

Final Report on the Project # 1648.2

**Examination of VVER fuel behaviour under severe
accident conditions, Quench state (VVER-QUENCH)**

STAGE C. Quench Model

Presented by

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Budapest, Hungary
Hungarian Academy of Sciences KFKI
Atomic Energy Research Institute AEKI
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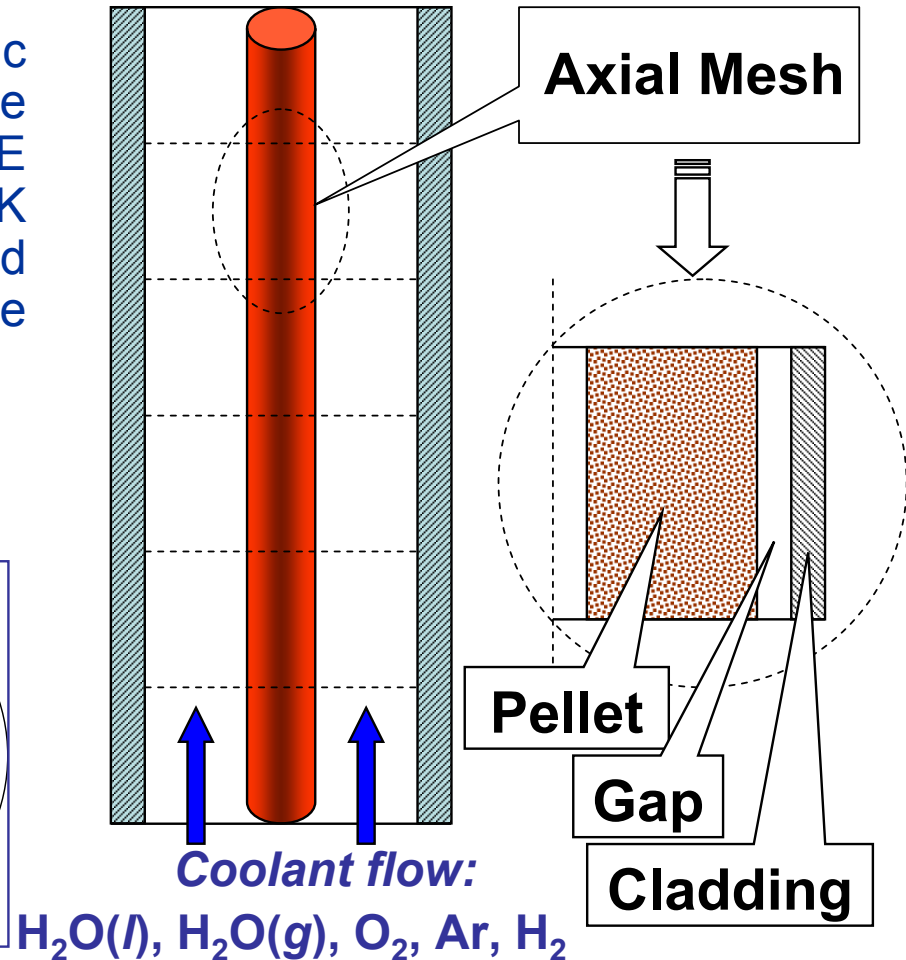
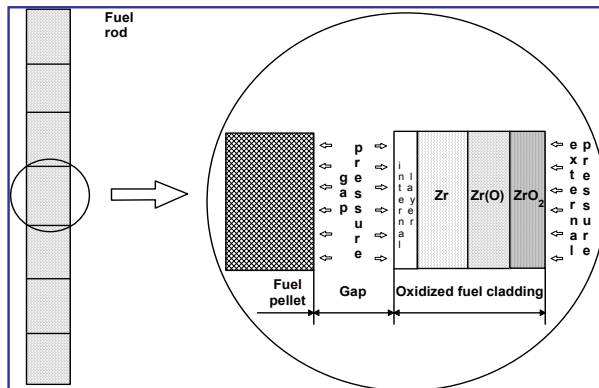
Stage C: Main Tasks

1. Application of the SVECHA/QUENCH code to simulation of the RIAR quenching tests with VVER fresh fuel rods
2. Application of the SVECHA/QUENCH and MFPR codes to simulation of the RIAR quenching tests with VVER irradiated fuel rods
3. Application of the SVECHA/QUENCH code to simulation of the QUENCH-12 bundle test with VVER fuel rod simulators (*included in presentation of J. Stuckert*)

**Part 1. Application of the SVECHA/QUENCH
code to simulation of
the RIAR quenching tests
with VVER fresh fuel rods**

Single Rod Code SVECHA/QUENCH

- Advanced mechanistic **SVECHA/QUENCH** code was developed by IBRAE in collaboration with FZK for theoretical support and modelling of FZK single rod quench tests



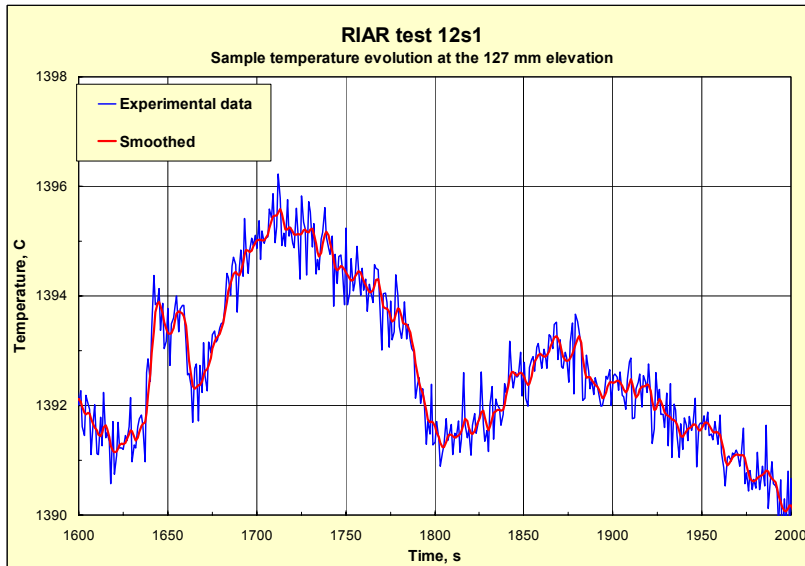
SVECHA/QUENCH code modification

- Initially the single-rod SVECHA/QUENCH (S/Q) code was developed for PWR fuel rod materials (in collaboration with FZK)
- To simulate the behaviour of the VVER fuel rod under severe accident conditions it was modified to take into account material properties of VVER fuel rods
- The necessary adaptations of the S/Q code were performed and the following modules were modified:
 - ✓ the heat exchange module,
 - ✓ the cladding oxidation module,
 - ✓ the mechanical behaviour module,
 - ✓ the driver module (to take into account all modifications).

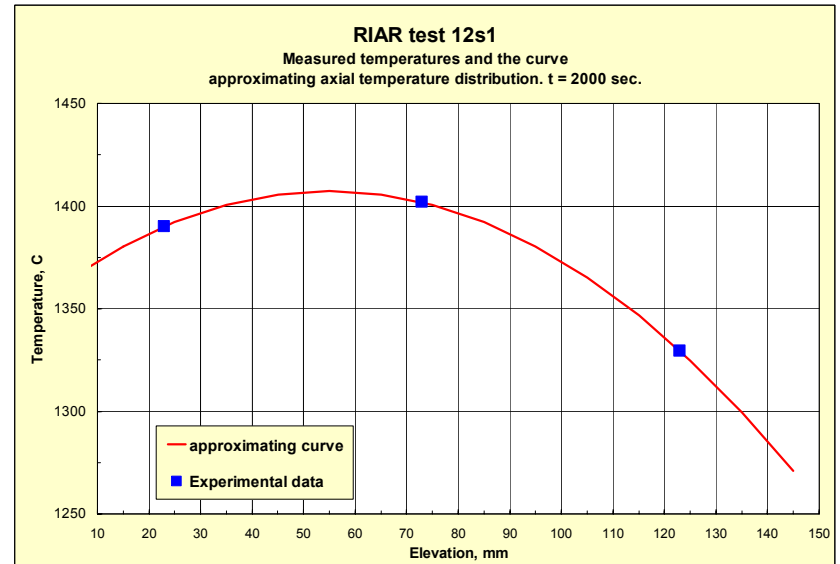
Analysis of the RIAR tests

- Temperature evolution of the sample is determined by its heat exchange with the operating channel internal surface, i.e. in calculations temperature of the operating channel internal surface represents the boundary condition for the heat exchange problem
- Temperature of the operating channel was adjusted to reproduce correctly the experimentally measured temperatures of the sample outer surface and of the fuel pellet centre (iteration procedure)
- Before using, the experimentally measured temperatures should be smoothed
- In the quench stage the operating channel temperature is fixed equal to steam temperature

Processing of the experimental data

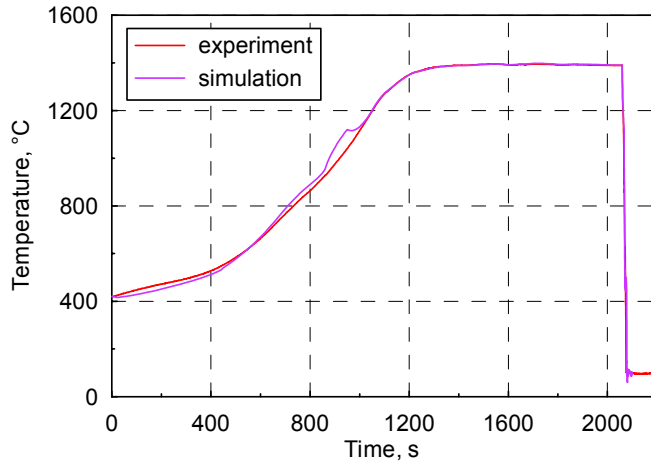


Smoothing of the experimental temperature curves (upper TC data)

