The properties of nuclear fuel cladding materials under normal operation and accident conditions

Extended summary of the PhD. thesis and the connected research

My name is Márton Király and I have been working in the Fuel and Reactor Materials Department of the Centre for Energy Research in Hungary for ten years. My original scientific background is in Chemical Engineering (BSc. and MSc. in chemical process engineering, BME). During my time at the university I started to engage in nuclear engineering topics, and after finishing my MSc. I applied for a job at the Centre for Energy Research. A significant part of my research is dedicated to the scientific support of the operation of Paks NPP (consisting of 4 units of VVER-440). During my time as a research fellow I engaged in various topics related to the safe operation of the NPP, especially from the point of view of the nuclear fuel cladding integrity. I designed experimental programs, developed models and performed evaluations that were well suited to the projects of the research institute. In this application, I try to summarize my research activities related to my PhD thesis. Although these listed tasks were mostly collaborative efforts, a significant portion of their design and implementation, the measurements and the evaluation of the results were my task, as well as writing numerous research reports and scientific articles. I took part in all the measurements themselves, while the evaluation and writing reports and articles were my own accomplishments.

One of the key questions in safety analyses of the nuclear power plants is whether the zirconium cladding of the nuclear fuel - as an important engineering barrier - remains intact, as damage to the cladding and loss of integrity can lead to the escape of radioactive isotopes. The introduction of new cladding materials requires verification of compliance with the criteria developed for the previous cladding, as well as verification of the numerical models used to model processes in the cladding and describe changes in the cladding material under both normal and off-normal conditions and, if necessary, their modification. The development and validation of the computer code models used for the calculations require measurement data that is representative of the various states of the fuel elements and provides information on the processes that take place in the fuel.

Over the past two decades, fuel manufacturers have begun intensive development to meet market needs and have developed, approved and introduced several new cladding alloys (such as optZIRLO™, M5™) with improved properties. The Russian fuel manufacturer has also developed a new variant of the relatively well-known Zr1%Nb cladding alloy E110 which is currently used in several VVER-type pressurized water reactors, and it was expected to be introduced in Hungary as well. Despite their very similar composition, significant differences were found between the newly developed Russian cladding material E110G (sometimes called E110M sponge base metal variant) and those of the E110 and Zircaloy alloys used in PWRs to the extent that new code-additions were required (Figure 1). I investigated several aspects of the normal and off-normal conditions of the fuel in order to characterize its behaviour and supply data for the fuel performance codes.

Figure 1: The two claddings after oxidation in high temperature steam. The E110 has spalling oxide layer (left), while the oxide on the E110G remains compact (right).

In addition to the information supplied by the fuel manufacturer, it is very useful to for both licensing and operation to have independent measurements and modelling experience in the country where the fuel is used. Therefore, based on the available literature and our own experimental data, I decided to determine and compare the mechanical properties of the currently used E110 alloy and the new E110G alloy tubes. Axial and ring tensile tests were performed with both cladding specimens at room temperature, elevated temperature (150 °C) and near the operating temperature (300 °C) to determine their axial and tangential ultimate tensile strength. In developing the new geometry of the specimens to be used for the tensile tests, I relied on the results of research carried out abroad and the data available on the topic in the domestic and foreign literature. In order to gain a deeper understanding of the processes involved in the measurements and to support the planned experiments, the sample geometries selected for tensile tests were also modelled by my colleagues using Finite Element method and the results of the tensile test simulations were evaluated (Figure 2).

In addition to the analysis of the untreated, as-received samples, the effect of treatments (inert heat treatment, oxidation in steam, hydrogenation) on the tensile strength was also investigated. The experimental design included slight oxidation in steam and hydrogen uptake resembling normal operating conditions. The high temperature heat treatment itself changes the structure of the cladding material, especially when the temperature reaches the phase transition temperature range (850-900 °C depending on the alloy), above which the crystalline structure of the zirconium is transformed. The axial and ring specimens were exposed to oxidation and inert atmosphere heat treatment at various temperatures and times to test the effects of the heat treatment and the oxidation separately. I evaluated the effects of the above treatments on the tensile strength of the two investigated cladding alloys. In the end over 300 tensile tests were performed at various temperatures and degrees of treatment. The evaluation of all the data was also part of my assignment, and it was also a significant part of my PhD thesis. I investigated each set of measurement data, using statistical analysis to identify the significant effects.

