Experiments With VVER Fuels to Confirm Safety Criteria

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Introduction

During loss-of-coolant (LOCA) accidents the primary reactor coolant system shall be maintained in a safe state so that the coolability of fuel rods was maintained and the emergency core cooling criteria must be met. These criteria were accepted originally for Western design reactors and the same criteria were applied in Hungary to the Paks NPP as well. However the VVER type reactor has different core materials and geometrical arrangement, so the Western experimental results may not be directly applied. That is why experimental series were conducted to check the following safety criteria:

- peak cladding temperature (PCT) is not allowed to exceed 1200°C,
- during oxidation the equivalent cladding reacted (ECR) is limited to 17% of the total cladding thickness at any local position on the fuel rod,
- fuel rods must keep a coolable geometry.

The embrittlement and the mechanical properties of the cladding plays an important role in the first two points. The criteria were established for Western Zircaloy type cladding [1]. In the VVER reactor Zr1%Nb cladding is used. There are some differencies between the mechanical properties of the two claddings (yield stress, ultimate strength, uniform elongation), the mechanical strength of the Zr1%Nb cladding is lower. In the same time the corrosion resistance of the Zr1%Nb cladding is higher and takes up less hydrogen and forms thinner external oxide layer during normal operation even at high burnup. Preliminary tests with Zr1%Nb and Zircaloy-4 cladding tube samples indicated differences between the two alloys during steam oxidation as well. The mechanical testing of the samples with ring compression method indicated that at the same level of oxidation the Zr1%Nb cladding became more brittle: the relative deformation before crushing was smaller for the VVER material than for the Zircaloy-4 (Fig. 1.). Further studies indicated that the hydrogen up-take in steam oxidation conditions is much higher in the Zr1%Nb cladding than in the Zircaloy-4. The external oxide layer has different structures for the two materials and the existence of cracks in the oxide can facilitate the hydrogen absorption by the metal and change the oxidation kinetics. All these facts indicated that the ECR and PCT criteria will not be satisfied automatically for the Zr1%Nb alloy and thus detailed investigation was needed for the VVER cladding. These criteria were traditionally checked by thermal shock tests and the ring compression tests were not considered as representative for reactor conditions. The Russian fuel supplier provided some related experimental data. The main goal of the thermal shock test series carried out in Hungary was to extend the database with independent and complementary experiments with Zr1%Nb cladding.

The third point is related to the ballooning phenomena, which can reduce the flow cross section between the fuel rods and if the reduction is too high the coolability of rods can be unsatisfactory and local melting can take place. In the Western PWRs the fuel rods are arranged on a square lattice in the assembly and several tests with such arrangement showed that even very strong deformation of the cladding can not lead to blockage formation in the bundle. The VVER reactors have hexagonal fuel arrangements and in this geometry the coolant channel between the rods is connected only to three rods. At the first glance it seems that the blockage formation in the hexagonal arrangement takes place easier than in the square lattice, for the cross section geometry is more simple and even one of the three rods could close the gap. Furthermore the VVER-440 assemblies are covered by zirconium shroud, which practically prevents the cross flows between the assemblies and so make more difficult the cooling in case of local blockage.

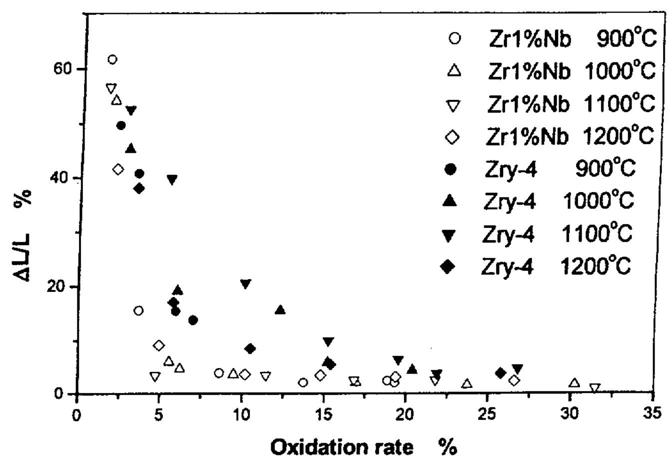

Fig. 1.
Results of ring compression tests with Zr1%Nb and Zircaloy-4 claddings preoxidised in steam: relative deformation until crushing of ring samples as function of oxidation ratio. Preoxidation took place under isothermal conditions at the indicated temperatures.