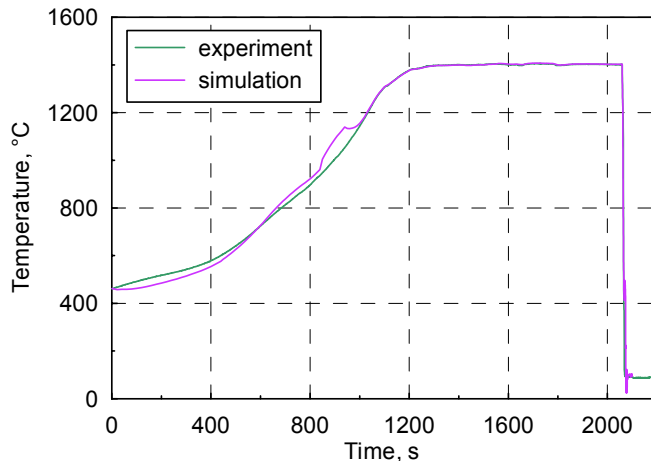
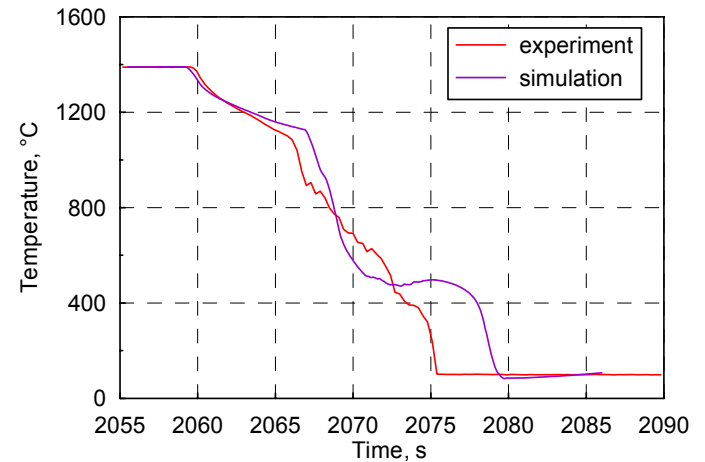


Extrapolation of the axial temperature distribution on the basis of the test data

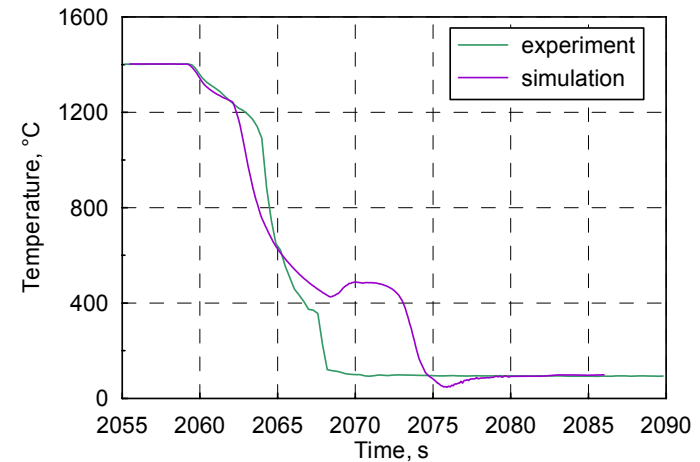
Simulation of the temperature evolution in the RIAR test #12



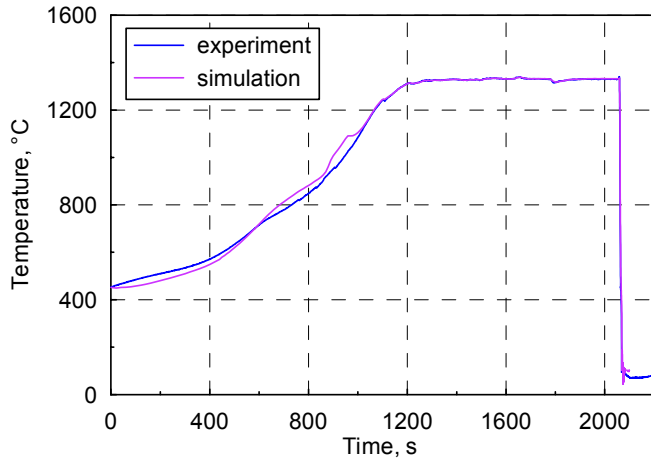
upper TC location



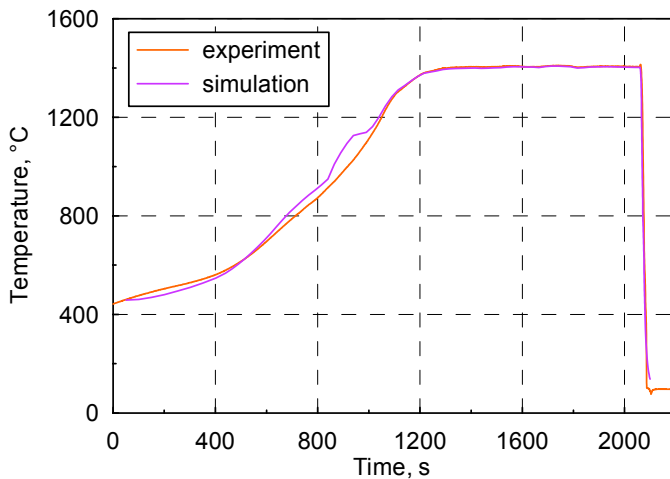
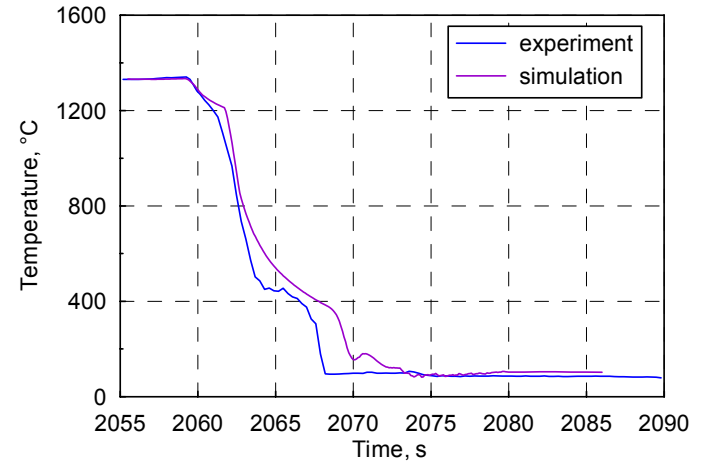
central TC location



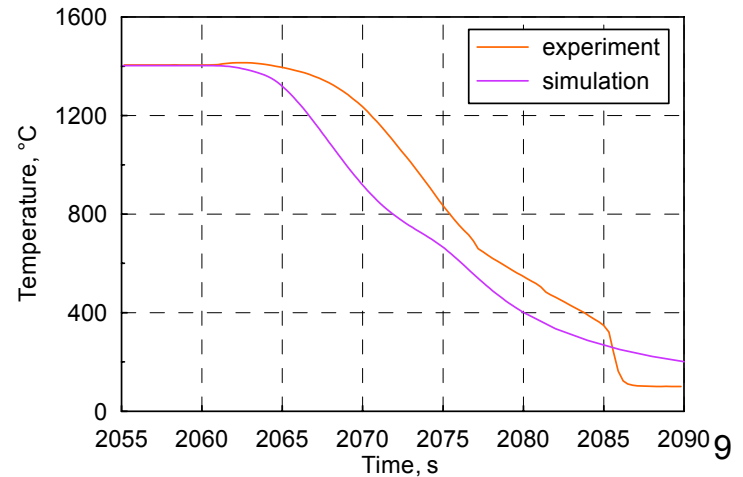
Simulation of the temperature evolution in the RIAR test #12



lower TC
location



pellet
central TC
location



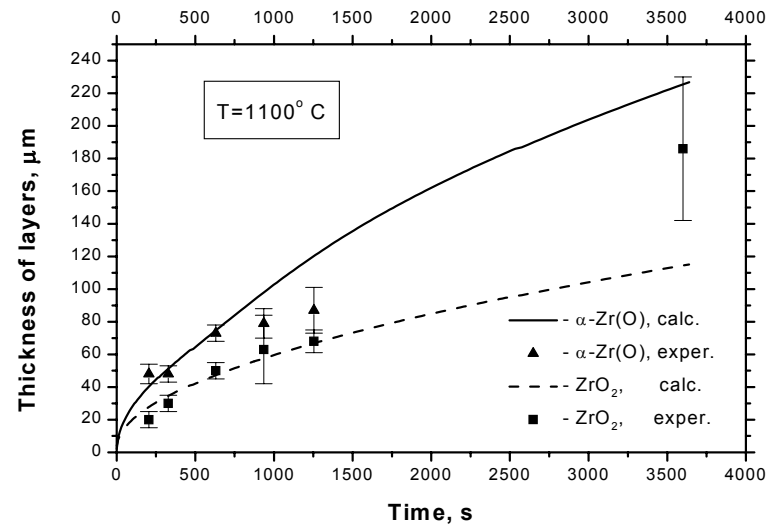
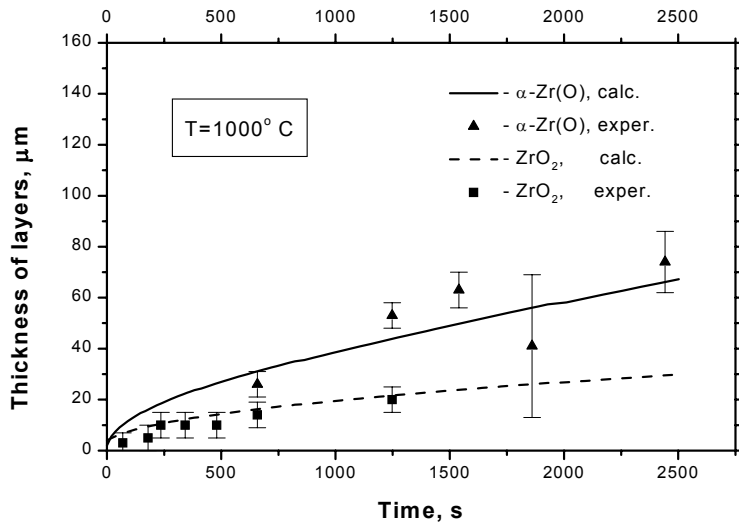
Oxidation module development (1/5)

Comparison of Zircaloy-4 and Zr-1%Nb oxidation kinetics [*]

- In general, the high temperature oxidation kinetics of non-irradiated VVER fuel rod cladding is very similar to that of non-irradiated Zircaloy-4 cladding.
- The main difference is the “breakaway” oxidation effect occurred in the VVER cladding in the range of 900 ÷ 1000 °C.
- The breakaway oxidation influences the mechanical deformation behaviour due to further embrittlement of the cladding. The most important factor is the hydrogen embrittlement.
- The hydrogen embrittlement occurs at very low temperature (less than 100 – 150 °C) and it does not affect oxidized fuel cladding fracture at high temperature (>1000 °C).
- New technologies of Nb-bearing alloys manufacturing can decrease or prevent this phenomenon.