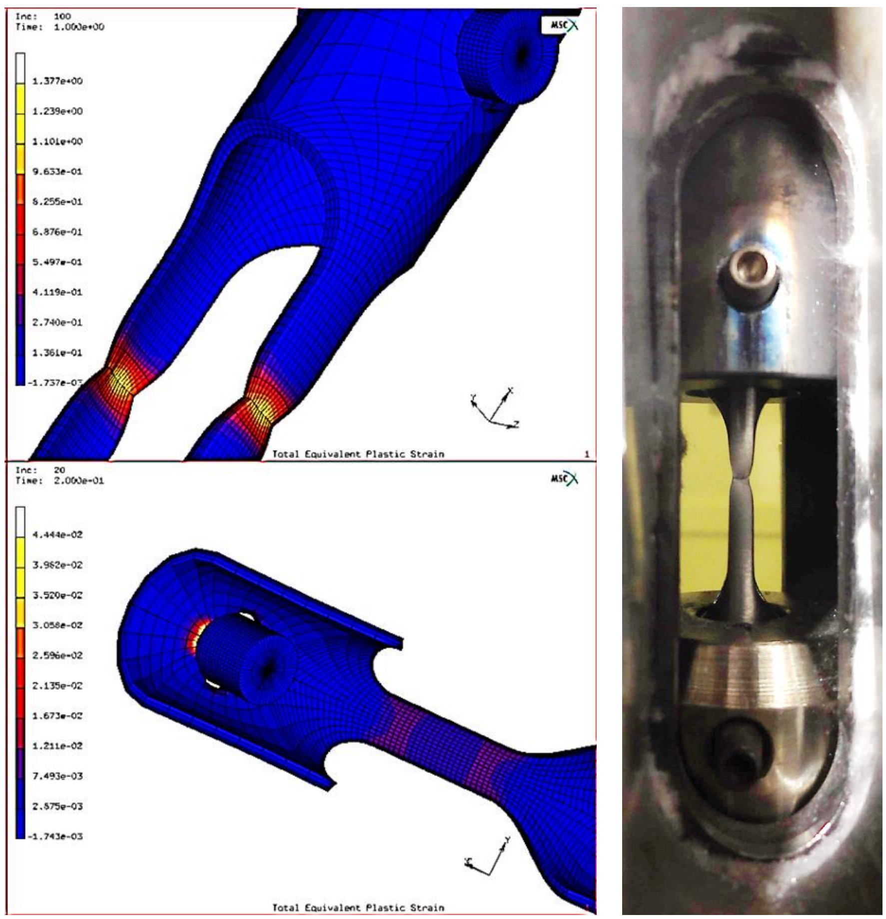

Figure 2: The Finite Element model of the axial test specimen (left) and the prepared sample during tensile testing (right).

During normal operation the gap between the fuel pellet and the cladding closes and in case of sudden changes in power during normal operation or reactivity accidents the thermal expansion of the pellet can rupture the cladding wall. Air ingress during high temperature dry storage can oxidize the fuel pellet and this also results in volumetric expansion as the higher oxides of uranium have lower density. This also can result in cladding rupture. The ductility of the cladding is an important parameter in both transport and storage conditions.

The mandrel test simulates the pellet-cladding mechanical interaction (PCMI) by expanding segmented dies (mandrels) inside cladding samples (Figure 3). This setup represents the actual mechanical conditions of the cladding better than the ring compression tests widely used to investigate cladding ductility. The mandrels are driven apart by a pyramidal taper connected to the crosshead of a universal testing machine, the force is measured under the mandrels. The friction coefficient is kept constant between the mandrels and the base plate they move on by applying graphite-based high temperature bearing grease. The results are the maximum force, the force-displacement curve integral, the maximal reached diameter and the mode of failure, which all give important information about the ductility of the sample.

Several sets of experiments were conducted in this setup, measuring the ductility of various cladding types and treatments. These included heat treatment, oxidation and hydrogenation as well. It was determined that the measured ductility of the cladding samples decreases with the amount of hydrogen content almost linearly up to about 2000 ppm absorbed hydrogen, after that the samples showed only minimal ductility before brittle fracture. Some new cladding materials with accident tolerant coatings were also tested to see how the coating behaves under load and if it changes the mechanical parameters. The length of the samples was 8 mm, but as of recently there is a new setup which can accept tube samples up to 30 mm with contact between the mandrels and the sample only in the middle of the tube to be able to recreate the axial cracking observed on damaged spent nuclear fuel.

