Behaviour of the cladding oxide layer under steam starvation conditions

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Abstract

Results of the tests on the long duration annealing of pre-oxidized Zircaloy cladding tubes in the inert atmosphere with the annealing temperature 1400 °C are presented. The objective of the tests was the investigation of the reduction kinetic of the oxide layer during the steam starvation phase by the progression of core drying during the severe accident. The homogeneous formation of the α-Zr(O) precipitations inside of the oxide layer was detected in addition to the oxide layer thickness decrease. The phenomenon should have a strong influence on the intensive hydrogen release during the following quench phase.

1. Motivation and Objectives

The steam starvation conditions on the fuel rods surface are possible due to the drying of the reactor core and blockages formation during the severe accident. The oxide layer on the cladding tube surface will be reduced under these conditions. Therefore the knowledge about the oxide layer reduction processes is important for the severe accident measures. The detailed investigations there are up to now only for the description of the oxide layer *growth* during the cladding oxidation [1]. The oxide layer reduction processes were not investigated. The presented work describes the first results of the FZK corresponding experiments, which detect the complicate character of the oxide layer degradation process.

2. Applied Tools

Fifteen empty Zry-4 sealed tubes with a length of 15 cm were used. The experimental facility used for the pre-oxidation and for the following annealing of these probes was the IMF Quench rig [2]. The probes were pre-oxidised in argon (90 l/h) – steam (100 g/h) mixture flux at 1400 °C to reach a different initial oxide layer thickness (100 – 400 µm). Next stage was the annealing of pre-oxidised probe at 1400 °C during 300 – 10800 s. Three other probes were cool-downed after pre-oxidation and used as reference probes.

3 Conclusions and Future Work

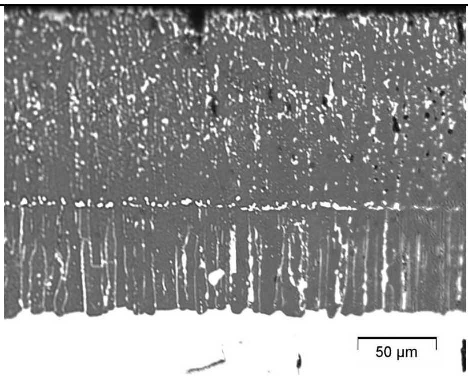
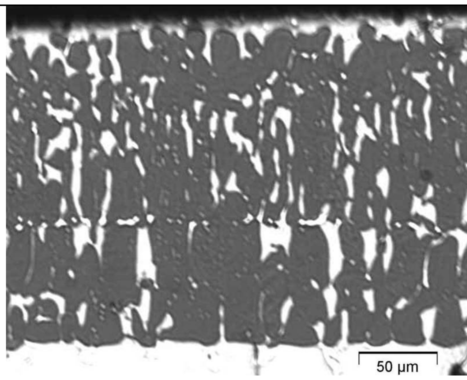
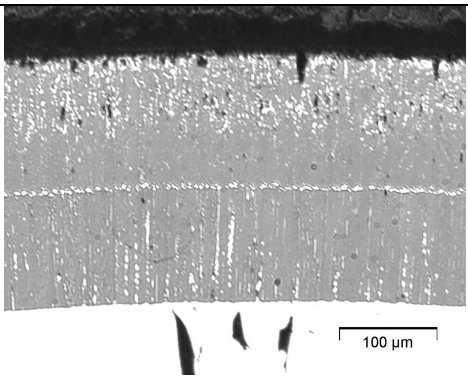
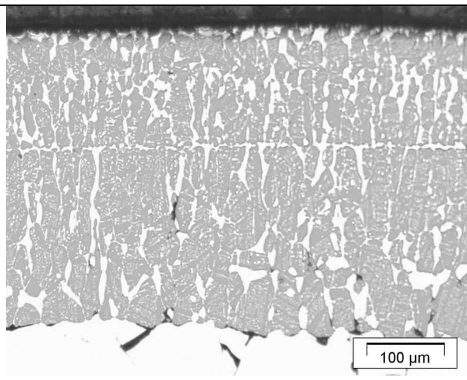
- The change of the oxide layer thickness during the reduction deviates dramatically from the results of the conventional oxidation model.
- The experimental results show the formation of the two-phase structure in the oxide layer during the reduction process. The metallic precipitates α-Zr(O) were formed inside of the oxide layer. Thin α-Zr(O) layer developed on the surface of oxide layer.
- The content of the metallic phase in the oxide layer can reach 25%, that considerably change the mechanical and diffusion properties of this layer.

- The formation of the α-Zr(O) precipitates in the oxide layer will lead to the intensification of oxidation under reflooding conditions.
- The detected phenomenon of the metal precipitations formation inside of the oxide layer combined with the decrease of the oxide layer thickness during the steam starvation phase causes the temperature escalation due to exothermic zirconium-steam reaction and the intensive hydrogen release during the quench process. The importance of the discovered phenomenon concerning the influence on the quench processes requires a detail theoretical activity.

4. Relevant Publications

[1] P. Hofmann, V. Noack, M.S. Veshchunov, A.V. Berdyshev, A.V. Boldyrev, L.V. Matweev, A.V. Palagin, V.E. Shestak, Physico-chemical behavior of Zircaloy fuel rod cladding tubes during LWR severe accident reflood. Wissenschaftliche Berichte, FZKA-5846 (Mai 97)

[2] J. Stuckert; M. Steinbrück; U. Stegmaier; A. Palagin. On the thermo-physical properties of Zircaloy-4 and ZrO_2 at high temperatures. Wissenschaftliche Berichte, FZKA-6739 (Oktober 2002)

pre-oxidation time 660 s initial δox ~210 µm		
	δ_{ox}=191 µm, metal precip.=21%	δ_{ox}=215 µm, metal precip.=19%
pre-oxidation time 1080 s initial δox ~320 µm		
	δ_{ox}=268 µm, metal precip.=13%	δ_{ox}=407 µm, metal precip.=19%
	annealing time 1200 s	**annealing time 3600 s**

Oxide layer structure.
Change of precipitates morphology vs. annealing duration