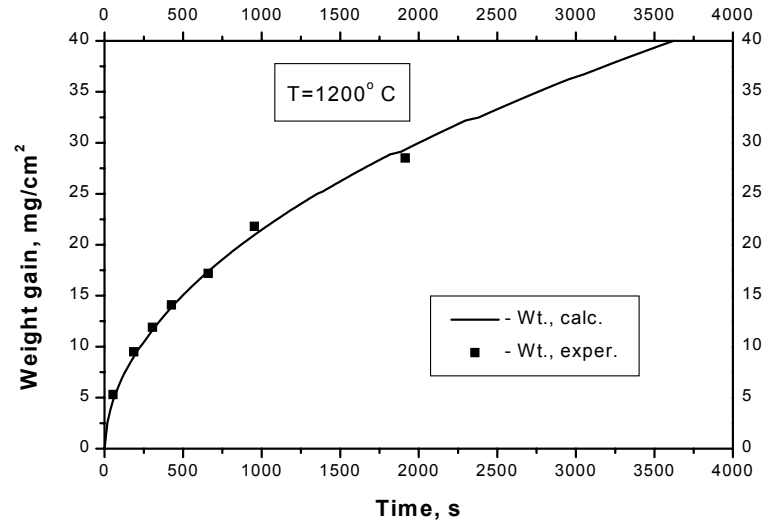
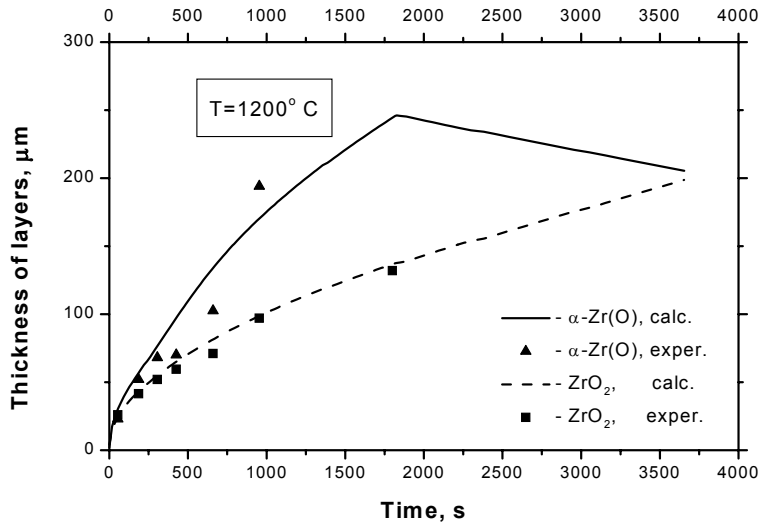
[*] L. Yegorova, K. Lioutov, N. Jouravkova, A. Konobeev, V. Smirnov, V. Chesanov, A. Goryachev Experimental Study of Embrittlement of Zr-1%Nb VVER Cladding under LOCA-Relevant Conditions. NUREG/IA-0211, IRSN 2005-194, NSI RRC KI 3188. March 2005.

Analysis of RIAR separate-effect tests



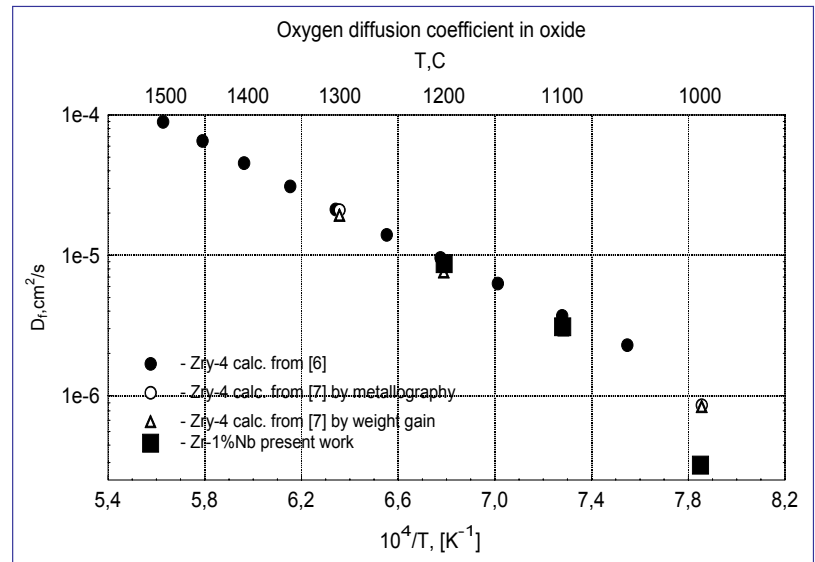
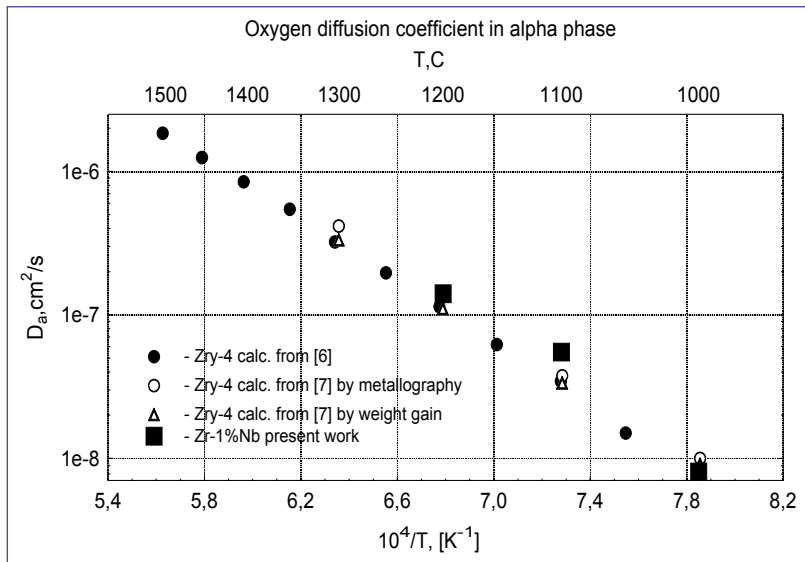
Simulation of the α -Zr and ZrO₂ layers growth kinetics in the RIAR isothermal tests on Zr-1%Nb cladding oxidation in steam at 1000 and 1100°C

Analysis of RIAR separate-effect tests



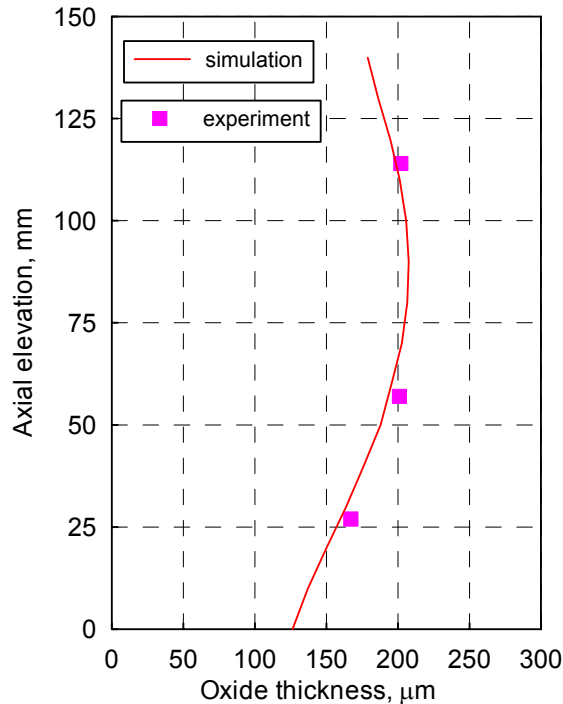
Simulation of the $\alpha\text{-Zr}$ and ZrO_2 layers growth and weight gain kinetics in the RIAR isothermal tests on Zr-1%Nb cladding oxidation in steam at 1200°C

Analysis of RIAR separate-effect tests

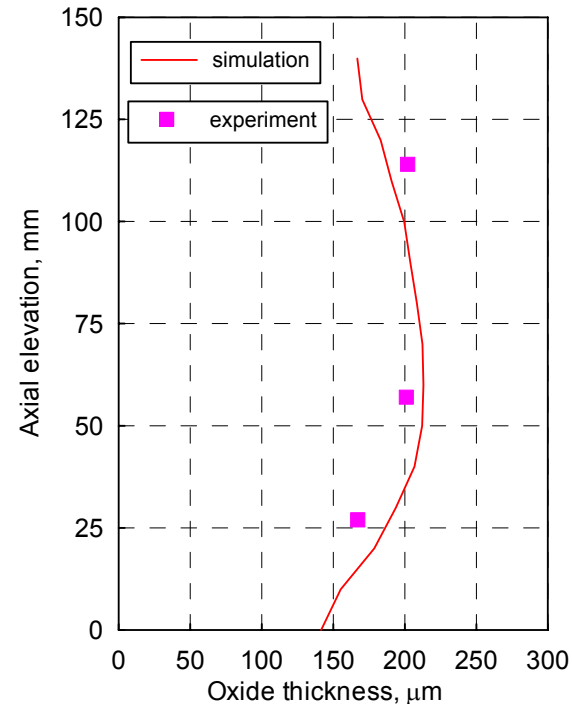


Calculated oxygen diffusion coefficients in α -Zr and ZrO_2 phases of Zr-1%Nb and Zry cladding materials

Zr-1%Nb cladding oxidation in the RIAR quench tests



Test #12



Test #14

Comparison of experimental and simulated axial profile of ZrO_2 layer thickness

Mechanical deformation module development (1/6)

Comparison of Zry-4 and Zr-1%Nb deformation behaviour [*]

- The main difference in the deformation behaviour of the non-oxidized Zry-4 and Zr-1%Nb cladding materials occurs at temperature below ~ 1000 °C and depends on the phase content, which is a temperature dependant function. At higher temperatures the difference in the deformation behaviour is negligible.
- At temperatures above the ($\alpha \rightarrow \beta$) phase transition in the non-oxidized Zr-1%Nb alloy (> 900 °C) mechanical properties do not depend on the strain rate.
- Mechanical properties of the non-oxidized Zr-1%Nb alloy are affected by irradiation at low temperature. At high temperatures (> 600 °C) mechanical properties of irradiated and non-irradiated claddings are similar owing to the effect of radiation damage annealing (see Fig. 1). Similar results were obtained for Zry-4.

[*] L. Yegorova, K. Lioutov at al. Experimental Study of Narrow Pulse Effects on the Behavior of High Burnup Fuel Rods with Zr-1%Nb Cladding and UO₂ Fuel (VVER Type) under Reactivity-Initiated Accident Conditions. NUREG/IA-0213, Vol. 1-2, IRSN/DPAM 2005-275, NSI RRC KI 3230. May 2006.