Thermal shock tests

In large break LOCA accidents most of the coolant flows out from the primary system through the break at the beginning of the process. After the blowdown period part of the fuel elements in the reactor can be heated up to high temperatures and oxidised in steam atmosphere before the emergency core cooling system injects enough water into the reactor vessel to reflood the core. The 1200 °C PCT criterion limits the temperature in order to avoid the autocatalytic Zr-steam reaction. This phenomena was earlier investigated in VVER integral tests, which indicated that temperature escalation took place above 1300-1400 °C [2]. The 17% ECR criterion shows where mechanical breakup of the cladding can be expected during the reflood of the hot and oxidised cladding below 1200 °C.

Thermal shock tests were performed to investigate the behaviour and failure of oxidised VVER cladding after initial heat-up and then quenching by water. The 5 cm long specimens were made of original Zr1%Nb cladding and the fuel pellets were replaced by alumina ones. The experiments started with heating up and continued with oxidation in steam atmosphere at constant temperatures. The on-line hydrogen concentration measurement provided detailed information on progress of oxidation process. After reaching the requested extent of oxidation the specimens were dropped into cold water to simulate the quenching process. The test matrix covered the most critical parameter ranges: temperatures between 1000-1250°C and oxidation time between 8-240 min (Fig. 2., Table 1.).

After the tests the mass gains due to oxidation were measured and metallographic examinations were performed to determine the changes in the cladding structure. The extent of cladding oxidation was calculated using the conservative oxidation correlation for Zr1%Nb alloy [3].

$$\Delta m = 920 \times \exp(-10410/T) \times \sqrt{\tau},$$

where Δm - weight gain, mg / cm^2;
T - temperature, K
τ - time, s.

Due to the oxidation and the heat treatment the zirconium alloy cladding became brittle, which could result in the failure of the cladding during quenching or during handling of

the samples after the tests. The oxidation created not only an external oxide layer on the surface of the cladding, but the oxygen diffused into the metal and formed oxygen stabilised α-Zr(O) layers, which replaced the original β-phase. The absorbed hydrogen increased the embrittlement of the alloy as well. Correlation was found between the Zr metallic characteristics and the cladding failures: specimens with β-layers were much less brittle than the specimens where the β-layers disappeared and only α-Zr(O) metallic phase was present. Fig. 3 shows the cross section of the sample oxidised at 1200 °C for 480 s, the existance of β-layer can be clearly seen and it saved the integrity of the rod. In case of much higher oxidation (e.g. 1200 °C for 7578 s, Fig. 4.) the β-layer disappeared and the remaining α-Zr(O) was unable to withstand the thermal shock.

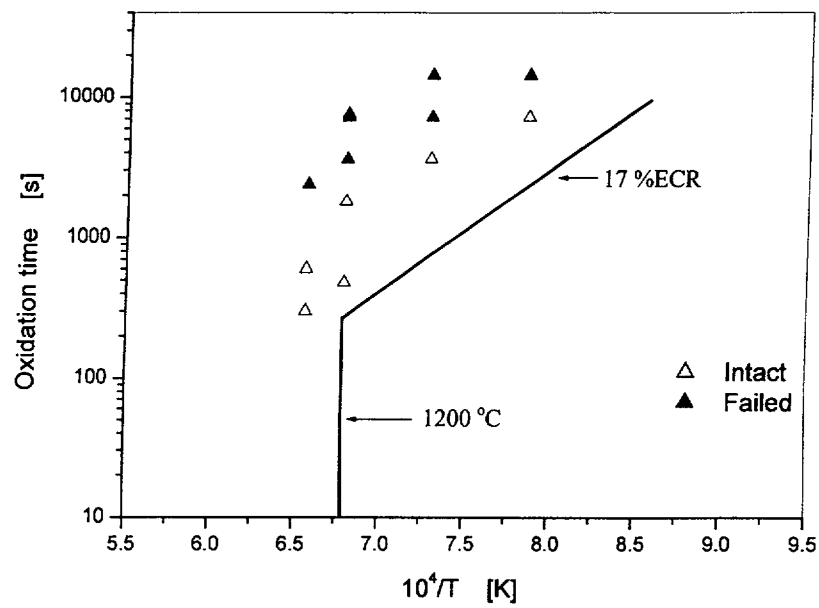

Fig. 2
Results of AEKI thermal shock tests

Table 1.
Results of thermal shock tests with Zr1%Nb cladding

Oxidation temperature °C	Oxidation time, s	State of specimen after the test
1200	1800	intact
1200	3600	failed during handling
1200	7578	failed during handling
1100	14400	failed during handling
1200	480	intact
1100	3600	intact
1100	7200	failed during handling
1250	2400	failed during quenching
1250	600	intact
1000	14400	failed during quenching
1000	7200	intact
1200	7200	failed during handling
1250	600	intact
1250	300	intact

