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Changes in the material structure of nuclear power  
plant fuel elements under accident conditions

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# 1 Background of the research

Nuclear energy is one of the key pillars of global electricity generation, providing large amounts of continuous power with low carbon-dioxide emissions. The application of nuclear technology, however, entails stringent safety requirements, since the potential consequences of operational disturbances and accidents at nuclear power plants may be extremely severe. From the safety point of view, fuel cladding plays a key role: its primary function is to retain fission products and to prevent direct contact between the fuel and the coolant.

In light water reactors, zirconium alloys have become widely used as cladding materials owing to their unique nuclear and materials-science properties. Their low neutron-absorption cross section, good corrosion resistance, and high melting point contribute to the efficient and safe operation of reactors [Northwood, 1985; Ivanov et al., 1958; Olander et al., 2017] a,b,c. Under accident conditions, however - for example during loss-of-coolant accidents (LOCA), reactivity-initiated accidents (RIA), or station blackouts (SBO) - the cladding is exposed to extreme mechanical, thermal and chemical loads. Oxidation, nitride formation, hydrogen uptake and microstructural changes may together lead to loss of cladding integrity and severe damage to fuel structure, thereby endangering plant and public safety. The experience of the Fukushima accident clearly demonstrated that the accident behavior of cladding materials is a determining factor in the development of severe consequences. These lessons motivated the launch of new international research programs aimed at the development of so-called accident tolerant fuels (ATF) [Terrani, 2018; Tang et al., 2017; Brachet et al., 2020; Steinbrück et al., 2020] d,e,f,g. Compared with conventional zirconium alloys, ATF claddings are intended to provide higher resistance under extreme conditions and thereby increase the safety margins of nuclear power plants. The aim of the dissertation is the detailed investigation of changes in the material structure of zirconium-based claddings under accident conditions. The research focuses on the comparative analysis of claddings with different compositions, production technologies, and wall thicknesses. The experiments were carried out at the CODEX facility operated at the HUN-REN Centre for Energy Research, which makes it possible to perform integral laboratory experiments modeling the severe accident processes in nuclear reactors.

## 2 Objectives

The main objective of the present research was the comprehensive and systematic investigation of changes in the material structure of zirconium alloys - used as nuclear fuel claddings - under accident conditions. The extreme temperature, pressure and chemical effects arising during operational disturbances and severe accidents strongly influence the mechanical and chemical stability of the fuel cladding. Understanding these processes is of outstanding importance from the point of view of nuclear safety. The research aimed to reveal the behavior of cladding materials with different compositions, production technologies and wall thicknesses (E110, E110G, E110G SLIM, OptZirlo<sup>TM</sup>, as well as chromium-coated ATF variants) under beyond design basis accident conditions, and to provide a detailed picture of the changes occurring in their structure and microstructure, their oxidation and hydrogen uptake properties.

I set myself the goal of designing and establishing those experimental configurations in the CODEX facility that make it possible to investigate different operational occurrences and accident conditions. To this end, I assembled experimental bundles with different cladding alloys in different geometrical arrangements.

The objectives of the individual experiments were multifaceted. On the one hand, we investigated whether a loss-of-coolant event would lead to damage of the fuel elements. On the other hand, we modeled an air-ingress integral test under extreme conditions, in a steam- and oxygen-starved environment. Using metallographic examinations and online measurement data, we investigated which chemical reactions occurred in the zirconium cladding, to what extent zirconium oxidation and nitridation progressed during the process, and whether re-oxidation of zirconium nitrides also took place.

In the same experimental facility, I also modeled an accident scenario during which nitrogen enters the reactor core. In this case, I investigated whether heterogeneous nitride and oxide layers form in the fuel cladding in the presence of nitrogen, and whether this accelerates the oxidation of zirconium components in steam. I also examined whether nitrogen causes a serious chemical reaction with zirconium at high temperature (above 1000 °C), or whether it appears only as an inert gas.

By means of an integral experiment, I investigated what kind of degradation the cladding exhibits under high temperature severe-accident conditions. Two different cladding types were used in the experimental bundle, and I then compared the behavior of the two alloys during the very high temperature transient. My goal was to determine the extent of their oxidation.

I performed a LOCA test with the newly introduced thin-walled cladding type used in VVER-440 reactors in order to investigate whether cladding rupture occurs under accident conditions with the thinner cladding. In addition, we investigated the oxidation behavior of the new type of cladding manufactured from base material produced on sponge-based technology.

I also performed measurements with bundles assembled from chromium-coated (accident-tolerant) and conventional zirconium claddings, to investigate the extent of damage above 1300 °C, and to determine what degradation processes occur in each case. I intended to demonstrate that, whereas for conventional claddings, the embrittlement due to zirconium oxidation is the dominant process, in chromium-coated claddings the formation of a chromium-zirconium eutectic is what leads to cladding failure.