Mechanical deformation module development (2/6)

Dependence of ultimate strength (MPa) of irradiated and unirradiated Zr-1%Nb cladding on temperature (K)

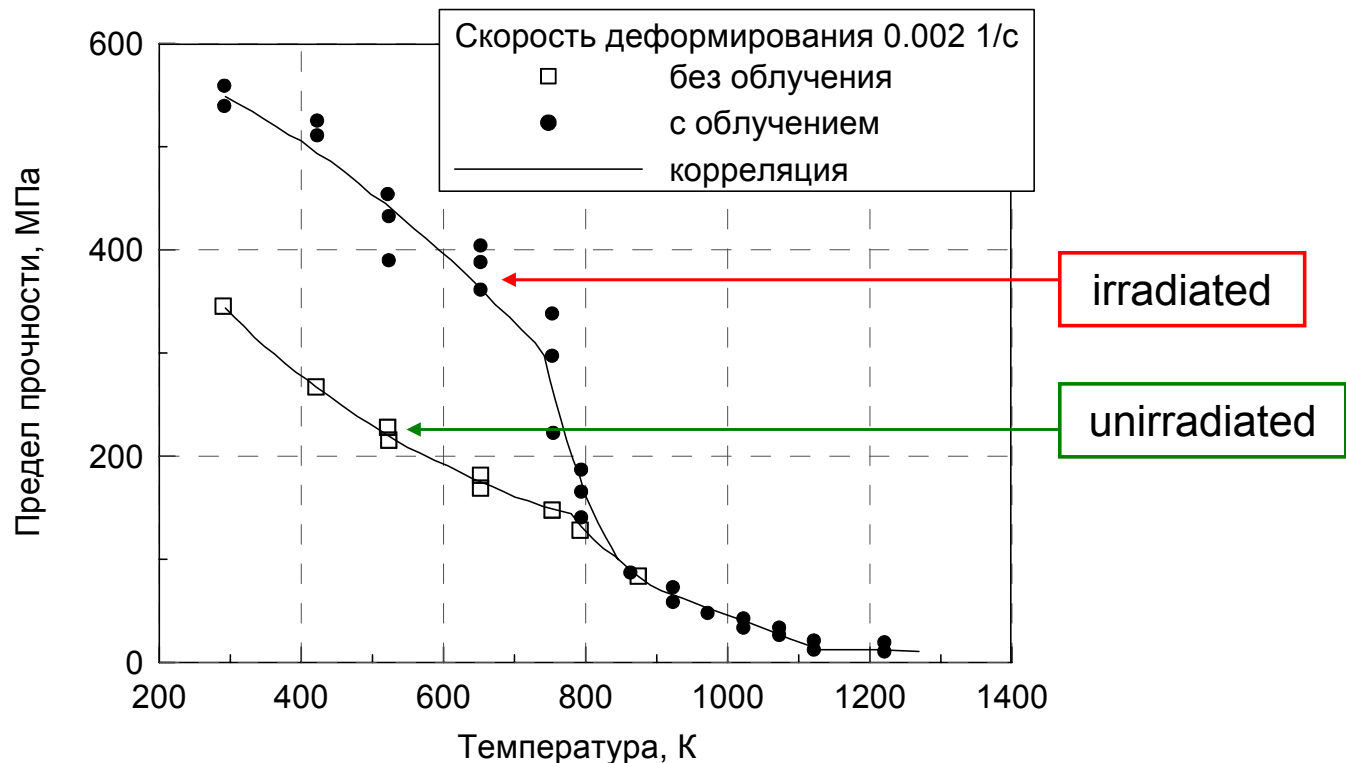


Figure 1

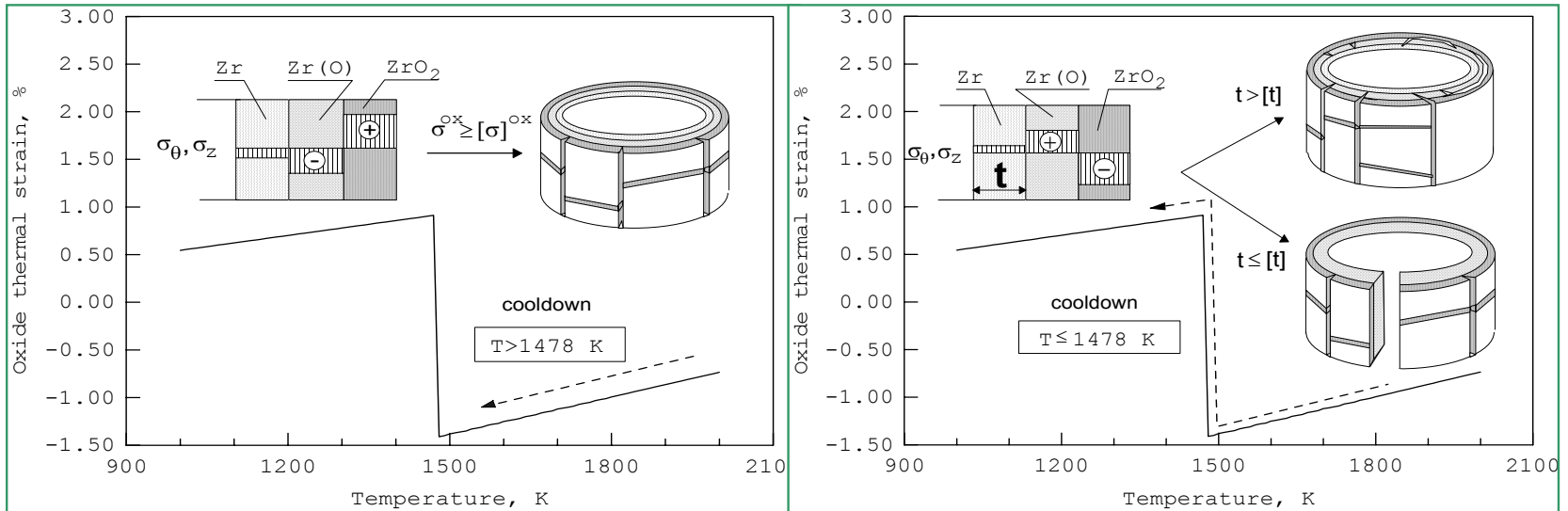
Mechanical deformation module development (3/6)

Zr-1%Nb mechanical properties

- Taking into account similar deformation behaviour of Zry-4 and Zr-1%Nb cladding at high temperatures and lacking data for mechanical properties of the α -Zr(O) and ZrO_2 phases of Zr-1%Nb, the corresponding mechanical properties of Zry-4 are used in calculations.
- At low temperatures (< 1000 °C) and in the case of an inert atmosphere (i.e. when oxidation is small) the mechanical properties of the metal Zr-1%Nb phase are used.
- For these reasons, formation of the net of through wall cracks under quench conditions in the case of the oxidized VVER cladding are foreseen owing to the same mechanism: tetragonal-to-monoclinic phase transition in the ZrO_2 scale which induces a strong increase of thermal strains in the ZrO_2 material.
- Therefore, the main peculiarities derived from the analysis of the results of FZK small scale quench test program are expected to be valid for the RIAR tests with non-irradiated VVER fuel rod simulators.

Mechanical deformation module development (4/6)

Through-wall crack formation mechanism

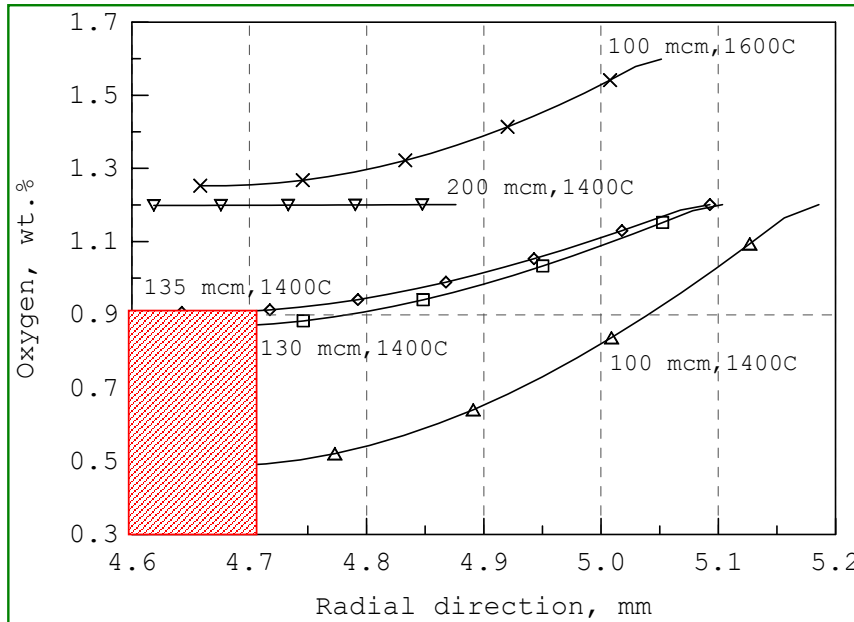


Cladding deformation behaviour on cool-down above and below oxide phase-transition temperature (tetragonal → monoclinic)

Mechanical deformation module development (5/6)

Chung-Kassner criterion:

determines possibility of oxidised rods to withstand thermal shock



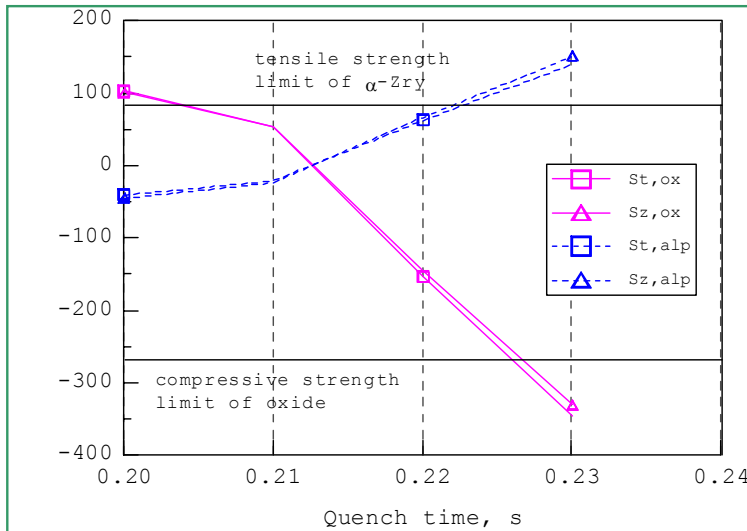
calculated thickness of cladding with oxygen content < 0.9 wt.% should be greater than 0.1 mm

SVECHA simulations for various quenching temperatures and preoxidation

Mechanical deformation module development (6/6)

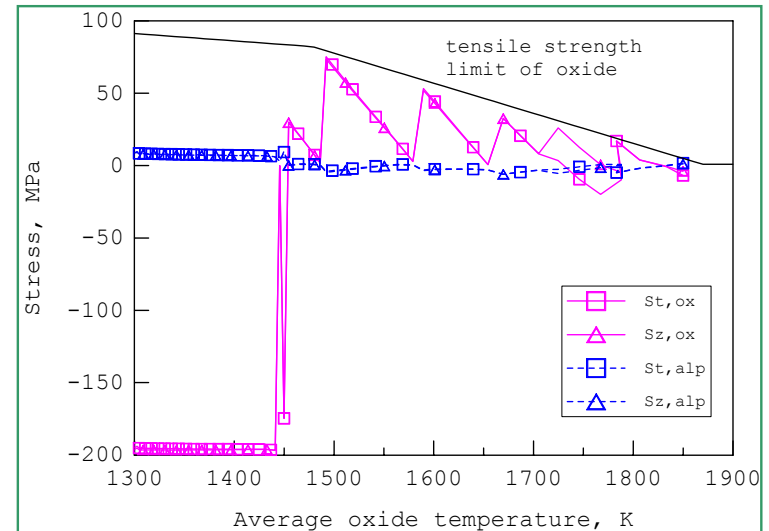
Conservatism of the Chung-Kassner criterion:

T=1200 °C, pre-oxidation 300 μm



Tensile stresses in α -phase attain critical value \Rightarrow **onset** of through-wall cracks

T=1600 °C, pre-oxidation 300 μm

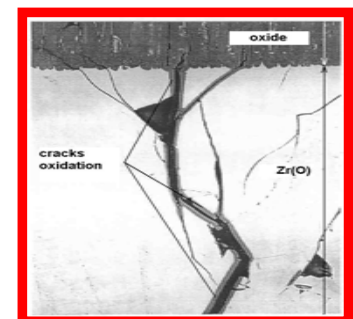
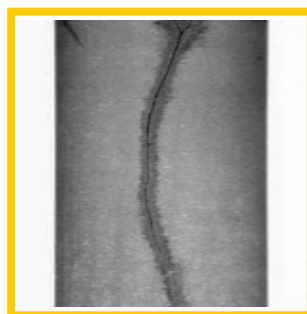
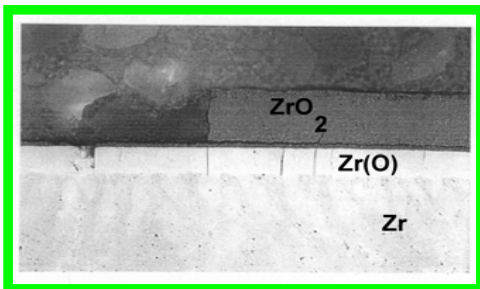


Tensile stresses in α -layer are small due to continuous oxide microcracking during pre-oxidation \Rightarrow **no** through-wall cracks

Conclusion: despite the criterion is satisfied in both cases, S/Q thermo-mechanical calculations predict different behaviour (no through-wall cracks in the case of high temperatures and thick oxides)

Crack formation in FZK Quench Rig Tests

Oxide thickness, μm	Initial temperature, $^{\circ}\text{C}$		
	1200	1400	1600
0	Cracks in Zr(O).	Cracks in Zr(O). Local spalling (water).	Cracks in Zr(O). Local spalling (water).
100÷150	No through wall cracks. Cracks in Zr(O). Local spalling (water).	No through wall cracks. Cracks in Zr(O). Local spalling (water).	A few through wall cracks. No cracks and inner surface oxidation.
150÷350	Net of through wall cracks. Cracks and inner surface oxidation.	Net of through wall cracks. Cracks and inner surface oxidation.	A few through wall cracks. Crack surfaces partially oxidized.

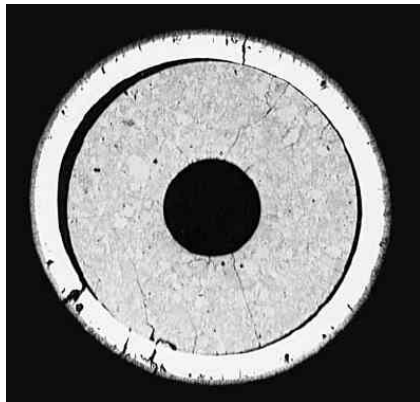


Simulation of Zr-1%Nb fresh fuel rod behaviour in RIAR tests

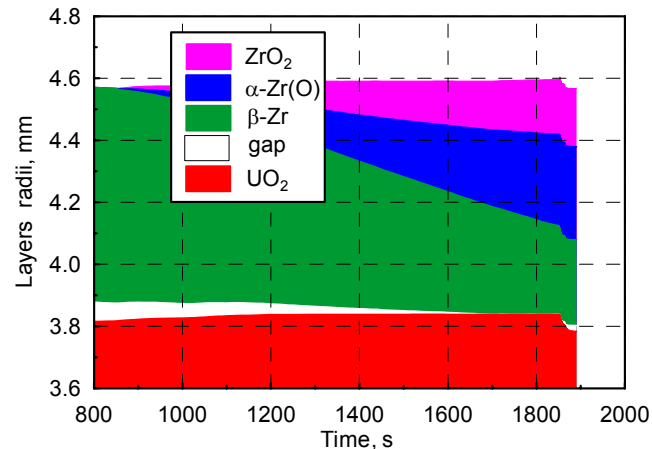
Test #12 observations:

- Gap disappearance and fuel-to-cladding interaction were observed in some areas of the cladding inner surface
- Sample had a net of through wall cracks in the centre, which were visualized by wetting with acetone.

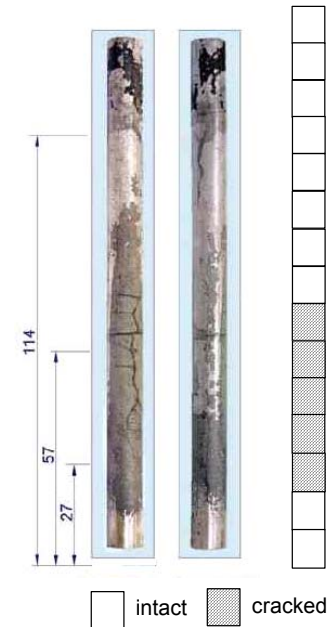
Both observations are correctly predicted by S/Q calculations



Cross section at elevation 57 mm.

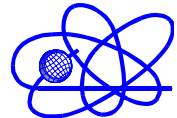


Evolution of the cladding layers at elevation 57 mm.



Comparison of experimental and calculated states of the quenched fuel rod simulator

Crack formation in RIAR Tests with Fresh Fuel



Group	Test №	Initial temp., °C	Oxide thickness, μm	Metal thickness, μm	Elevation, mm	Cladding state after the test
I	12	1400	201	574	57	Net of through wall cracks in the central part of the rod.
	14	1600	216	571	77	One circumferential through wall crack.
	15	1600	-	-	-	One circumferential through wall crack. Breakdown into 2 parts.
II	21	1400	13	703	127	Intact. No through wall cracks.
	22	1400	-	-	-	
	23	1400	155	605	27	
	24	1400	127	618	127	
III	25	1700	251	530	80	Breakdown into 3 parts due to heater failure.
	26	1700	144	614	77	Intact.
	27	1700	83	656	141	Intact after test. Broken during handling.
	28	1700	124	619	77	Intact.
IV	31	1700	140	636	75	Broken during handling. Net of the cracks is observed for wetted with water sample.
	32	1700	288	508	77	Breakdown into 2 parts. Net of the cracks is observed for wetted with water sample.
	35	1400	99	636	77	Intact. No through wall cracks.

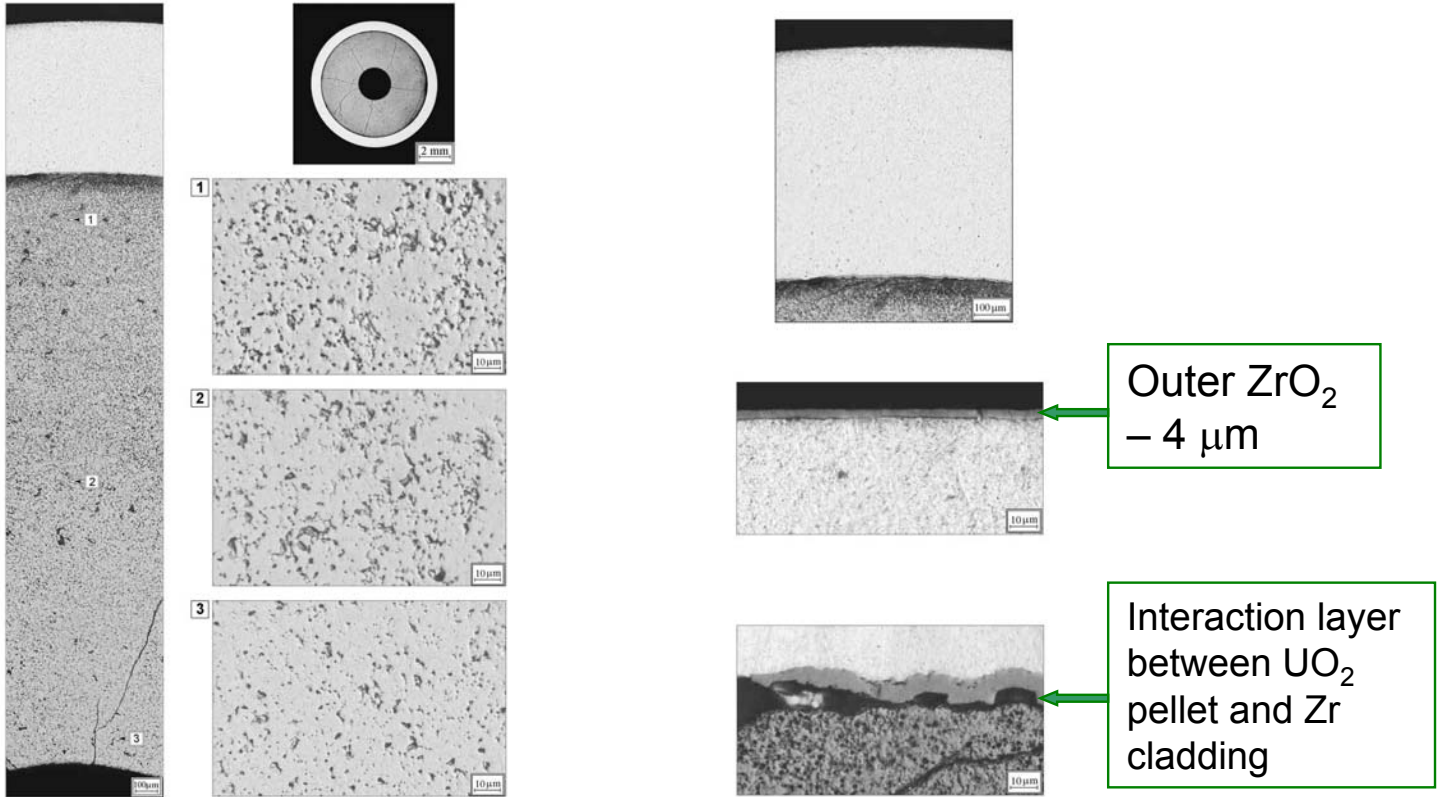
Conclusions

on the modelling of RIAR tests with VVER fresh fuel rod simulators

- SVECHA/QUENCH (S/Q) code initially designed for simulation of PWR fuel rods simulators in FZK quench tests, was successfully modified and adapted to simulate behaviour of the VVER fuel rod simulators in the RIAR quenching tests.
- The modified S/Q code well predicts the temperature evolution during quenching, the maximum extent of Zr-1%Nb cladding oxidation and the final mechanical state of the oxidized Zr-1%Nb cladding in the tests with fresh uranium fuel rods.
- The S/Q correctly predicts similar behaviour of unirradiated fuel rod simulators in FZK (with Zry-4 cladding) and RIAR (with Zr-1%Nb cladding) tests under similar test conditions (i.e. similar test matrices)
- The Chung-Kassner criterion for determination of possibility of oxidised rods to withstand thermal shock was shown to be reliable, however, rather conservative. Application of the criterion to quenching tests requires mechanistic calculation of oxygen distribution in the cladding layers (from diffusion equations).