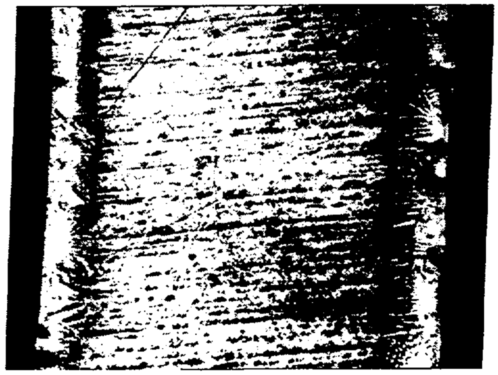

Fig.2
Cross section of Zr1%Nb caldding treated at 1200 °C for 480 s

Fig.3
Cross section of Zr1%Nb caldding treated at 1200 °C for 7578 s

The real oxidation limit between the failed and intact specimens was at ~40% ECR according to calculations with double sided oxidation. The experimental results were in good agreement with data of the Bochvar Institute, Moscow carried out with VVER cladding [3] (Fig. 5). The Russian tests indicated cladding failure below 40% several cases, but no failure was observed below the limiting 17%. The experiments confirmed that no fuel failure can be expected below the 1200°C temperature and 17% oxidation range calculated by the conservative correlation of safety criteria if only thermal stresses due to quenching are considered.

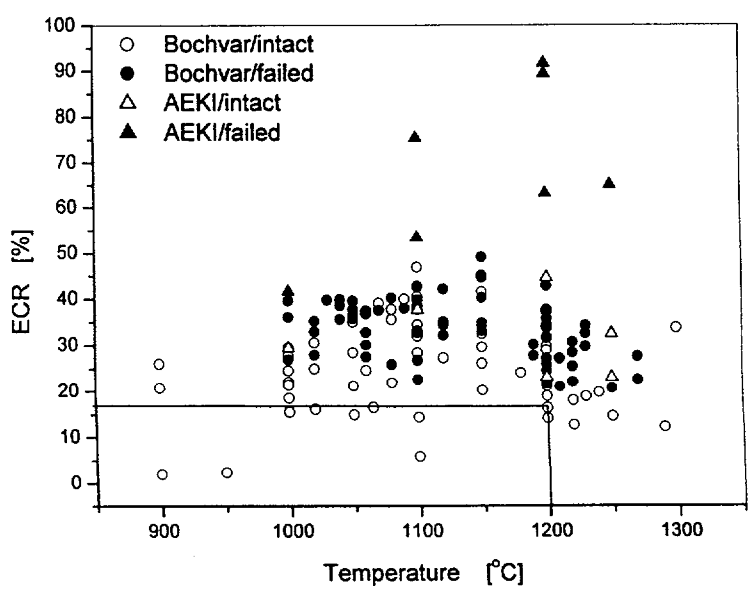

Fig. 5.
Results of Russain (Bochvar Institute [3]) and Hungarian thermal shock tests with Zr1%Nb cladding

Ballooning tests

During LOCA type accidents the fuel cladding can suffer strong deformation at high temperature due to high internal pressure. The ballooning may lead to the failure of the fuel rod integrity and to the reduction of the coolant flow cross section. For VVER reactor fuel single rod ballooning tests were performed earlier, which indicated large deformations before cladding failure [4]. Later the checking of blockage formation and the criteria for coolable geometry were carried out with 7-rod VVER bundles.

The experimental facility included a high temperature furnace with temperature regulation capabilities. On-line data acquisition of temperatures at three different elevations and pressures in each cladding tube were recorded. Each tube of the bundle was pressurised before the test by argon and the tubes were separated from each other.

Table 2.
Summary of 7-rod ballooning test parameters

No.	atmosphere	Inital pressure	Max. pressure	Temperature at the failure of the first rod	Max. temperature	Blockage rate
1	argon	10 bar	23 bar	800 °C	1000 °C	72 %
2	argon	3 bar	7.5 bar	1100 °C	1200 °C	57 %
3	argon	20 bar	35 bar	900 °C	900 °C	59 %
4	argon	30 bar	46 bar	800 °C	900 °C	-
5	argon	30 bar	53 bar	800 °C	900 °C	76 %
6	steam	30 bar	55 bar	900 °C	900 °C	43 %
7	steam	20 bar	36 bar	800 °C	900 °C	55 %
8	steam	10 bar	20 bar	900 °C	900 °C	57%
9	steam	3 bar	7 bar	-	1300 °C	34 %
10	argon	1 bar	11 bar	900 °C	900 °C	-

