^b DEN, CEA, Université Paris-Saclay, Service de Rech. Métall. Appliquées, F-91191, Gif-sur-Yvette, France

^c DEN, CEA, Université Paris-Saclay, Service d'Etudes Analytiques et de Réactivité des Surfaces, F-91191, Gif-sur-Yvette, France

Context: Chromium coated zirconium based nuclear fuel claddings are developed and studied within the CEA-Framatome-EDF joint research program as "Enhanced Accident Tolerant Fuel" (EATF) cladding short-term concept for nuclear light water reactors [1] [2].

Objective: To investigate the influence of the initial chromium grain size and morphology on the High Temperature (HT) steam oxidation behavior of 6-8µm thick Cr-coated zirconium based specimens, by coupling several *post-mortem* experimental techniques, that is, FEG-SEM-EBSD, Glow discharge Mass Spectrometry (GDMS), Electron Probe Micro Analysis (EPMA) and Raman spectroscopy.

Materials and experimental conditions:

- ✓ Zr based alloys: Sheets specimens (thickness ~1.2 mm)
Tubular cladding geometry (thickness ~0.6 mm)
- ✓ 6-8 µm thick chromium coatings deposited by a special Physical Vapor Deposition (PVD) process:
=> very good bonding with the Zr substrate - dense coating without apparent porosity - good thickness homogeneity

Experimental conditions:

- 1- Pre-annealing thermal treatments – 2h at 700 and 800°C, under vacuum
- 2- HT steam oxidation at 1200°C in steam and final water quenching (using reference CEA "DEZIROX" facility [3])

Post-mortem measurements:

- Weight gains and Post-Quenching (PQ) examination/analysis: FEG-SEM-EBSD, GDMS, EPMA and Raman spectroscopy

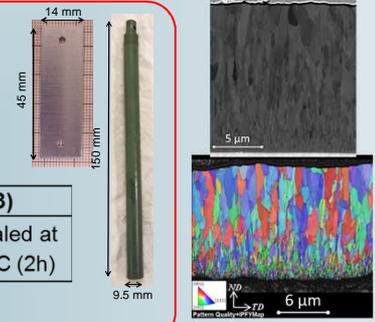
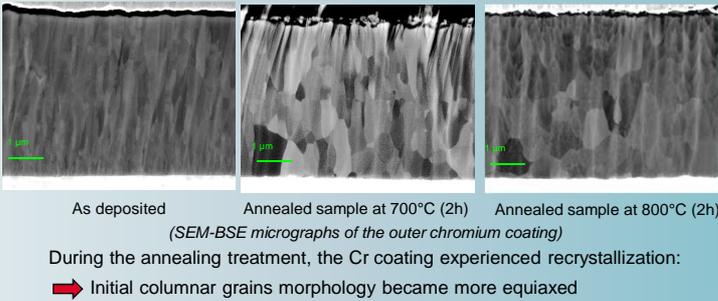


Fig. 1 : Typical microstructures of Cr coating on Zr substrate (a- FEG-SEM micrograph; b- EBSD map)

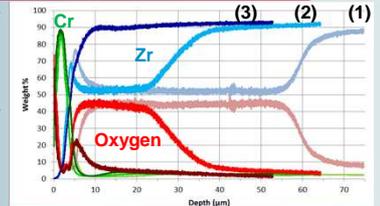
Samples	(1) Virgin	(2) Annealed at 700 °C (2h)	(3) Annealed at 800 °C (2h)
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Pre-annealing thermal treatment