**Part 2. Application of the SVECHA/QUENCH
and MFPR codes to simulation of
the RIAR quenching tests
with VVER irradiated fuel rods**

Initial state of irradiated fuel rods



53.5 MWd/kg U

FZK tests on chemical interactions of Zry-4 tubing with UO_2 fuel

Test objective [*]:

- An investigation of the combined H_2O /Zry and UO_2 chemical interactions as a function of time and temperature.

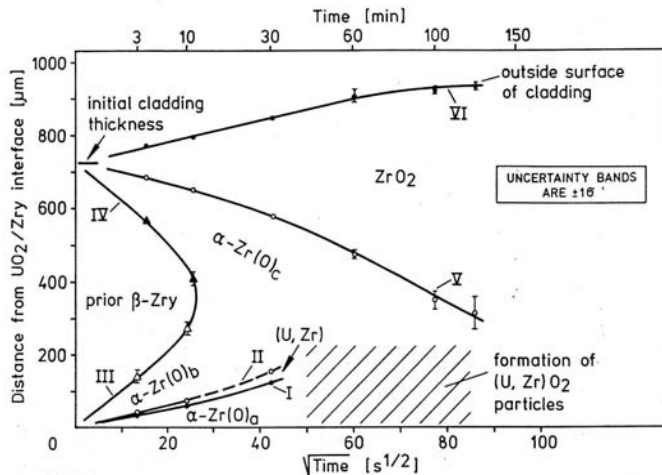
Test procedure:

- The out-of-pile experiments were performed in Ar + 25 vol % O_2 gas mixture.
- The short cladding tube specimens filled with stoichiometric high-density UO_2 pellets were used.
- The specimen was contained in a high-pressure vessel of the test rig MONA and inductively heated, with the cladding acting as susceptor.
- Cladding temperature was continuously monitored by a pyrometer and measured at three different axial elevations and orientations of the specimen.
- The maximum cladding temperature was 2000°C and the exposure times varied between 1 and 150 min.
- The external pressure was about 40 bar.

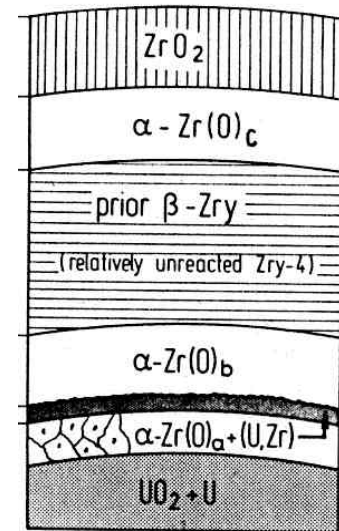
[*] P. Hofmann, Chemical Interactions of Zircaloy-4 Tubing with UO_2 Fuel and Oxygen at Temperatures between 900 and 2000°C (Experiments and PECLOX Code), Part I: Experimental Results, Kernforschungszentrum Karlsruhe, KfK 4422, CNEA NT-36/87, Oktober 1988

Main results of FZK tests on chemical interactions of Zry-4 tubing with UO_2 fuel

- In the case of solid contact between pellet and cladding, Zry reduces UO_2 to form oxygen stabilized $\alpha\text{-Zr(O)}$ (internal) and (U,Zr)-alloy.
- The external cladding interaction with oxygen or steam results in the formation of $\alpha\text{-Zr(O)}$ (external) and ZrO_2 .
- The internal and external $\alpha\text{-Zr(O)}$ layers of the cladding grow \approx with the same rates. Initially the reaction-layer growth obeys a parabolic rate law. After the disappearance of β -phase, the cladding tube is completely embrittled and no more mechanically stable.



T = 1300 °C
Pext. = 40 bar, atmosphere = Ar + 25 vol.% O₂

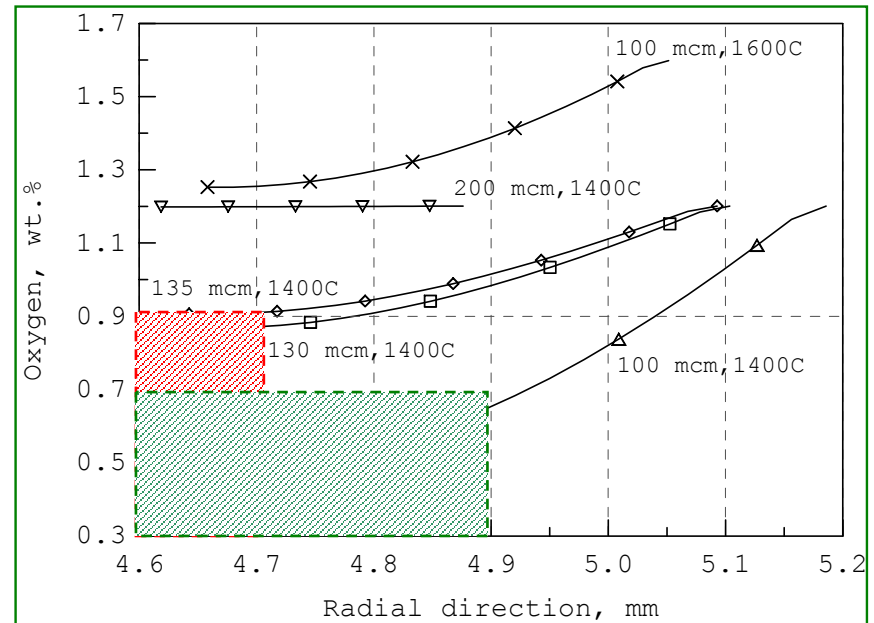


Cladding Embrittlement Criteria (1/2)

- Capability to withstand the different loading modes depends on thickness of and oxygen distribution in β -Zr layer:

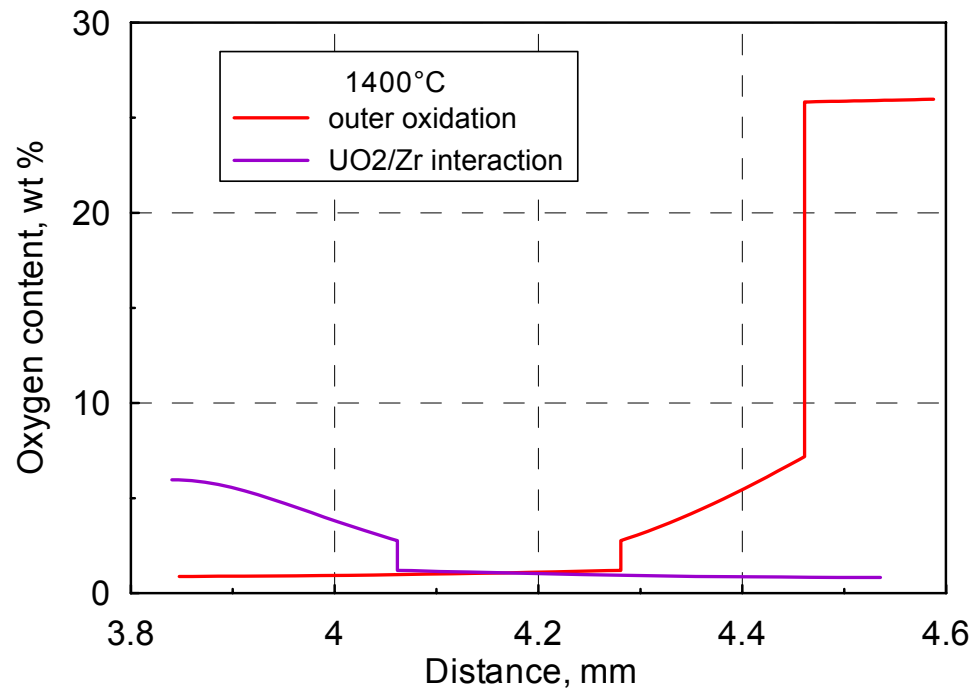
Chung-Kassner criteria

1. **Capability to withstand thermal shock during LOCA reflooding:** calculated thickness of the cladding with $\leq 0.9 \text{ wt } \%$ oxygen should be greater than **0.1 mm**.
2. **Capability to withstand fuel handling, transport and storage:** calculated thickness of the cladding with $\leq 0.7 \text{ wt } \%$ oxygen should be greater than **0.3 mm**.



Cladding Embrittlement Criteria (2/2)

- Formation of Zr(O) layer at the inner surface of the cladding as a result of UO_2/Zr chemical interaction leads to cladding embrittlement even in the absence of oxidation atmosphere at the outer cladding surface.



Maximum extent of oxidation and fuel-pellet interaction to remain intact under thermal shock.