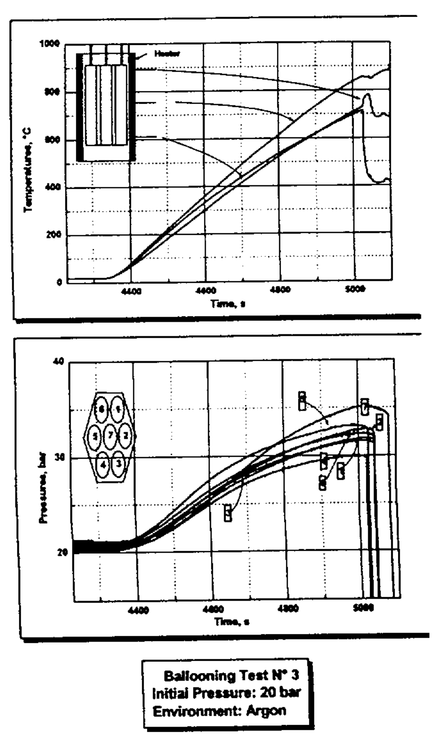

Fig. 6.
Pressure and temperature history of test No.3

Ten tests were performed with 7-rod bundles of hexagonal VVER geometry. The cladding was made of Zr1%Nb alloy, the length of the bundle was 15 cm. The experiments applied linear temperature increase until the failure of all rods. One series was performed in Ar atmosphere and another series in steam in order to study the effect of clad oxidation during the transient. The initial pressure varied between 3 and 30 bars. The maximum temperature reached in the tests was between 900-1300 °C, the rod failures were observed between 800-1100 °C. The rods with higher inital pressure failed earlier at lower temperatures. The rod failures happened in a narrow time period. Usually the central rod failed last, for the higher temperatures appeared at the peripheral rods. The main characteristics of the tests are summarised in Table 2. In test No. 4 the spacer grid was displaced during the heat up period and for this reason the conditions were repeated in test No. 5. The initial pressure in test No. 9 was too low to reach failure of the rods and the experiment was stopped at maximum temperature of the furnace. Test No. 10 was carried out under isothermal conditions with linear pressure increase. In this tests the 7 rods were connected to a common collector and the test ended with failure of the first rod.

After the tests the bundles were filled up with epoxy and several cross sections were prepared from each bundle for post-test examination (Fig. 8.a and. 8.b). The axial distribution of deformation of each rod of each bundle was measured. The typical blockage rate (which shows the cross section reduction compared to the nominal value) was between 40-50% and the maximum was below 80%. The ballooning took place on the hottest 4-5 cm long part of the bundle (Fig. 7.a and 7.b).

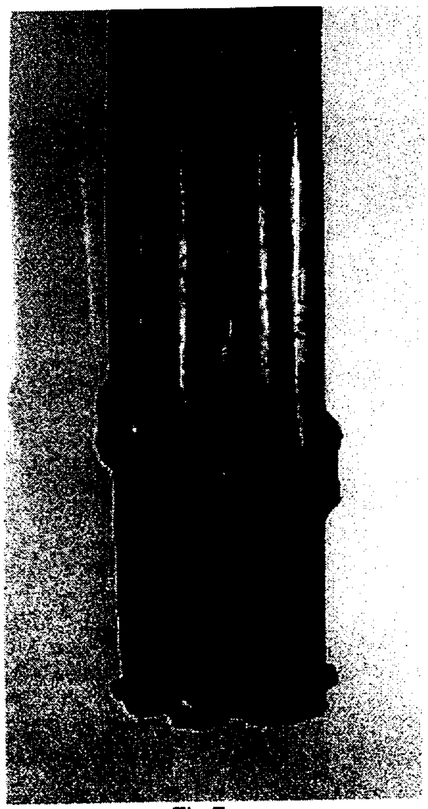

Fig.7.a
7-rod ballooning test in argon, 20 bar initial pressure

Fig.7.b
7-rod ballooning test in steam, 10 bar initial pressure

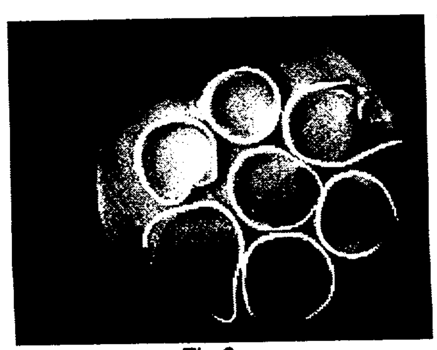

Fig.8.a
Cross section of the bundle after ballooning test in argon, 30 bar initial pressure