### **3 Experimental methods**

To perform the integral experiments, I used the CODEX experimental facility. I carried out the necessary modifications of the facility for each test based on the chosen scenarios of the individual experiments. The measuring section contains the investigated fuel rods that model the real-life reactor geometry and thermal-hydraulic behavior of the fuel as accurately as possible. The experimental bundle consists of seven electrically heated fuel rods. Before each experiment, I assembled the experimental bundle from different claddings. A significant part of the experimental work was the assembly of the bundle itself. The rods were equipped with internal heating using U shaped tungsten wires. As spacers between the cladding and the tungsten, I selected  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  pellets, whose thermal properties are similar to those of  $\text{UO}_2$  pellets. To ensure pressure resistance, one end of each rod was welded shut with a zirconium plug and the other end was sealed with a graphite seal. Each rod was connected to capillary tubes through which the internal pressure could be varied and measured online with individual pressure transducers. The surfaces of the rods were instrumented at different positions with K-type and high temperature W-Re thermocouples, which provide accurate data on the temperatures established in the different parts of the bundle. It was necessary to build a steam generator and a superheater for the facility, which supplied superheated steam during the experiment. The electrical power of the heaters was measured by means of a calibrated analog multiplier circuit. The composition of the gas leaving the bundle was measured with a Pfeiffer OmniStar GSD 320  $\text{O}_2$  quadrupole mass spectrometer. During the experiments, the data-acquisition system collected and stored the measurement data and also provided means to control the facility. After the experiment, the bundle was inspected with a PCE-VE endoscope,

then embedded in epoxy resin and cut. After appropriate polishing of the transverse sections, metallographic examinations were performed.

## **4 New scientific results**

### **Thesis point 1**

In the CODEX facility, I designed and established the experimental configurations that enabled the investigation of different operational occurrences and accident conditions. I created experimental bundles from cladding materials used in different nuclear power plants (E110, E110G, optZIRLO™, chromium coated optZIRLO™) in different geometrical arrangements. I developed the technical solutions that made it possible to ensure the required boundary conditions, and I set up the online data acquisition and control for the experiments. [S1][S2][S3][S4][S5][S6][S7][S8][S9][S10][S11][S12]

### **Thesis point 2**

Based on a series of experiments performed with the E110 and E110G cladding materials used in VVER-440 reactors, I confirmed that - under design basis accident conditions - a loss-of-coolant accident (LOCA) does not lead to damage of the fuel elements. In the CODEX-LOCA-200, CODEX-LOCA-200B, and CODEX-LOCA-E4 experiments, cladding failures were observed only under conditions in which the internal pressure exceeded the value expected during a real accident, or when the high temperature steam oxidation lasted longer than would be conceivable according to the reference scenario. The CODEX-LOCA-E4 experiment also indicated that the sponge-based fuel cladding oxidizes less and preserves its integrity for a longer time under beyond design basis accident conditions. [S1][S2]

### **Thesis point 3**

With the CODEX-SLIM loss-of-coolant accident measurement performed on the newly introduced thin-walled E110G (Zr1%Nb) alloy cladding used in VVER-440 reactors, I demonstrated that fuel rod rupture does not occur under accident conditions even with lower cladding thickness. Furthermore, the sponge-based cladding material showed much better oxidation behavior than the conventional E110 alloy that was used previously. [S8][S9]

### **Thesis point 4**

I modeled a severe accident scenario involving air ingress under extreme conditions, in a steam- and oxygen-starved environment. As a consequence of the high temperature chemical reactions, complex oxide and nitride structures formed in the zirconium cladding made of E110G alloy.

The online measurement data and the metallographic examination of the bundle confirmed that in the CODEX-AIT-3 experiment, not only zirconium oxidation and nitridation took place during the process, but re-oxidation of zirconium nitrides also occurred. [S3]

### **Thesis point 5**

In the CODEX-NITRO experiment, I modeled an accident during which nitrogen enters the reactor core under loss-of-coolant accident conditions. I showed that in the presence of nitrogen, heterogeneous nitride and oxide layers form on fuel cladding made of both E110 and E110G alloys, and this accelerates the oxidation of zirconium components in steam. The experiment also indicated that, whereas at lower temperatures (below 500 °C) nitrogen appears in the accident process only as an inert gas, at higher temperatures (above 1000 °C) it produces a serious chemical reaction with the zirconium cladding. [S4][S5][S6]

