Degradation 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.

Zusammenfassung

Es werden die Ergebnisse von Dauerglühversuche mit voroxidierten Zircaloy Hüllrohren in der Inertgasatmosphäre mit der Glühtemperatur von 1400 °C dargestellt. Das Ziel der Tests war die Untersuchung der Kinetik der Oxidschichtreduzierung während der Dampfmangelphase bei der Entwicklung der Reaktorkerntrocknung während des schweren Störfalles. Neben dem Abnahme der Oxidschichtstärke wurde die homogene Entwicklung der α-Zr(O) Ausscheidungen innerhalb der Oxidschicht nachgewiesen. Das Phänomen sollte einen starken Einfluss auf die intensive Wasserstofffreisetzung während der folgenden Abschreckphase haben.

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. Status of Work

2.1 Applied Tools

Six empty Zry-4 sealed tubes with a length of 15 cm were used. The experimental facility used for the pre-oxidation of these probes and for the following annealing of the each second probe was the IMF Quench rig [2]. Three probes were pre-oxidised in argon (90 l/h) – steam (100 g/h) mixture flux at 1400 °C during 250 s, 750 s and 2000 s. These probes were cool-downed after pre-oxidation and used as reference probes. The other three probes were annealed at 1400 °C during three hours after the pre-oxidation periods with the same durations, corresponding to the reference probes.

2.2 Main Results Achieved in 2002

Fig. 1 shows the post-test appearance of the probes. All reference probes have a dark surface, whereas the surface of the annealed probes depends on the initial oxide layer thickness before the annealing. The metallic appearance of the probe with the thin initial oxide layer evidences the complete reduction of the oxide layer with dissolution of the oxygen in the metal. The grey surfaces of the other two annealed probes show not complete disappearance of the oxide structures.

The metallographic analysis of the middle cross-sections (**Fig. 2**) evidences the complete disappearance of the thin oxide layer (initial thickness ~130 µm) and formation of the homogeneous α-Zr(O) layer. The thicker oxide layers (initial thickness >200 µm) have been decreased with the time, but the degree of the decrease is relative small due to development of *metallic precipitations inside* of the oxide layer. The formation of these precipitations has a stochastic character and they distribute homogeneously across the oxide layer. The image analysis of the oxide layer region shows that the part of the metallic phase in the annealed oxide layer can achieve up to 25%. Such volume of the metallic phase near from the cladding tube surface will intensive interact with a steam during quench phase, following after the possible steam starvation phase during the severe accident measures.

2.3 Conclusions and Future Work

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 experimental and 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)

Fig. 1. Appearance of the pre-oxidised and annealed LWR cladding tubes. Annealing temperature 1400 °C, annealing duration 3 h

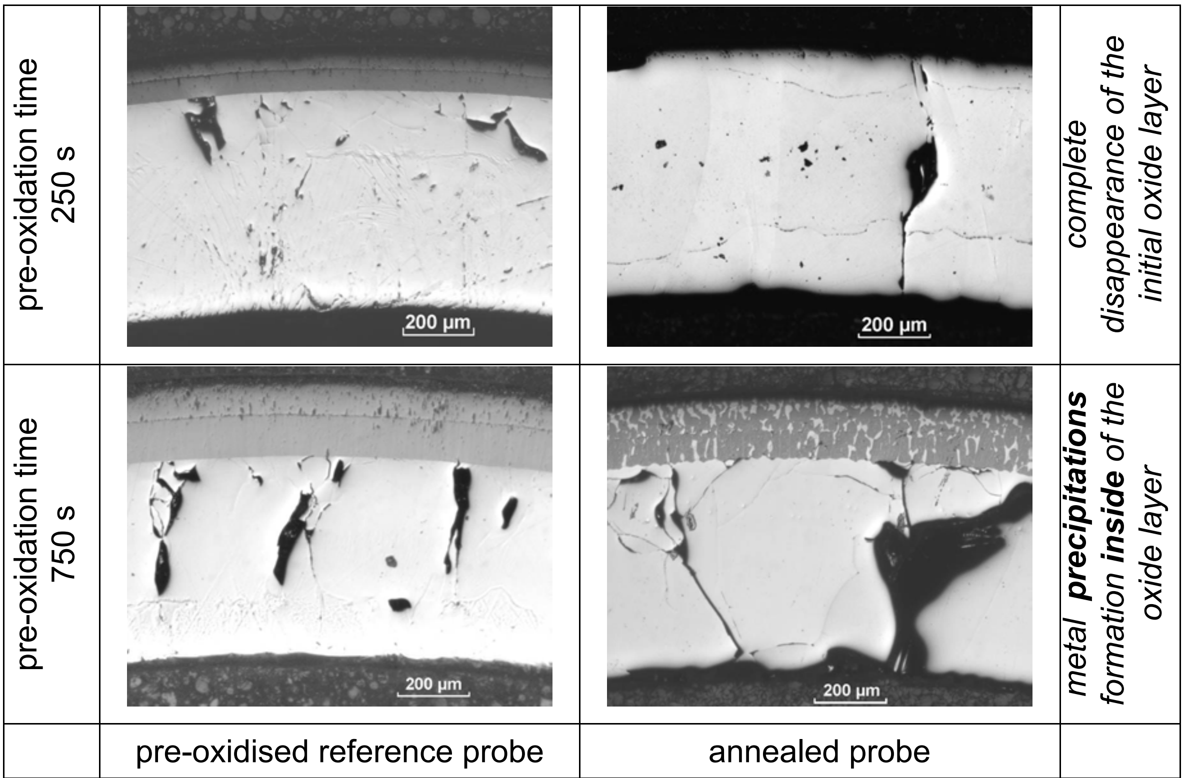

Fig. 2. Appearance of the cladding cross-section before and after annealing for thin (~130 µm) and thick (>200 µm) initial oxide layer