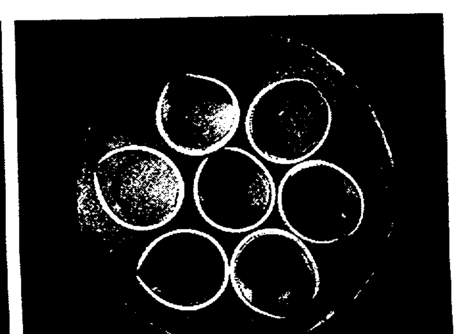

Fig.8.b
Cross section of the bundle after ballooning test in steam, 20 bar initial pressure

The reduction of the coolant flow cross section did not lead to total blockage formation, so the experiments confirmed that the VVER type fuel bundles with hexagonal arrangement of fuel rods will keep coolable geometry in LOCA accidents. Parallel to the 7-rod

Hydrogen up-take of Zr alloys

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Objective

Earlier experimental work showed that the embrittlement of Western type Zircaloy and the Russian type Zr1%Nb alloys during steam oxidation are different. Detailed investigations pointed out that first of all the H content was responsible for the embrittlement process. The objective of the present work was to determine the amount of hydrogen absorbed in the Zr metal and to evaluate its effect on the cladding strength.

Results

The experiments showed that the Zr1%Nb cladding takes up several times more H than the Zircaloy cladding under similar steam oxidation conditions. The higher H uptake resulted in earlier embrittlement of the VVER type cladding. The on-line H measurements indicated the formation of cracks in the Zr1%Nb cladding oxide, which facilitated the absorption of H in the metall phase.

No H absorption was observed below 7-14 μm thick oxide layer. Crack fomation (break up effect) took place in thicker oxide layers only and initiated the H up-take. The Zircaloy cladding showed break up and took up H only at 1000 °C.

During mechanical testing the Zr1%Nb samples crushed at lower force and at lower deformation than the Zircaloy samples oxidised up to the same extent.

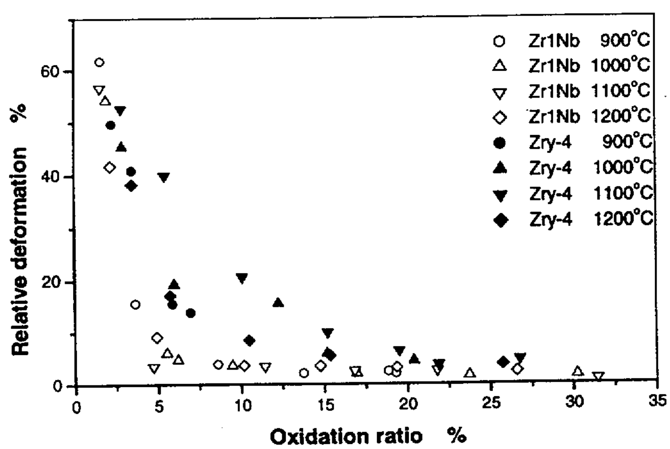

Fig. 1.
Relative deformation of oxidised cladding until failure in ring compression tests.
The low deformation of Zr1%Nb cladding (empty signs) indicates higher embrittlement.

Methods

A separate effect test faciltity was developed for the oxidation of Zr samples. It included a high temperature vertical furnace with steam supply and an on-line H monitoring system. The ring type cladding segments were oxidised between 900-1200 °C in steam/argon atmosphere under isothermal conditions. The integrated H content was determined using high temperature desorption method. The mechanical investigations were performed on tensile test machine.

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Remaining work

The numerical simulation of tensile tests is forseen with finite-element codes.

ballooning test in AEKI similar experiments were carried out in Russia. The Bochvar Institute [5] and the Gidropress Company [6] conducted 3 and 16 test with 19-rod bundles of VVER type. The experimental conditions covered a wide range of parameters considering pressure, temperature, atmosphere and the highest blockage rate in those cases remained below 80%.

Conclusions

The thermal shock and ballooning tests with VVER materials proved that the fuel rods with Zr1%Nb cladding material fulfil the emergency core cooling safety criteria used in Hungary. The experiments confirmed that with Zr1%Nb cladding

- no fuel failure can be expected due to thermal stresses during quenching below the 1200°C temperature and 17% oxidation range calculated by the conservative correlation of safety criteria,
- the reduction of the coolant flow cross section due to ballooning does not lead to blockage formation.

The test results were in good agreement with similar Russian experimental data concerning both thermal shock and bundle ballooning tests.

Acknowledgements

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