Understanding LOCA-Related Ductility in E110 Cladding

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Background

One of major economic requirements to the world nuclear industry is to increase the fuel cycle length and fuel burnup (60 MW d/kg U and higher)

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Numerous investigations performed during the last ten years allowed to understand that the key factor of this problem is mechanical properties of irradiated claddings

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Moreover, the analysis of associated test data has shown that zirconium niobium alloys are as a best candidates for the cladding material of the high burnup fuel

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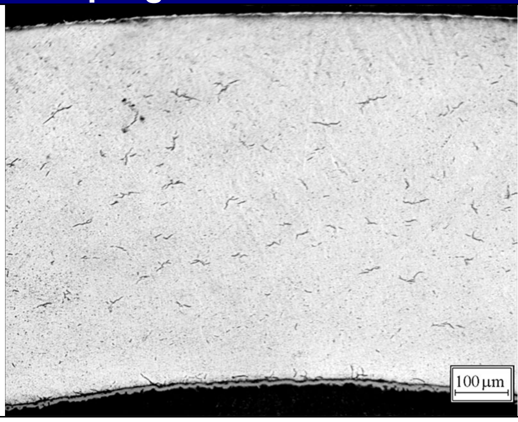
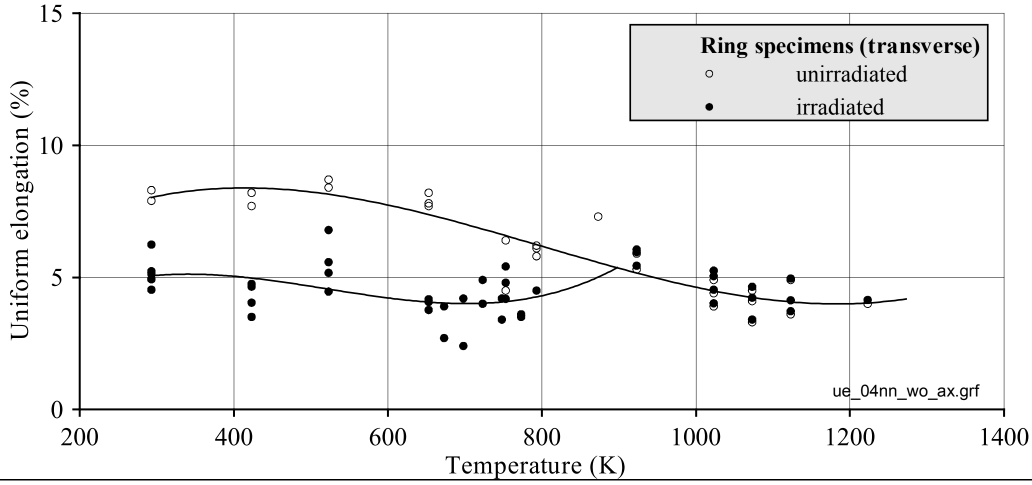
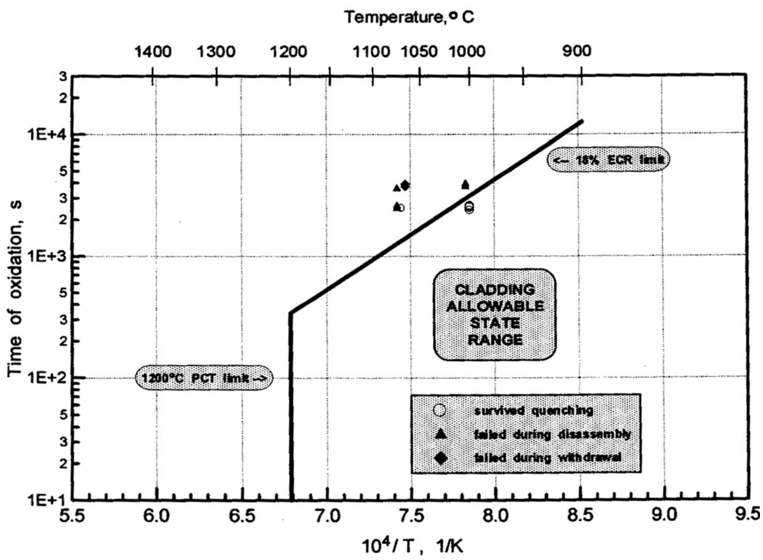
Important components for this conclusion are:

- results of more than thirty years of the Russian experience of the operation of VVER NPPs with Zr-1%Nb (E110) claddings of fuel rods
- results of special tests performed to justify the behavior of VVER high burnup fuel under the design basis accident conditions

Background (continued)

<table>
<tr><th colspan="3">Major findings of special test programs with E110 commercial irradiated claddings</th></tr>
<tr><td rowspan="2">The end of operation</td><td>Corrosion behavior</td><td>

← Typical appearance of the E110 irradiated cladding (~60 MW d/kg U)</td></tr>
<tr><td>Mechani-cal properties</td><td>
</td></tr>
<tr><td>Behavior under RIA condi-tions</td><td colspan="2">
Appearance of the E110 high burnup fuel rod (~60 MW d/kg U, 0.1 MPa) after the narrow pulse in the BIGR reactor at the peak fuel enthalpy ~165 cal/g → no fragmentation, no fuel dispersal</td></tr>
<tr><td>Behavior under LOCA condi-tions</td><td colspan="2">Bochvar institute/RIAR thermal shock tests with E110oxidized simulators of fuel rods refabricated from VVER commercial high burnup fuel rods (~50 MW d/kg U)

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Zirconium niobium alloys in the context of the international discussion on the LOCA safety analysis

1. Major issues of the discussion on the prevention of fuel fragmentation under LOCA conditions:

- Are the current safety criteria (1200/1204 C, 18(17, 15)% of the ECR) developed approximately thirty years ago representative now (especially for irradiated zircaloy claddings and new cladding materials (unirradiated and irradiated))?
- What types of tests should be used to validate the mechanical behavior of different oxidized claddings?

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2. Separate topic of the discussion:

- Can we consider that the mechanical behavior (margin of ductility) of oxidized unirradiated zirconium niobium cladding vs. the ECR is similar to that of the unirradiated zircaloy cladding?