### **Thesis point 6**

By means of an integral experiment, I demonstrated that high temperature severe accident conditions caused by a total loss of electrical power in a nuclear power plant (station blackout) may lead to significant degradation of the fuel cladding. Comparison of the two cladding types (E110 and E110G) used in the CODEX-SBO experimental bundle did not indicate a significant difference during the very-high-temperature transient. Both cladding types underwent extensive oxidation. [S6][S7]

### **Thesis point 7**

From the integral measurement performed with the CODEX-ATF bundle assembled from chromium coated (accident tolerant) and conventional optZirlo™ zirconium claddings, I concluded that both cladding types may be damaged above 1300 °C. The degradation processes, however, are different: whereas in the conventional cladding the embrittlement due to zirconium oxidation is the dominant degradation process, in the case of chromium-coated cladding, the formation of a chromium-zirconium eutectic leads to the failure of the cladding. [S10][S11][S12]

## 5 Potential applications of the results

The experimental results collected during my work from integral experiments performed with E110, E110G, E110G SLIM, optZIRLO™, and chromium-coated optZIRLO™ alloys support, on the one hand, the work of the nuclear safety authority and, on the other hand, provide a basis for the computer-aided development of fuel behavior models. They help the understanding of severe accident processes because they provide detailed data on the oxidation, nitride formation, and hydrogen uptake of zirconium claddings in different atmospheres (steam, air, H<sub>2</sub>, N<sub>2</sub>), and they also help to reveal the mechanisms of cladding degradation (oxide layer cracking, breakaway oxidation, nitridation–reoxidation cycle).

The CODEX measurement data can also be used for fuel performance code development and validation (ASTEC, MELCOR, ATHLET-CD, FRAPTRAN, TRANSURANUS). The CODEX-ATF and CODEX-ATF-AIT experiments provide indispensable measurement data for the simulation of the latest generation of accident tolerant fuels. These data were also used by researchers at Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) GmbH in Munich to improve oxidation calculations in the ATHLET-CD code and to develop the oxidation calculation of chromium-coated claddings in the latest AC2 program package (AC2 = ATHLET + ATHLET-CD + COCOSYS).

The measurements may contribute to the refinement of LOCA criteria, which may in turn provide a basis for the modification of Hungarian and international regulations (for example, regarding the fraction of fuel rods that may rupture during LOCA). In this way, they assist authorities in making well founded decisions in fuel licensing and safety matters.

The CODEX measurement databases have been used in several EU and IAEA projects. International comparison tests (e.g. parallel QUENCH–CODEX measurements) serve as a validation basis and make it possible to evaluate the effects of several important phenomena.

The comparison of E110 and E110G alloys clearly demonstrates that the application of the E110G cladding introduced at the Paks Nuclear Power Plant is more advantageous from the safety point of view. It also provides a reassuring confirmation that for the fuel rods with the thinner cladding burst failure may not occur under design basis accident conditions.

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- g M. Steinbrück, U. Stegmaier, M. Große, L. Czerniak, E. Lahoda, R. Daum, K. Yueh, High-temperature oxidation and quenching of chromium-coated zirconium alloy ATF cladding tubes with and w/o pre-damage, J. Nucl. Mater., 559 (2022), Article 153470

## 7 Publications

### 7.1 Scientific publications related to the thesis points

- [S1] Zoltán Hózer, Imre Nagy, Nóra Vér, Róbert Farkas: Simulation of Loss-of-Coolant Accidents in the CODEX integral test facility, EHPG, Lillehammer, Norway, 25-28. September, 2017, paper F2.7
- [S2] Hózer, Zoltán; Nagy, Imre; Farkas, Róbert; Vér, Nóra; Horváth, Márta; Novotny, Tamás; Perez-Feró, Erzsébet; Király, Márton; Kis, Zoltán; Maróti, Boglárka et al. Experimental Simulation of the Behavior of E110 Claddings under Accident Conditions Using Electrically Heated Bundles In: Yagnik, Suresh K.; Motta, Arthur T., Zirconium in the Nuclear Industry: 19th International Symposium ASTM International (2021) pp. 813-832. Paper: STP162220190010, 20 p.
- [S3] R. Farkas, Z. Hózer, I. Nagy, N. Vér, M. Horváth, M. Steinbrück, J. Stuckert, M. Grosse: Effect of steam and oxygen starvation on severe accident progression with air ingress, Nuclear Engineering and Design 396, 2022
- [S4] Nagy, I ; Farkas, R ; Vér, N ; Hózer, Z ; Szabó, P ; Szabó, G ; Kostka, P ; Lajtha, G ; Téchy, Zs: A CODEX-NITRO integrális atomerőművi súlyos baleseti kísérlet NUKLEON 12 Paper: 222 (2019)
- [S5] R. Farkas, I. Nagy, N. Vér, Z. Hózer, M. Horváth, P. Szabó: The CODEX-NITRO experiment, 25th International QUENCH Workshop 22-24 October 2019 Karlsruhe, Institute of Technology Karlsruhe, Germany, Editor: Martin Steinbrück DOI: 10.5445/IR/1000100298
- [S6] R. Farkas, Z. Hózer, I. Nagy, N. Vér, P. Szabó, M. Horváth, P. Kostka, G. Lajtha: Experimental simulation of selected design extension condition scenarios without core meltdown (DEC-A) in the CODEX facility, Progress in Nuclear Energy, 161, 2023