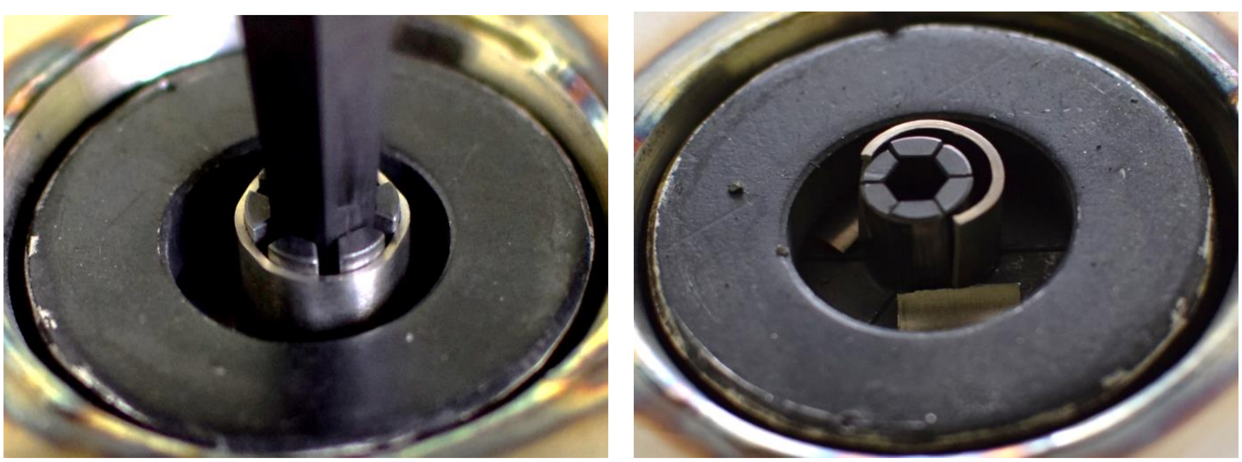

Figure 3: The mandrel test performed with a ductile (left) and a brittle sample (right).

The cladding endures different stresses during operation and as spent fuel in the interim storage. Long term pressure at high temperatures can lead to permanent deformation even if the stress was under the yield strength. This thermo-mechanical creep is an important parameter of the cladding tubes. In Hungary the maximum allowed temperature of the spent VVER fuel cladding during drying and dry storage is 400 °C. At this temperature the cladding may undergo thermal creep driven by the inner pressure of the initial helium, the fission gases and the helium resulting from the alpha decay. The spent fuel, which has undergone some corrosion during operation, may contain some dissolved hydrogen or precipitated hydrides, and it is important to quantify the effect of the hydrogen content on the creep properties.

The hydrogen gas was dissolved in the cladding samples in a vacuum furnace, the absorbed hydrogen content was verified by the mass increase of the sample and the distribution of hydrogen was measured by neutron-induced prompt gamma-ray spectroscopy. The structure and orientation of the hydrides before (on a parallel sample) and after the creep test was inspected by metallography. As the heat treatment during sample preparation may also have an effect, a heat treated control sample (treated in the same setup in pure argon atmosphere) was also be measured. The measurements included heat treated and hydrogenated samples with 1000 ppm hydrogen dissolved, and also as-received (untreated) samples and non-pressurized samples to quantify the diameter change due to oxidation by air or water vapour entering the furnace.

The tube furnace in which the tests took place is a three-zone electric resistance furnace, with a vacuum pump and a high purity argon gas canister to provide the inert atmosphere. The samples were pressurized through steel impulse pipes and capillaries connected to them by hydraulic fittings (Figure 4). The samples were removed from the furnace every 3-7 days and a self-built laser profilometer scanned the outer diameter at a given azimuthal orientation. The precision of the diameter measurement is 0.5 μm, the axial precision on the whole sample is about 20 μm. As a result of the tests it was concluded that the high amount of hydrogen absorbed in the cladding forms zirconium-hydrides in the cladding wall and these harden the material, which results in significantly lower creep rates. From these creep tests a number of parameters were calculated, some were implemented in the FUROM fuel performance code.