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3. Basis for the question:

- German mechanical tests (J.Böhmert) and Hungarian mechanical tests (AEKI) with oxidized Zr-1%Nb (E110) claddings have demonstrated significant differences in the behavior of zirconium niobium and zircaloy alloys
- French mechanical tests (CEA-Saclay, EDF, FRAMATOME) with oxidized Zr-1%Nb (M5) claddings have demonstrated that there are no general differences in the mechanical behavior of both types of oxidized claddings (M5, Zry-4)

Goal of the work

1. Outlines of the work

The following issues were not the subject of this research:

- analysis of the representativity of current safety criteria
- selection of the best test procedures to validate the mechanical behavior of oxidized claddings under LOCA conditions
- analysis of LOCA scenarios

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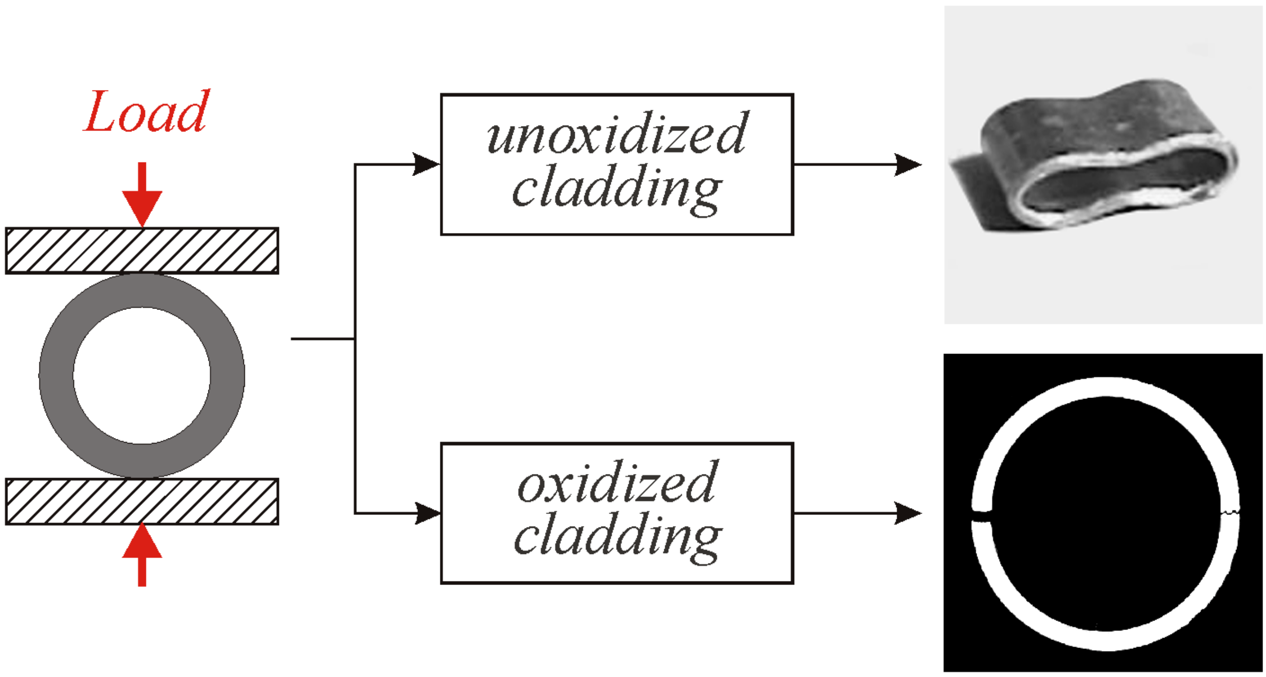
2. Goal of the work

To obtain the multifactor test data base characterizing the sensitivity of the mechanical properties of the E110 oxidized cladding to specially selected parameters. To use ring compression tests for this goal because:

- such type of tests was used to develop fuel safety criteria for zircaloy cladding (1204°C, 17%) in 1973 (NRC's approach)
- such type of tests allows to perform the comparative analysis of results obtained by different researchers for this cladding material and other cladding alloys

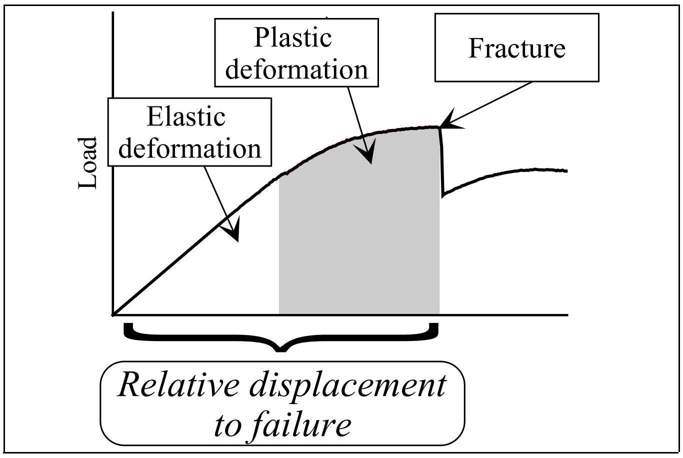
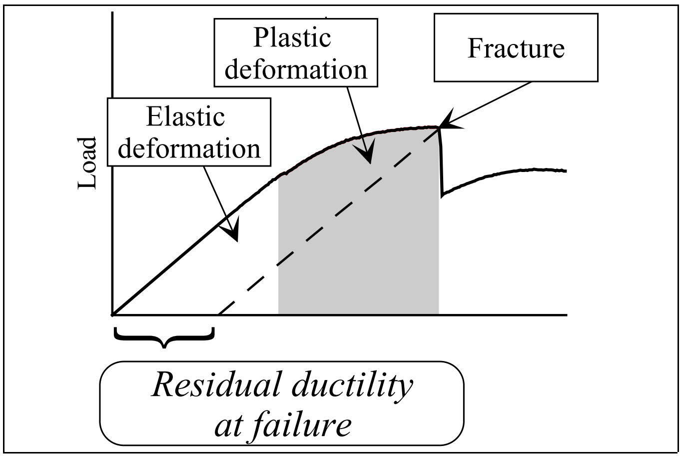
Interpretation of results of mechanical tests

1. Scheme of ring compression tests:



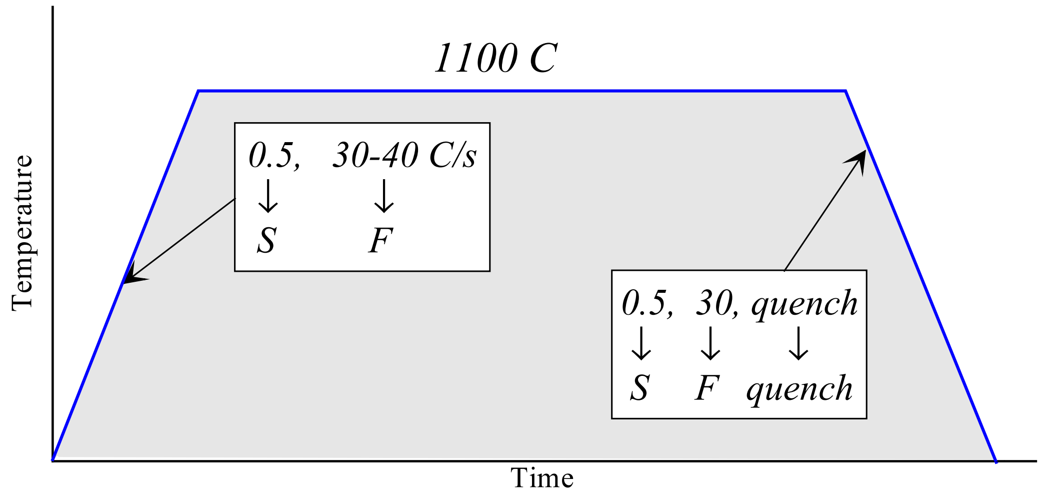
2. Processing of the load-displacement diagram:

The major revealed problem ⇒ there is not the common approach for the processing of ring compression test results:

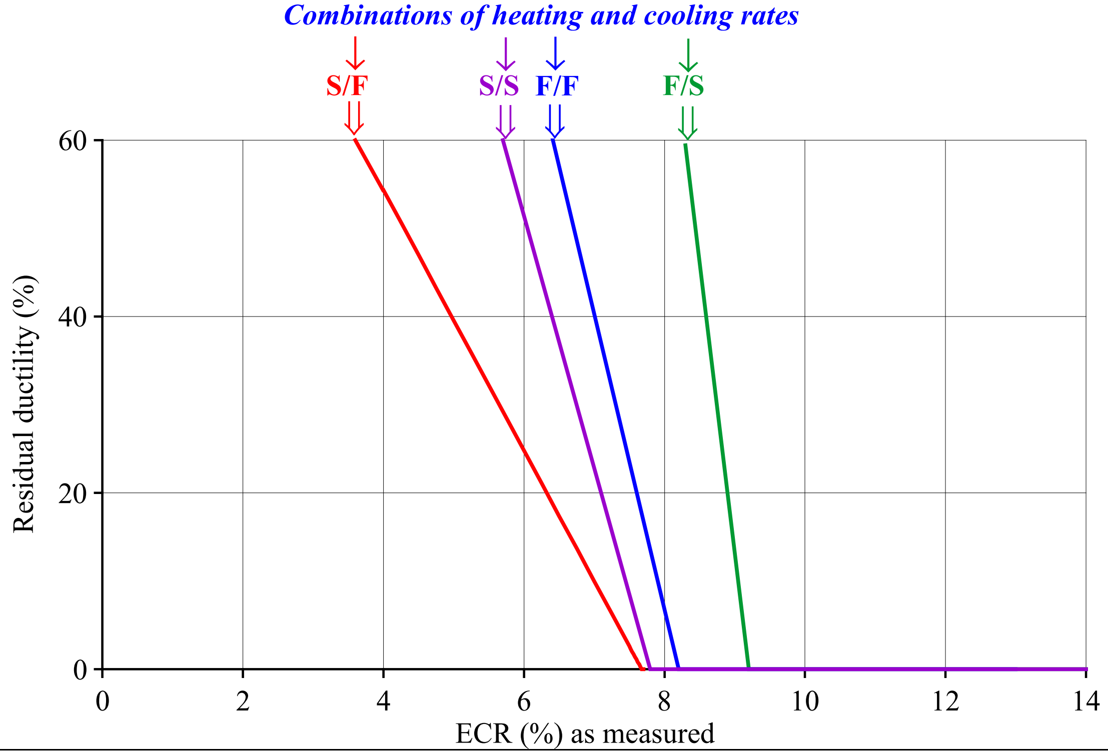
Previous popular approach	Approach proposed for this work
⇓	⇓
	
⇓	⇓
−	+
⇓	⇓
The zero ductility threshold is not determined in the explicit form	The residual ductility at failure of each brittle cladding equals to zero

First stage of the program: Sensitivity of the E110 oxidized cladding residual ductility to time dependent parameters of the LOCA

1. Variation of heating and cooling rates under high temperature oxidation conditions



2. Results of mechanical tests with E110 double sided oxidized claddings



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First conclusions:

- zero ductility threshold has a weak dependence on the heating/cooling rates
- E110 embrittlement threshold is about of 8.3% of the ECR for the typical test combination of heating and cooling rates (F/F)

Second stage of the program:
E110/Zry-4 comparative data

Double sided oxidation
T=1100 C
F/F combination of heating/cooling rates
E110, Zry-4 unirradiated claddings

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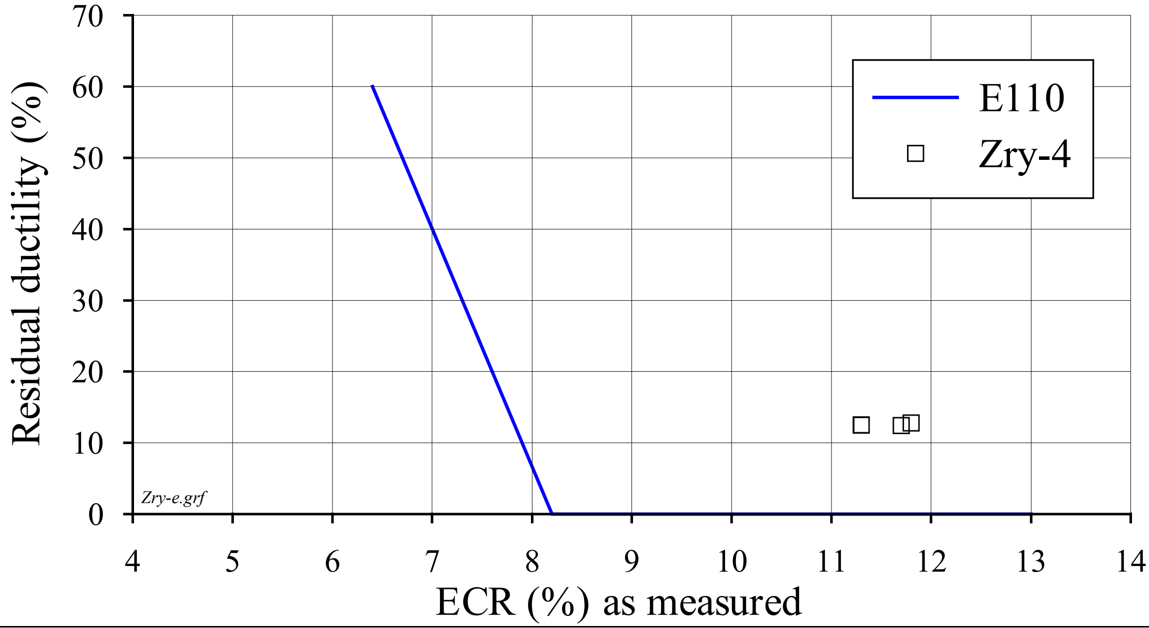