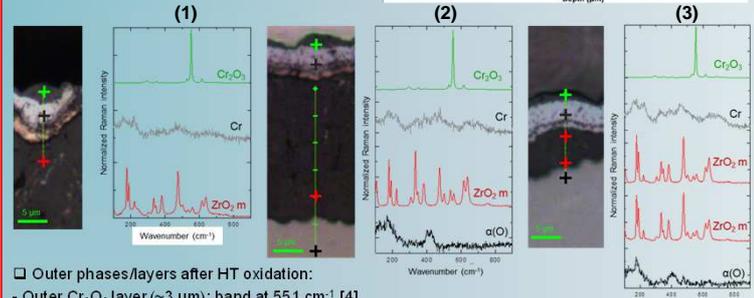


PQ microstructures and oxygen diffusion

- ➔ Glow discharge mass spectrometry measurements
- ✓ Interdiffusion between Cr and Zr
- ✓ Decrease of oxygen diffusion into the Zr substrate for pre-annealed samples



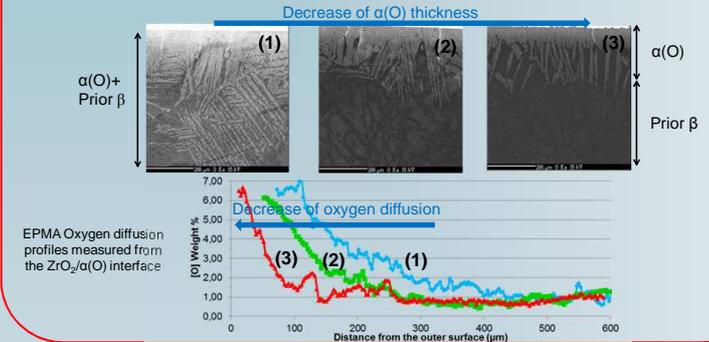
Cr₂O₃, Cr and ZrO₂ layers:



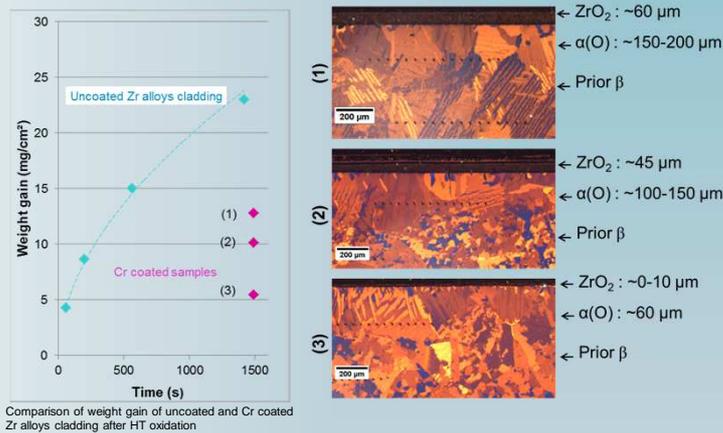
Raman spectroscopy measurements

- Outer phases/layers after HT oxidation:
 - Outer Cr₂O₃ layer (~3 µm): band at 551 cm⁻¹ [4]
 - Residual (non oxidized) metallic Cr
 - Monoclinic Zirconia (prior-quadratic phase retransformed to monoclinic one upon final quenching)

Metallic Zr substrate, α(O) and Prior-β:



HT oxidation and PQ microstructures



Conclusions and on-going/further work

- Thermal pre-annealing treatment results in an increase of Cr size grains before the HT oxidation
- Weight gains of pre-annealed Cr-coated specimen were lower than the non-annealed ones.
- Thickness of ZrO₂ and αZr(O) phases were significantly reduced for the pre-annealed samples, indicating that the grain size and morphological evolutions of the chromium coating upon the pre-annealing thermal treatment may influence its further HT oxidation behavior, thus inducing:
 - Overall, higher resistance to HT oxydation
 - Significant delay in the oxygen ingress into the Zr based substrate
- On-going: complementary EBSD, TEM... analysis for more detailed informations on the associated oxygen diffusion mechanisms...

Acknowledgments: Framatome and EDF for financial support and fruitful discussions

Some references:

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2. J. Bischoff, et al., "AREVA NP's Enhanced Accident Tolerant Fuel Developments: Focus on Cr-coated M5 Cladding", Nuclear Engineering and Technology 50, (2018), 223-2283
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