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

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HAL Authorization

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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

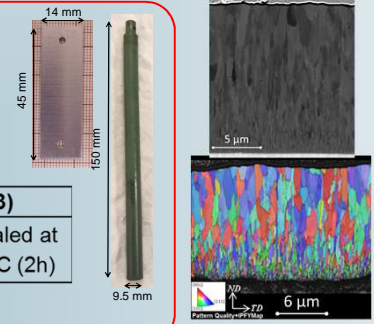
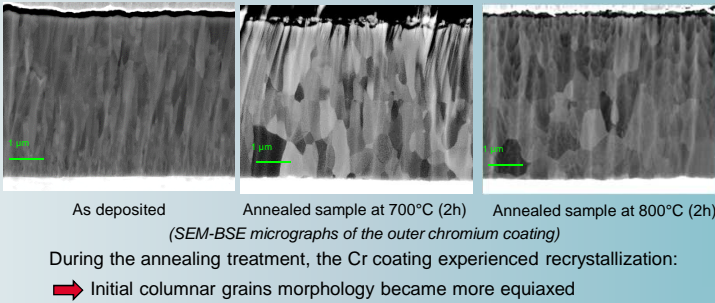


Fig. 1: Typical microstructures of Cr coating on Zr substrate (a- FEG-SEM micrograph; b- EBSD map)

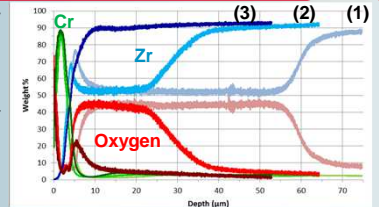
Samples	(1) Virgin	(2) Annealed at 700 °C (2h)	(3) Annealed at 800 °C (2h)
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Pre-annealing thermal treatment

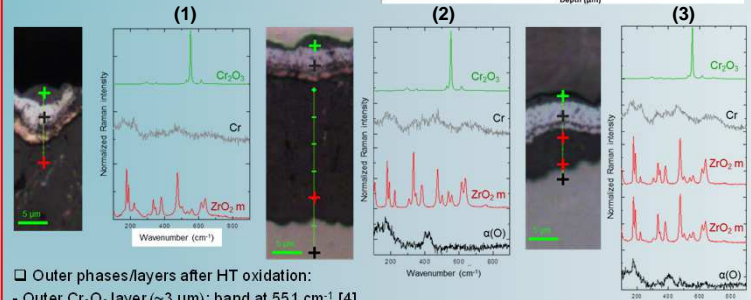


PQ microstructures and oxygen diffusion

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

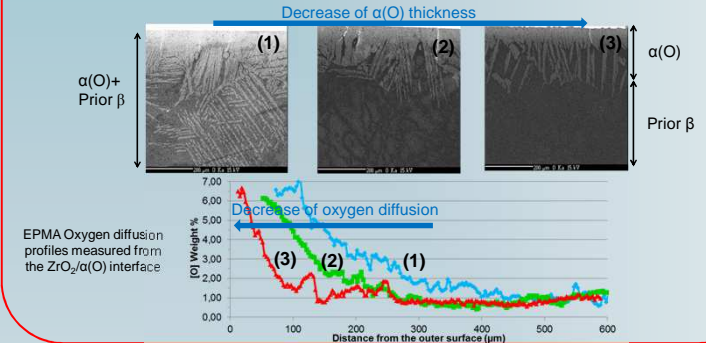


Cr₂O₃, Cr and ZrO₂ layers:

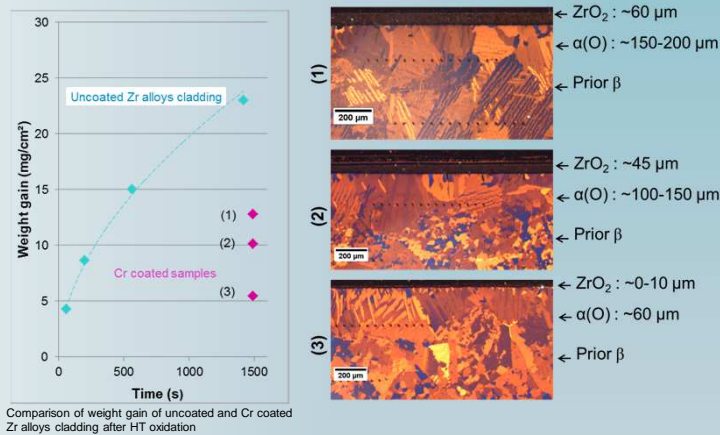


- Outer phases/layers after HT oxidation:
 - Outer Cr₂O₃ layer (~3 µm): band at 551 cm⁻¹ [4]
 - Residual (non oxidized) metallic Cr
 - Monoclinic Zirconia (prior-quadratic phase retransformed to monoclinic one upon final quenching)

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...

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Some references:

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4. I. Idarraga, et al., "Raman investigation of pre- and post-breakaway oxide scales formed on Zircaloy-4 and M5R in air at high temperature", Journal of Nuclear Mat. (2012) 421 – 160-171