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Second conclusion:

the zero ductility threshold of E110 unirradiated cladding is lower than that of Zry-4 unirradiated cladding under ring compression test conditions

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Additional observations:

- the ductility of Zry-4 cladding vs. the ECR is decreased monotonely (in accordance with the published data)
- a substantial decrease of the ductility of E110 cladding occurs in the narrow range of the ECR

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Assumption:

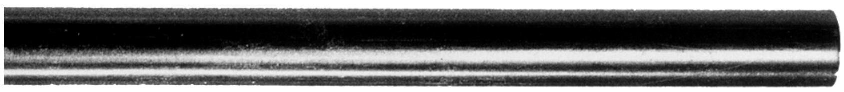
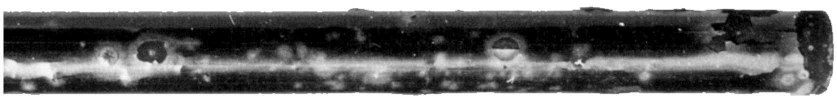
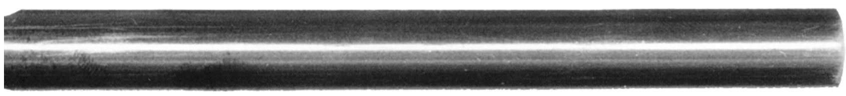
- different physical mechanisms are responsible for the oxidation behavior of E110 cladding before and after the criterial ECR

Third stage of the program: Development of the data base to clarify physical phenomena accompanying high temperature oxidation of the E110 cladding

The major directions of research:

1. Visual identification of ZrO_2 oxide type
2. Measurements of H_2 concentration in the E110 oxidized cladding vs. the ECR
3. Determination of the sensitivity of the cladding residual ductility to the H_2 concentration
4. Microstructure measurements and analysis

1. Type of the oxide in the function of the ECR

<u>E110</u> ECR=6.5%		Double sided oxidation at 1100 C and F/F combination of heating and cooling rates
<u>E110</u> ECR=10.5%		
<u>Zry-4</u> ECR=11.3%		

Some observations:

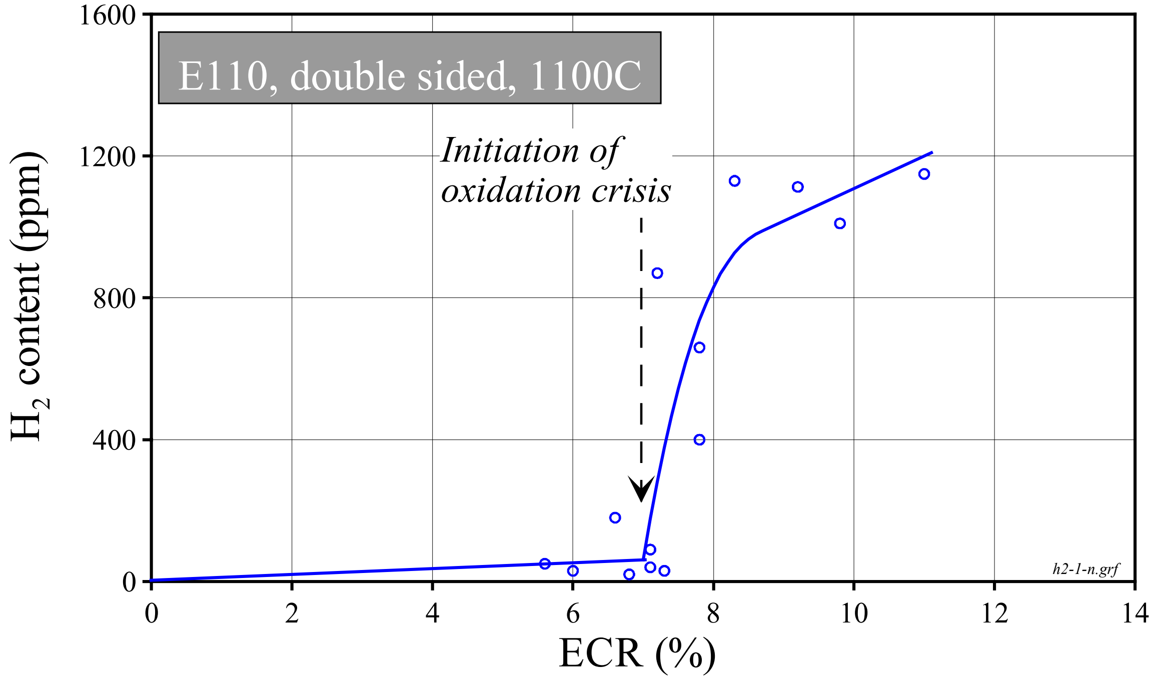
Type of cladding	Type of ZrO_2 oxide
E110 (ECR=6.5%)	Nonstoichiometric black protective oxide
E110 (ECR=10.5%)	Spalled light stoichiometric oxide
Zry-4 (ECR=11.3%)	Nonstoichiometric black bright oxide

Conclusion:

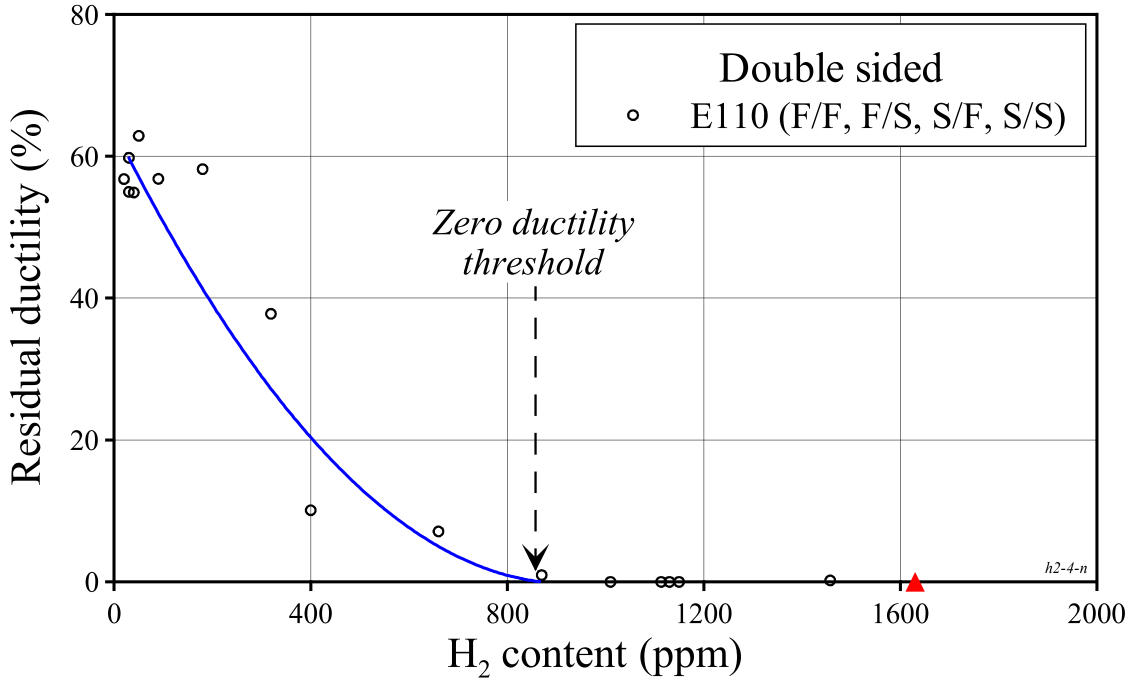
Breakaway oxidation occurs with the E110 cladding within this range of the ECR (6.5 – 10.5%)