Main results of RIAR quenching tests with irradiated VVER fuel rod simulators

Tests without pre-oxidation

Test #		40	46	45	47	37	49
Atmosphere		Ar	Ar	Ar	Ar	Ar	Ar
Exposure at 1400 °C	s	0	240	240	240	240	240
Quenching temperature	°C	1400	1400	1600	1600	1700	1700
State of the cladding after the test *		1	1	2	1	2	2
Thickness of outer ZrO ₂	μm	9	8	14	17	19	-
Thickness of outer α-Zr(O)	μm	23	26	41	43	31	-
Thickness of inner α-Zr(O)	μm	90	113	75	191	337	-
Total thickness of metal layer	μm	674	696	696	676	648	-
Total hydrogen release	mg	13.6	9.4	20.6	20.5	14.7	26

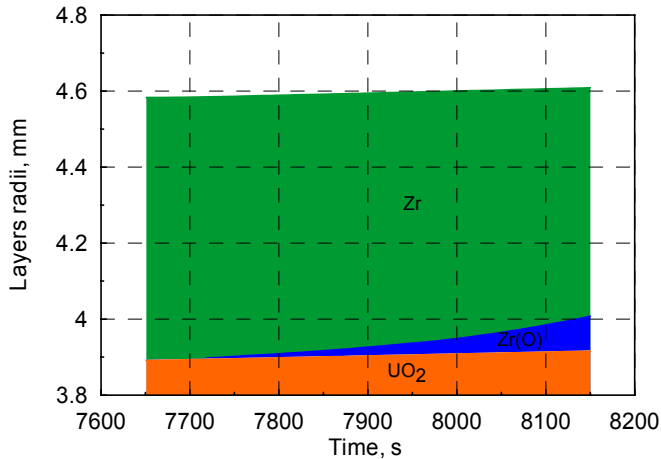
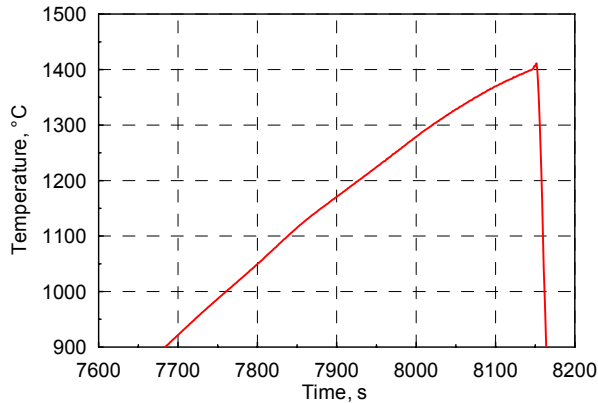
* Cladding state after the test:

1 – intact after the test

2 – broken after the test

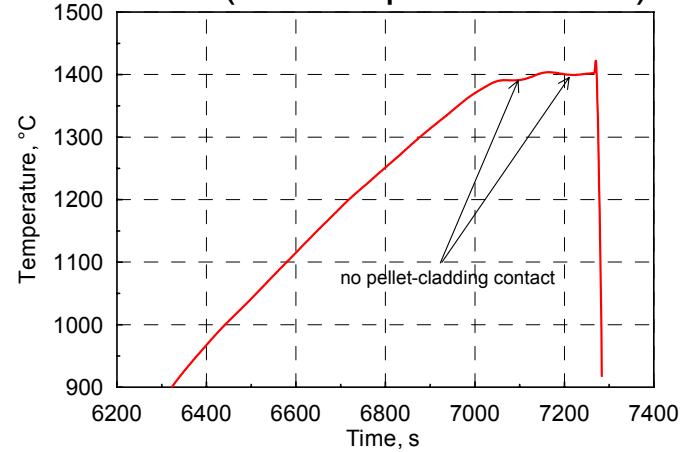
Simulation of RIAR quench tests with irradiated fuel rods without pre-oxidation (1/3)

Test #40 (without pre-oxidation)



Inner α -Zr(O) layer growth

Test #46 (without pre-oxidation)



Temperature evolution

Test	40	46
Measured	90	113
Simulated	92	110

Thickness of inner α -Zr(O) layer, μm

Simulation of RIAR quench tests with irradiated fuel rods without pre-oxidation (2/3)

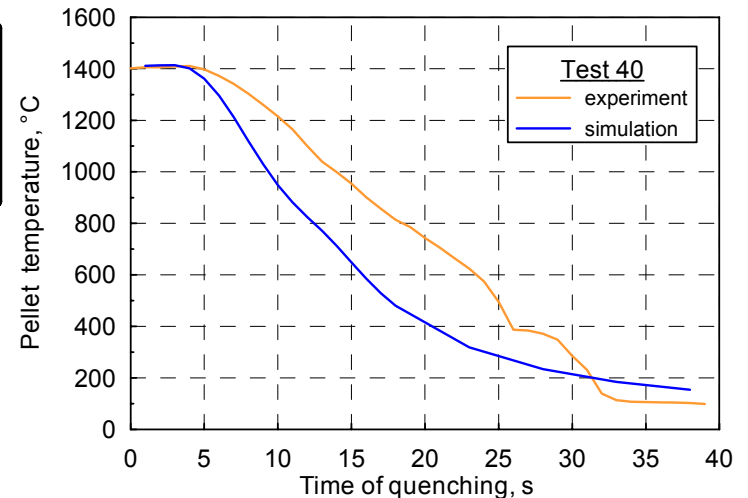
Test #40

Layers thickness, μm

Elevation n, mm	Measurement		Simulation	
	Outer Zr(O)	Outer ZrO ₂	Outer Zr(O)	Outer ZrO ₂
127	-	-	18	13
77	23	9	19	14
27	5	6	11	8

Hydrogen release, mg





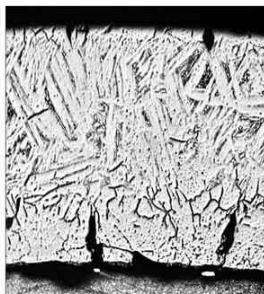

Measurement		Simulation	
Total	Quench	Total	Quench
13.6	13.6	12.6	12.6



Comparison of the calculated and measured temperatures in the pellet centre at elevation 77 mm during quenching.

Simulation of RIAR quench tests with irradiated fuel rods without pre-oxidation (3/3)

Application of Chung-Kassner criterions for capability to **withstand handling (± 1)** or to **withstand thermal shock (± 2)**

40	46	45	47	37	49
				<p>127 MM</p> 	
+1/+2	+1/+2	-1/+2	-1/+2	-1/+2	-1/+2

Conclusion: capability of irradiated fuel rods without additional pre-oxidation to withstand thermal shock under quenching and failure during post-test handling are correctly predicted by S/Q using the Chung-Kassner criterions

Main results of RIAR quenching tests with irradiated VVER fuel rod simulators

Tests with pre-oxidation

Test #		36	39	51	53	54	55	56
Atmosphere		Steam +Ar	Steam+ Ar	Steam +Ar	Steam +Ar	Steam +Ar	Steam +Ar	Steam +Ar
Exposure at 1400 °C	s	240	240	240	240	1050	1100	1100
Quenching temperature	°C	1400	1400	1600	1700	1400	1400	1600
Cladding state after the test *		1	2	2	3	3	3	3
Hydrogen release during quench phase	mg	6.7	7.0	22.5	39.2	14.0	17.2	43.2
Total hydrogen release	mg	164	173	210	252	386	497	686

* Cladding state after the test:

1 – intact after the test

2 – broken after the test

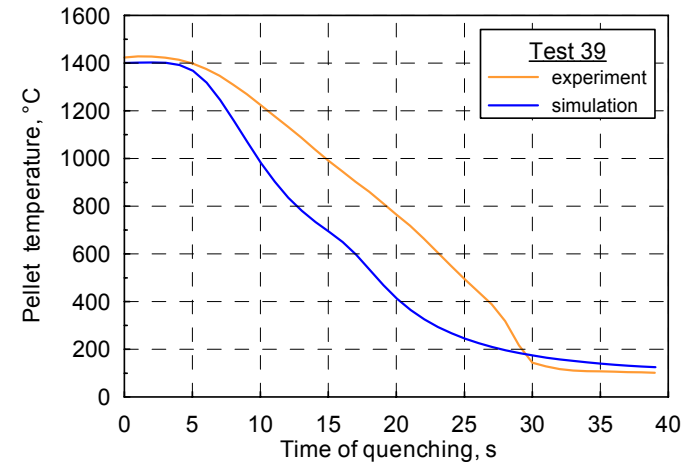
3 – broken during the test

Simulation of RIAR quench tests with irradiated fuel rods with pre-oxidation (1/3)

Test # 39

Layers thickness, μm

Elevation, mm	Measurement		Simulation	
	Outer/Inner Zr(O)	Outer ZrO ₂	Outer/Inner Zr(O)	Outer ZrO ₂
127	151/137	101	163/163	106
77	126/122	92	160/160	108
27	58/56	39	162/162	110



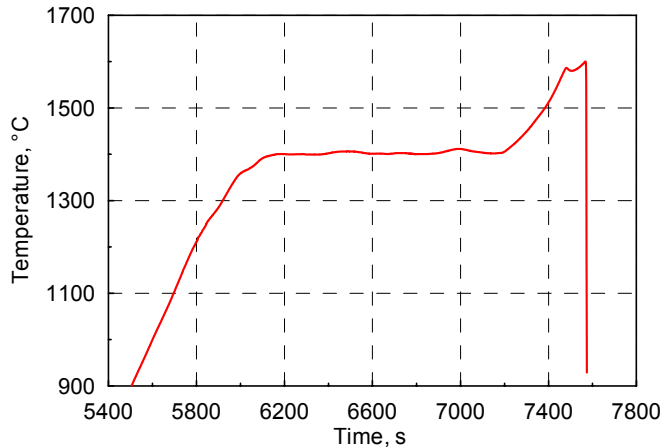
Hydrogen release, mg

Measurement		Simulation	
Total	Quench	Total	Quench
173.2	7.1	126.9	0.2

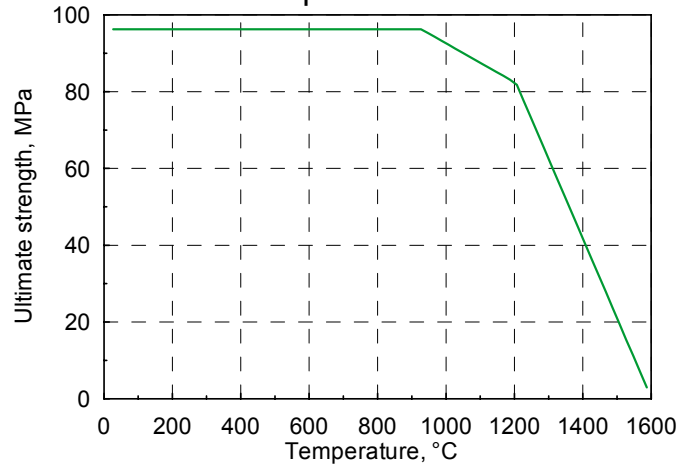
Comparison of the calculated and measured temperatures in the pellet centre at elevation 77 mm during quenching.