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- [S8] Farkas R., Hózer Z., Kis Z., Perez-Feró E., Király M., Horváth L., Novotny T., Szabó P., Bubonyi T.: Hűtőközegvesztéses atomerőművi üzemzavar kísérleti modellezése, Anyagvizsgálók Lapja 2023/III., 2023
- [S9] Farkas, R., Hózer, Z., Kis, Z., Perez-Feró, E., Király, M., Horváth, M., Novotny, T., Szabó, P., Bubonyi, T. (2024). Simulation of loss-of-coolant accident with thin-walled cladding tubes. *Nuclear Materials and Energy*, Volume 40, 101695.
- [S10] R. Farkas, N. Vér, B. Bürger, P. Szabó, Z. Hózer: The CODEX-ATF experiment, 28th International QUENCH Workshop 5-7 December, 2023 Karlsruhe, Institute of Technology Karlsruhe, Germany, Editor: Martin Steinbrück DOI: 10.5445/IR/1000152245
- [S11] Z. Hózer, R. Farkas, N. Vér, B. Bürger: CODEX-ATF: Integral Bundle Test With Accident Tolerant Fuel, Proceedings of TOPFUEL 2024, Grenoble, ENS
- [S12] Róbert Farkas, Nóra Vér, Berta Bürger, Péter Szabó, Anna Pintér Csordás, Levente Illés, Zoltán Kovács, Dávid Cinger, Zoltán Hózer: CODEX-ATF Test, Experimental Programme of Accident Tolerant and Advanced Technology Fuels (ATFs), Final Report of a Coordinated Research project (Vol. 1), IAEA TECDOC (megjelenés alatt)

## 7.2 Additional scientific publications

- [S13] Hózer, Z; Nagy, I; Kunstár, M; Szabó, P; Vér, N; Farkas, R; Trosztel, I; Vimi, A: Experimental investigation of the coolability of blocked hexagonal bundles, *Nuclear Engineering and Design* 317 pp. 51-58., 8 p. (2017)
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- [S15] Farkas, Róbert, Lenkey László: Visszasajtoló kutak által okozott hőmérséklet-változás modellezése, *Hidrológiai Közlöny* 92: 3 pp. 74-78., (2012)

## 8 Summary

The primary objective of my research was to design and development an experimental facility capable of performing integrated tests of nuclear fuel elements of nuclear power plants under accident conditions. Throughout the course of my work, I developed the necessary engineering solutions to reliably establish and maintain the required boundary conditions for such experiments.

A wide range of accident scenarios was simulated on fuel rod bundles comprising multiple fuel rods. In addition to varying the boundary conditions associated with the accident scenarios, I also altered the type of fuel claddings that make up the bundle. Experimental results confirmed that, under design basis accident conditions, a loss-of-coolant event does not result in mechanical failure of the fuel elements.

Loss-of-coolant accident measurements conducted on the newly introduced E110G SLIM cladding material for VVER-440 reactors revealed that even with reduced wall thickness, cladding rupture does not occur under loss-of-coolant accident conditions. Furthermore, the novel sponge-based E110G cladding material exhibited substantially improved oxidation resistance compared to the conventional E110 zirconium alloy.

I conducted severe accident simulations involving air ingress under extreme conditions, specifically in steam- and oxygen-starvation conditions. A LOCA scenario with nitrogen ingress into the reactor core was also examined. The results demonstrated that, in the presence of nitrogen, heterogeneous nitride and oxide layers form on the fuel cladding, which accelerate the oxidation kinetics of zirconium-based components in steam atmosphere.

Integral experiments performed on mixed bundles containing both chromium-coated (accident-tolerant) and conventional optZIRLO<sup>TM</sup> claddings indicated that both cladding types are susceptible to damage above 1300 °C. However, the underlying degradation mechanisms differ: for conventional zirconium claddings, the embrittlement due to the high-temperature oxidation of zirconium is the dominant cause of failure, whereas for the chromium-coated cladding, failure is driven by the formation of a chromium–zirconium eutectic.