Third stage of the program (continued)

2. Measurements of H_2 concentration in the E110 oxidized cladding vs. the ECR



3. Determination of the sensitivity of residual ductility to the hydrogen concentration



Conclusions:

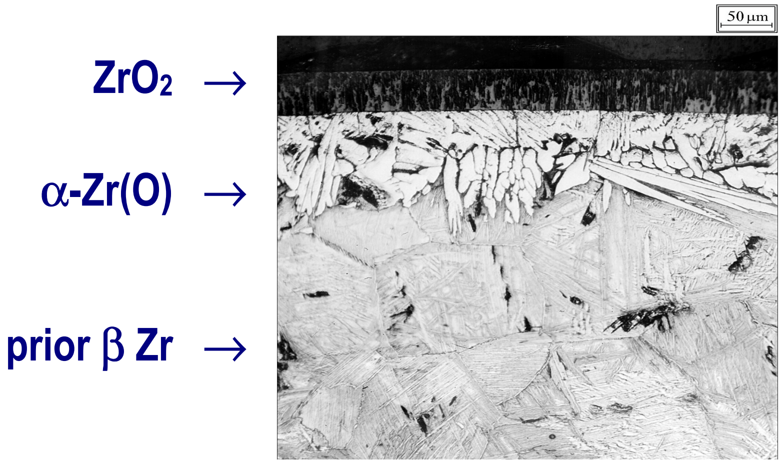
- **Fast increase of the hydrogen uptake occurs at the ECR>7%**
- **Zero ductility threshold corresponds to 900 ppm of hydrogen content (ECR≈8.3%)**

Third stage of the program (continued)

4. Microstructure measurements and analysis

4.1. Oxygen behavior in the E110 oxidized cladding

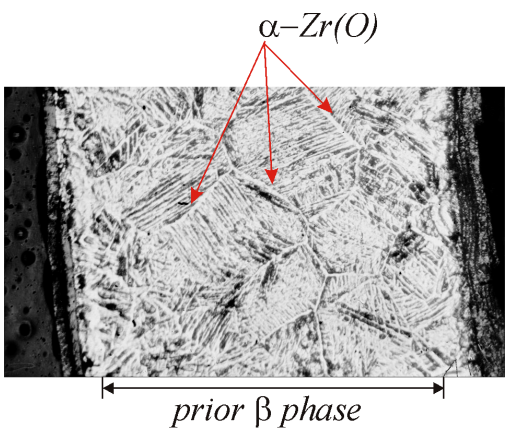
4.1.1 *Polish*



⇒ **The first observation:**

There is a fixed nonuniform boundary between the brittle α-Zr(O) layer and ductile cladding body (prior β phase)

4.1.2

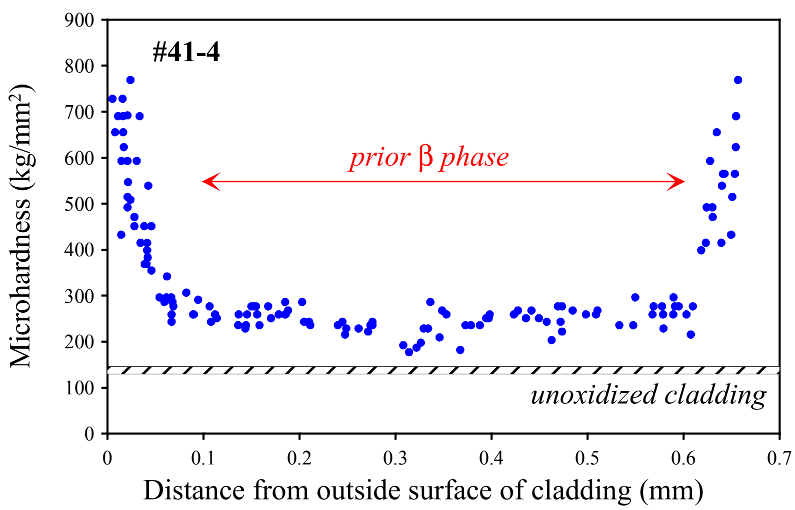


⇒ E110 specimen #41-4 after double sided oxidation at 1100 C to ECR=8.6%

The second observation:

α-Zr(O) layers are fixed on the boundaries of grains in the β-phase of the oxidized cladding