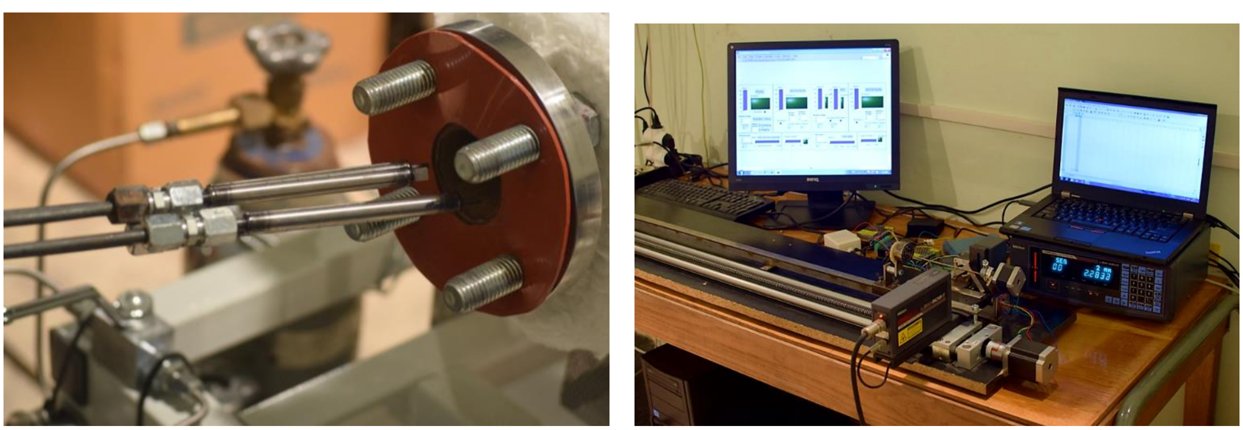

Figure 4: The prepared cladding samples (left) and a profilometry device (right).

In nuclear power plants, in case of a loss of coolant, the fuel elements may become heated to high temperatures and may intensively oxidize. Oxygen and hydrogen dissolved in the metal initiate changes in the material of the cladding which cause the plasticity of the metal to diminish and – above a certain concentration – the cladding may fail in a brittle way due to mechanical stresses. Several high temperature oxidation tests were performed in the Centre for Energy Research with both E110 and E110G claddings at temperatures between 600 °C and 1200 °C, oxidizing ring samples in an atmosphere containing argon and steam. In order to improve the transient fuel behaviour modelling codes and to verify existing models, I performed post-test calculations of the oxidation experiments. Based on the data, a new oxidation kinetics was established and supported by further experiments. The resulting best-estimate oxidation kinetics was included in both the FRAPTRAN and the TRANSURANUS fuel performance codes.

During a loss of coolant accident, the high temperature raises the inner pressure of the fuel that may lead to ballooning and burst of fuel cladding. These phenomena were investigated in a setup that ensures the high temperature and an isothermal constant pressurization of the cladding tubes We also wanted to observe the ballooning phenomenon and characterize the diameter evolution of the cladding samples. It required the design of a completely new experimental setup, with a large tube furnace that has viewing ports on its sides, a high-temperature lens system to allow a camera to record the tests, and a pressurization system to ensure the constant flow of argon gas into the samples at rates from 0.1 kPa/s to 1 MPa/s, and a data acquisition system to determine the burst pressure at different temperatures and pressurization rates. Different types of cameras were used for the imaging, regular HD cameras, high-speed cameras and an infrared camera with 350 frames/second to record the ballooning and crack initiation during the burst.

Several ballooning and burst tests were conducted in this setup and observations were made regarding the burst pressure, the diameter evaluation of the samples and the cracking behaviour. Several types of claddings were tested, including accident tolerant claddings that were coated in various ways to lessen the high temperature oxidation of the cladding and provide more time for the operators to react in case of accidents (Figure 5). A new correlation was fitted on the collected data to be able to estimate the burst pressure at a given temperature and pressure increase rate. Some samples were filled with ceramic balls arranged in a way to simulate the cracked fuel pellets that might escape the fuel rod after the integrity of the cladding is compromised.