Simulation of RIAR quench tests with irradiated fuel rods with pre-oxidation (2/3)

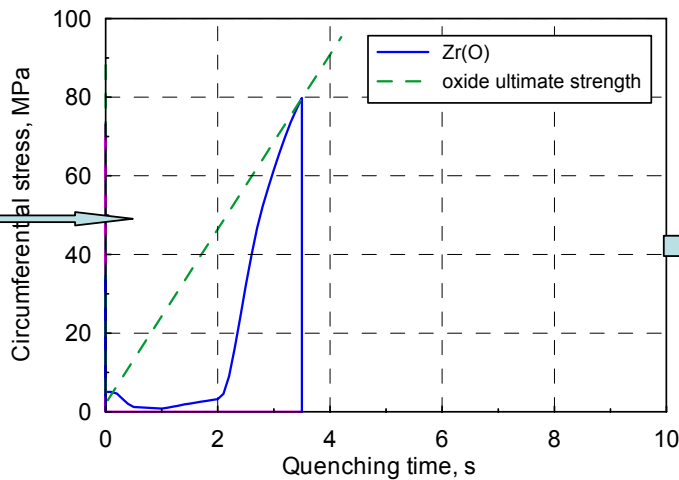
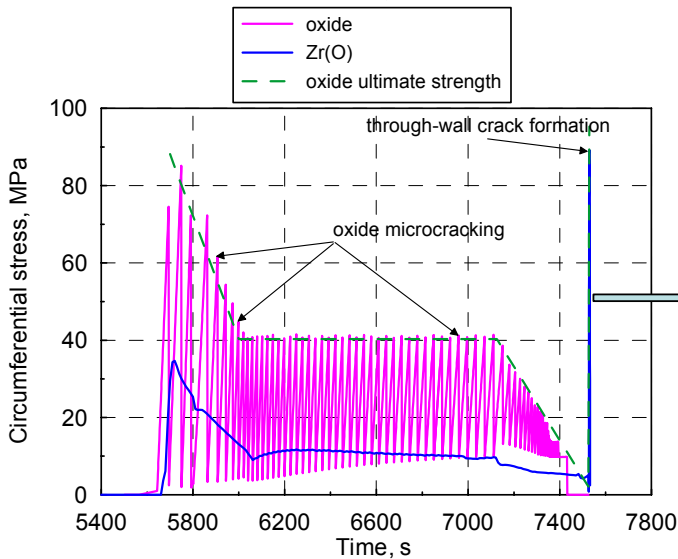
Test # 56



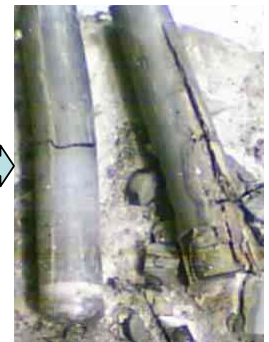
Oxide ultimate strength versus temperature



A similar dependence for α -Zr is assumed (lacking exact data)


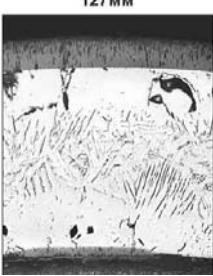
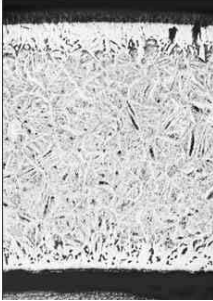




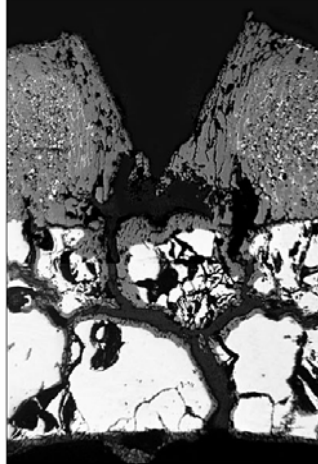


Post-test view



Simulation of RIAR quench tests with irradiated fuel rods with pre-oxidation (3/3)

Application of Chung-Kassner criterion for capability to **withstand handling (± 1)** or to **withstand thermal shock (± 2)**

36	39	51	53	55	54	56
	 					
-1/+2	-1/+2	-1/+2	-2	-2	-2	-2

Conclusion: conservatism of Chung-Kassner criterion is strongly reduced, owing to strong pellet-cladding mechanical interaction during quenching

Conclusions on simulation of RIAR quench tests with irradiated fuel rods

- Analysis of the results of RIAR quench tests with irradiated fuel rods show that for high burnups the gap between the fuel pellet and the cladding completely disappears at operation temperatures.
- Simulations by the SVECHA/QUENCH (S/Q) code confirm test observations of a good pellet-cladding contact in the case of further temperature escalation.
- On the base of the FZK separate-effect tests observations, S/Q predicts in this case chemical interactions of zirconium cladding with uranium dioxide that result in formation of internal α -Zr(O) layer and embrittlement of the cladding.
- Thus, the fuel rod cladding embrittlement can take place even lacking external oxidation. Basing on the Chung-Kassner criterions, for such test conditions the S/Q code correctly predicts fuel rods failure during handling after the test.
- Additional external cladding oxidation considerably enhances cladding embrittlement. Basing on the Chung-Kassner criterions, for such test conditions the S/Q code predicts fuel rods failure during quenching.
- Thermo-mechanical consideration of the S/Q code confirms this conclusion and demonstrates formation of through-wall cracks in fuel rods during quenching, owing to high tensile stresses induced by the fuel pellet in both α -Zr(O) and oxide layers of the cladding. This essentially reduces conservatism of the Chung-Kassner criterion (in comparison with the case of unirradiated fuel rods).

Development

- IBRAE-IRSN co-operation (1995-2007)

Mechanistic description

of FP behavior in irradiated UO_2 with intact geometry:

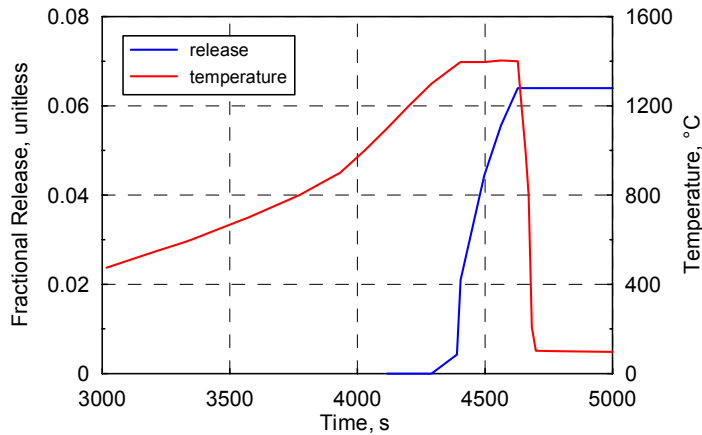
- In irradiation regime : steady state and transients
- In annealing regime : steady state and transients
- In accidental conditions: LOCA, severe accidents

Three groups of models (tightly coupled)

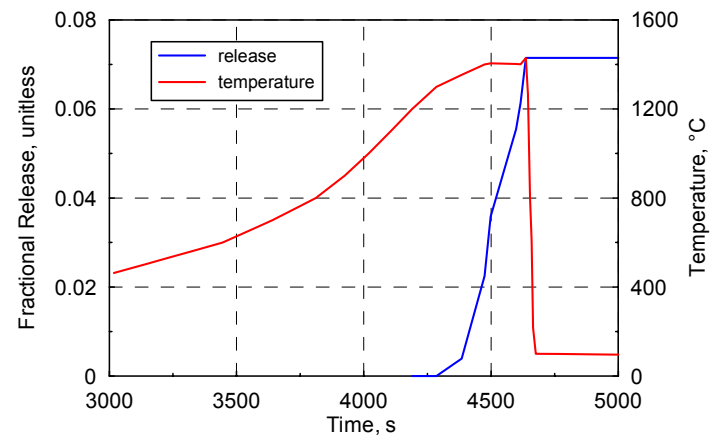
- Fission gases and gas bubbles
- Chemically active elements
 - Fuel oxidation/vaporisation in steam/hydrogen/air atmospheres
- Evolution of fuel microscopic defect structure

MFPR code simulation of the RIAR quench tests with irradiated fuel (1/2)

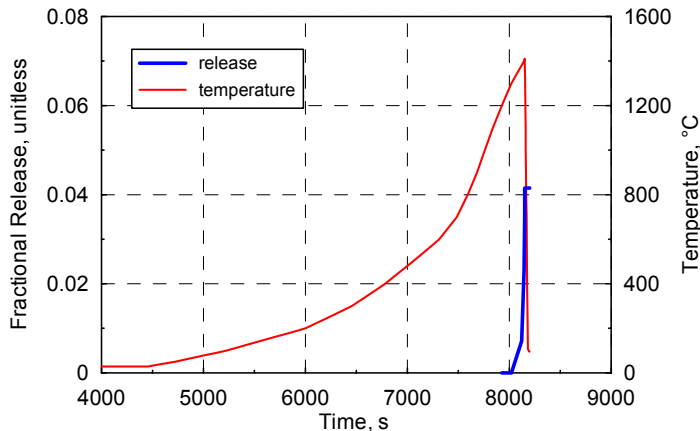
Test # 36 (53.5 MWd/kgU)



Test # 39 (65 MWd/kgU)



Test # 40 (65 MWd/kgU)



Cs fractional release, %

Test #	Experiment	MFPR calculation
36	4.6	7.5
39	8.8	7.7
40	2.6	4.3

MFPR code simulation of the RIAR quench tests with irradiated fuel (2/2)

Calculated and measured fractional releases of ^{137}Cs

Test #	Max. annealing temperature	^{137}Cs fractional release (%)	
		Experiment	MFPR calculation
36	1673 K	5	7.5
39	1673 K	9	7.7
40	1673 K	3	4.3
45	1873 K	14	22.0
47	1873 K	8	21.9

Calculated and measured fractional releases of ^{85}Kr

Test #	Max. annealing temperature	^{85}Kr fractional release (%)	
		Experiment	MFPR calculation
36	1673 K	16.5 ± 4.0	2.9
39	1673 K	8.0 ± 4.5	2.6
40	1673 K	4.0 ± 4.5	2.3
45	1873 K	4.9 ± 4.5	3.3
47	1873 K	4.2 ± 4.5	3.4

General Conclusions

on the modelling of RIAR tests (1/2)

- SVECHA/QUENCH (S/Q) and MFPR codes were adapted to simulate thermo-mechanical behaviour and failure of the VVER fuel rod simulators in the RIAR quench tests.
- The modified S/Q code well predicts the temperature evolution during quenching, the maximum extent of Zr-1%Nb cladding oxidation and the final mechanical state of the oxidized Zr-1%Nb cladding in the tests with fresh uranium fuel rods, demonstrating similarity with results of the FZK quench tests (with Zry-4 cladding).
- The S/Q code was extended for simulation of the RIAR quench tests with irradiated fuel rods. Analysis and simulations of the tests show that the gap collapse and chemical interactions between the burned fuel pellets and the cladding due to swelling lead to additional embrittlement of the cladding.
- The S/Q code reasonably predicts enhanced failure of the irradiated rods during the test or during handling after the test, owing to enhanced embrittlement and high tensile stresses induced in the cladding layers by the pellet. This allows reducing conservatism of the Chung-Kassner criteria for irradiated fuel rods.

General Conclusions

on the modelling of RIAR tests (2/2)

- The MFPR code reasonably predicts Cs and Kr release in the RIAR quench tests with irradiated fuel rods. The release from fuel pellets is associated mainly with high-temperature annealing stage rather than with short-termed quenching stage.
- On this base, in accident scenarios with temperature escalation during quenching (observed in some FZK QUENCH bundle tests) one should expect an enhanced fission products (FP) release from fuel pellets into the pellet-cladding gap during heat-up stage and further release outside cladding owing to cladding failure during fast cool-down stage of quenching.
- Coupling of the S/Q and MFPR codes for more self-consistent consideration of these complicated phenomena is foreseen in the close future (beyond the ISTC Project).