4.1.3 Oxygen distribution vs. cladding thickness

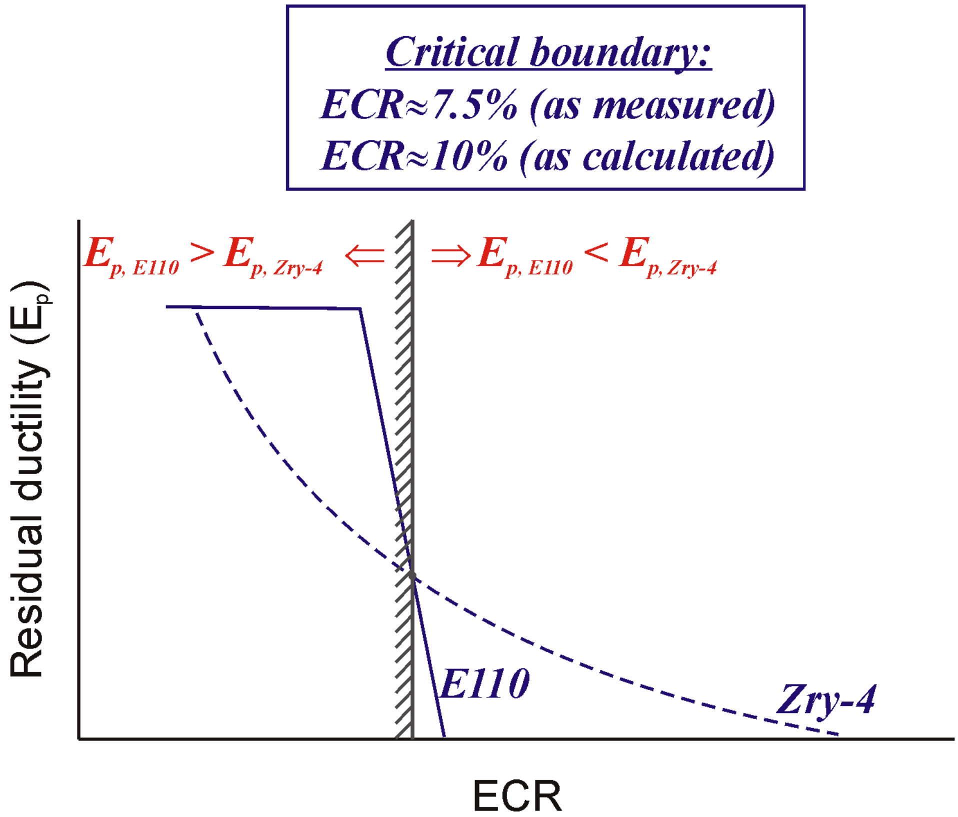


⇒ **The third observation:**

Relatively uniform distribution of microhardness (oxygen concentration) vs. β-phase thickness

Additional demonstration of some general differences in the mechanical behavior of the E110 and Zry-4 oxidized claddings

Comparative schematical data characterizing the ductility of both alloys versus the ECR (E110 data: this work; Zry-4 data are based on Böhmert's results)



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Observations	Possible explanations
Unlike Zry-4, the residual ductility of the E110 cladding is practically insensitive to the oxidation up to ECR=6.5%. Therefore, the margin of the E110 cladding residual ductility is higher than that of Zry-4 cladding up to ECR=7.5%	All absorbed oxygen is concentrated in surface layers of the E110 cladding
Unlike Zry-4, a sharp decrease of E110 ductility down to zero is noted at the ECR higher than 7.5%	The change of the mechanism of the E110 cladding oxidation results in substantial hydrogen (and oxygen?) uptake

Fourth stage of the program: Determination of the oxidized cladding residual ductility vs. the oxidation type (one sided, double sided)

Background:

- one sided oxidation of claddings characterizes the LOCA scenarios up to the cladding burst
- ring compression tests (performed in France) with the M5 one sided oxidized claddings did not reveal the sufficient differences in the residual ductility of zirconium niobium alloy (M5) and low-tin Zry-4

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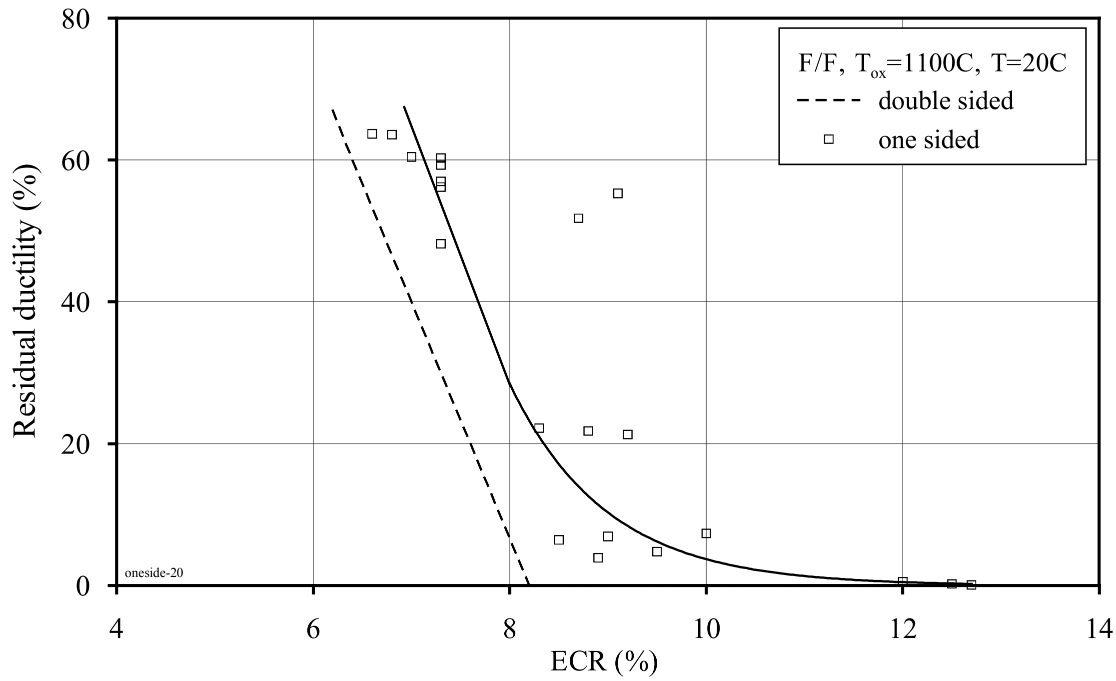
Comparative results of the E110 cladding residual ductility vs. the oxidation type



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Conclusion:

The residual ductility of Zr-1%Nb (E110) oxidized cladding is quite sensitive to the oxidation type

Fifth stage of the program: Sensitivity of the mechanical behavior of oxidized zirconium niobium claddings to the alloying components

Characteristic of some types of zirconium niobium alloys:

- Zr-1%Nb + low initial concentration of oxygen (~0.05%) ⇒ E110
- Zr-1%Nb + high initial concentration of oxygen (>0.11%) ⇒ E110K, M5
- Zr + Nb + Sn + Fe ⇒ E635, Zirlo

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Results of comparative tests

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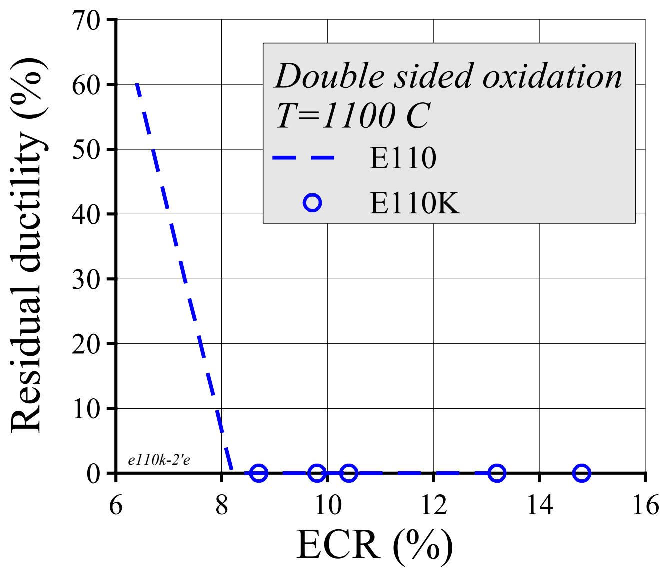
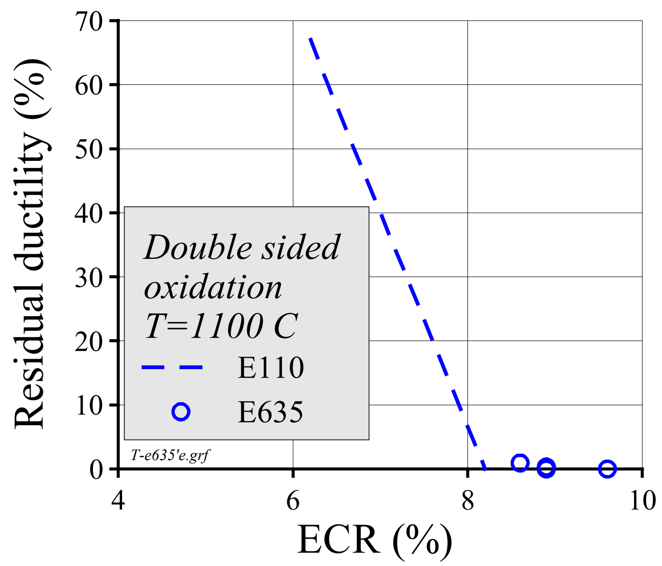
E110 and E110K