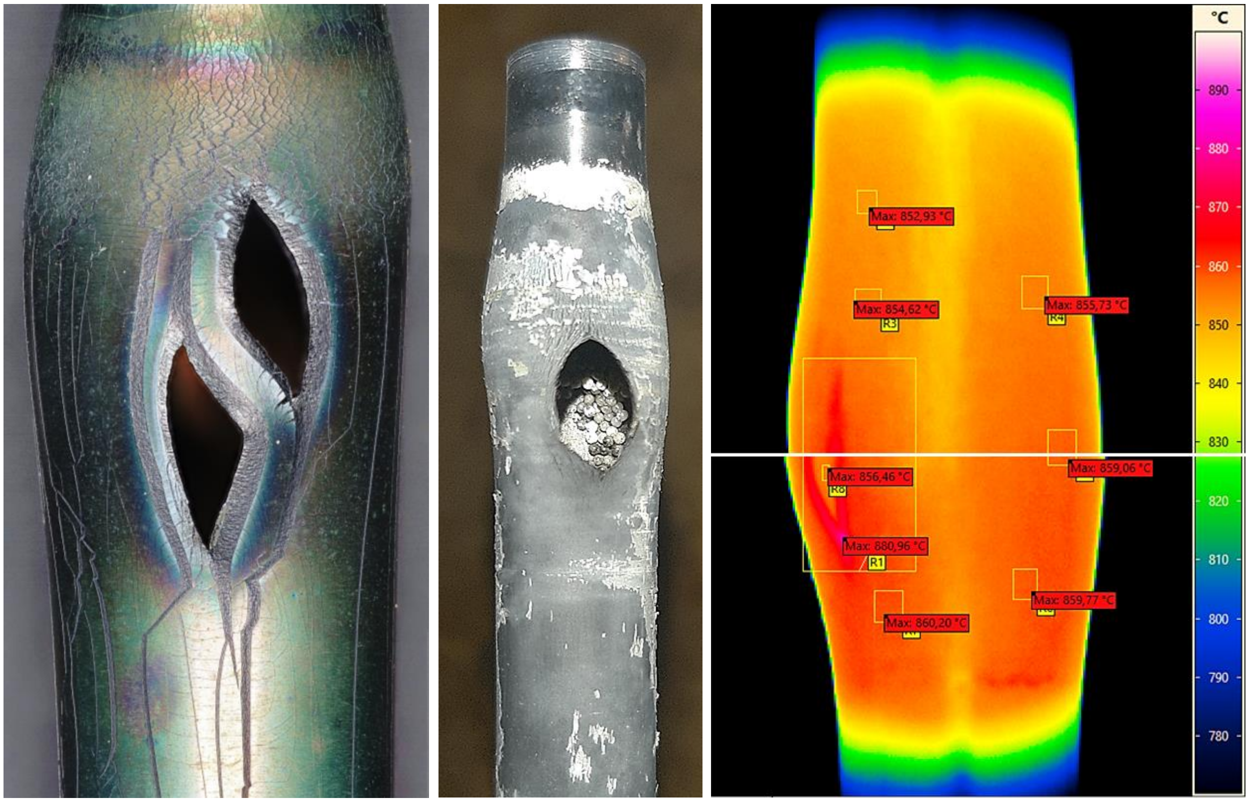

Figure 5: Cladding samples after ballooning and burst. Some samples were coated with accident-resistant coatings (left), some were filled with simulated fragmented fuel pellets (middle), and some were analysed in-situ using high-speed infrared cameras (right).

The cladding samples seemed to undergo uniform diameter increase, but right before the burst there is an asymmetric bulge formation and deflection on one side. Some samples were scanned post-test using CT and 3D profilometry to analyse the distribution of the wall thickness around the ballooned area. This included samples of various length, test temperature and pressure increase rate. Based on the results, a new estimation of the remaining strength of the inhermetic cladding tubes and the fuel assembly was implemented into the FRAPTRAN fuel performance code.

These different experiments were all parts of a collaborative effort to support the modellers in their effort to accurately and conservatively model the changes in the nuclear fuel during normal operation and design basis accidents for the safety analyses of Paks NPP submitted to the regulatory body, the Hungarian Atomic Energy Authority. These studies are required to ensure the safe operation and continuous improvement of the NPPs, for example licensing new fuel types, as well as to gain a deeper understanding of the mechanisms behind the physical changes in the fuel materials. The recent interest in new advanced and accident tolerant fuel designs is also an important research direction aimed to enhance the safety features of NPPs in the future and to test potential materials of the new generation advanced reactors.