E110 and E635



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Within the limits of these test procedures the qualitative influence of alloying components of Zr-1%Nb alloys on the zero ductility threshold has not been revealed

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It is possible to assume that general differences in the mechanical behavior of Zr-1%Nb and Zry-4 oxidized cladding are determined by specific features of the Sn and Nb behavior during cladding oxidation

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The appropriate research is in progress now

Sixth stage of the program: Sensitivity of the E110 oxidized cladding residual ductility to the temperature of mechanical tests

- All previous stages of the program were based on the analysis of results of mechanical tests performed at 20 C
- The current safety criterion was developed and validated for Zry-4 unirradiated oxidized cladding at 135 C (Hobson's ring compression tests)

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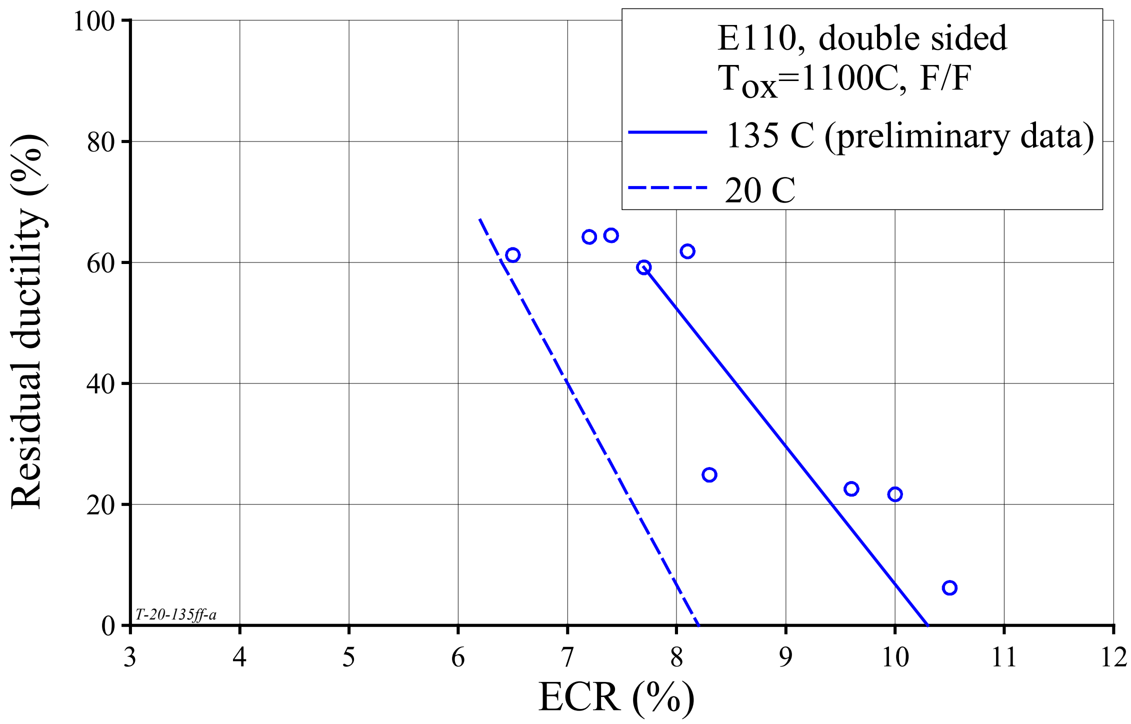
Comparative results of ring compression tests with E110 oxidized claddings at 20 C and 135 C



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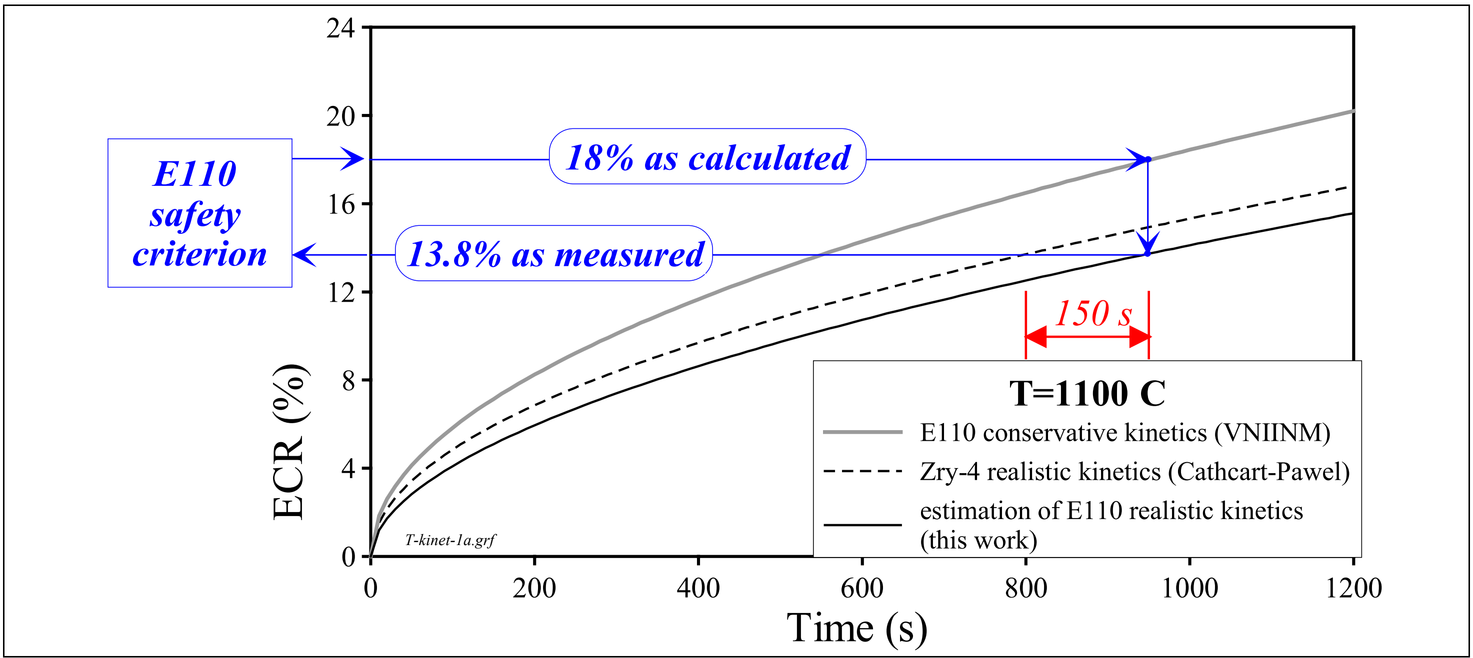
- The increase of the temperature of mechanical tests from 20 C up to 135 C leads to the increase of the E110 zero ductility threshold from ECR=8.3% up to ECR=10.3% (preliminary data)
- The possible explanation of this effect is based on the behavior of hydrides vs. the temperature

Seventh stage of the program: Estimation of the E110 realistic oxidation kinetic and determination of relationship between the values of the E110 safety criterion in conservative and realistic approaches



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- Results of 15 oxidation tests were used to obtain the estimation of the realistic oxidation kinetic for the E110 cladding at 1100 C
- Comparative analysis of conservative and realistic oxidation kinetics for E110 cladding allowed to determine the experimental value (13.8% as measured) of the E110 safety criterion
- Comparative analysis of Zry-4 and E110 realistic oxidation kinetics shown that the E110 cladding will reach the ECR=13.8% on 150 seconds later than the Zry-4 cladding

Eighth stage of the program: Sensitivity of the E110 cladding zero ductility threshold to the cladding irradiation

The oxidation tests and ring compression tests were performed with E110 irradiated claddings refabricated from VVER high burnup fuel rods (~50 MW d/kg U)

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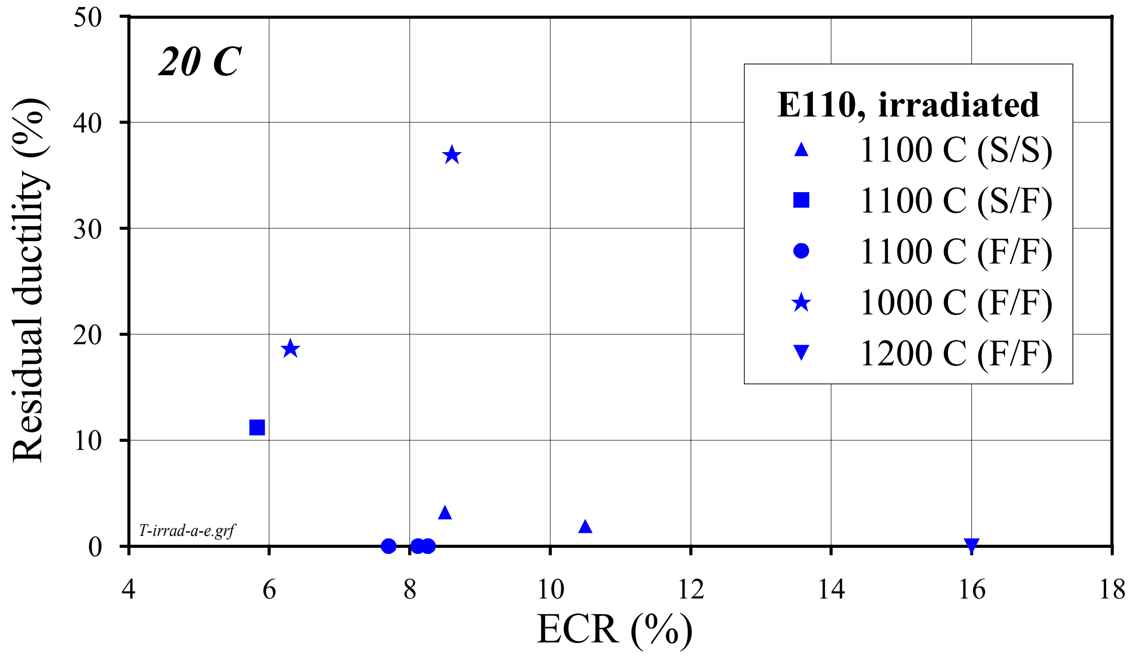

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- Obtained data allow to conclude that the zero ductility threshold of the E110 irradiated cladding is higher than ECR=6% at 20 C
- The increase of the temperature of mechanical tests from 20 C up to 135 C leads to the increase of the cladding residual ductility

Major conclusions

Experimental studies of the LOCA-related ductility in the E110 cladding were performed on the basis of ring compression mechanical tests. Results of several parts of this research program show that:

- the E110 unirradiated oxidized cladding (after double sided oxidation at 1100 C) keeps a very high level of ductility (>50%) up to:
  - ECR=8.5% for the best combination of heating and cooling rates
  - ECR=4.5% for the worst combination of heating and cooling rates
- After that, a sharp decrease of residual ductility is observed due to the breakaway effect
- A sharp increase of the hydrogen content in the prior β-phase of E110 cladding (>800 ppm) is responsible for the differences in mechanical behavior of the E110 and Zry-4 claddings
- The reference value for zero ductility threshold (at 20 C) of the E110 double sided oxidized cladding is 8.3% (ECR as measured)
- Comparative tests with Zry-4 cladding confirm that the Zry-4 oxidized cladding has the margin of residual ductility and low content of hydrogen at ECR=11.5% (as measured)
- The zero ductility threshold of E110 one sided oxidized claddings is increased up to ECR=12% (these data and French ones with M5 alloy may be compared directly only after the agreements of the test and processing procedures)
- The E110 cladding zero ductility threshold is sensitive to the mechanical test temperature. The increase of this temperature up to 135 C (so called ZDT temperature used on developing of Zry-4 safety criterion) leads to the increase of the embrittlement threshold up to ECR=10.3% (as measured)
- Special tests performed to reveal the ductility threshold sensitivity to the composition of zirconium niobium alloys (the increase of oxygen concentration (E110K alloy), the presence of such alloying components as Sn, Fe (E635 alloy)) show that there are no principle differences in the mechanical behavior of oxidized claddings from all tested alloys
- The zero ductility threshold of E110 irradiated oxidized cladding is higher than ECR=6% (as measured)