A Mechanistic Approach to Develop the Secondary Hydriding Criteria

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Abstract. Reliable criteria of secondary hydriding failures are important to assure safe operation of nuclear fuel in LWR power units. The present paper reviews available data on massive hydriding of zirconium claddings covering out-of-pile studies and in-pile tests in research reactors. Analyses of these experimental data give evidence that threshold conditions leading to the onset of massive hydriding are drastically changed under irradiation. The changes are caused mainly by irradiation damage of oxygen sublattice in ZrO_2 by fission fragments leaving the periphery of fuel pellets. The tests in research reactors provide a basis to develop a parametric dependency which relates the threshold of massive hydriding to composition of steam-hydrogen mixture, irradiation dose rate and temperature.

Key words: primary failure, massive hydriding, secondary fuel degradation, secondary failure, oxygen starvation, hydrogen-to-steam ratio, in-pile and out-of-pile experiments.

1. INTRODUCTION

Massive hydriding of zirconium cladding in defective LWR fuel rods may lead to severe secondary failures. Phenomenology of secondary hydriding is well known. Once a primary defect in cladding is formed, water enters the fuel rod and flashes into steam. Steam oxidation of both fuel and inner cladding surface can generate enough hydrogen to break down the protective properties of ZrO_2 and to cause excessive hydrogen pick-up. It is commonly accepted that 'enough' hydrogen means oxygen-starved conditions when hydrogen content in gas mixture is substantially higher than that of steam, [1].

To assure safe fuel operation at nuclear power units it is important to establish the reliable criteria of secondary failures to occur. One of the methods to build the criteria lies in making correlations with the available data on secondary hydriding in fuel of commercial reactors [2]. The advantages of this method arise from using the direct results of fuel operation. But detailed post-irradiation examinations (PIEs) of defective fuel assemblies in hot cells are rather scanty. By reason of limited PIE data, the universality of the established correlation criteria is questionable. Generally, application of correlations is not justified when fuel design or fuel operation regimes are changed.

The database for building the criteria of secondary failures can be widened due to out-of-pile experiments and tests in research reactors. Such experiments are performed under controlled conditions and give a detailed information on the final fuel/cladding state. The shortcomings of the model experiments are revealed when it is necessary to extend the obtained results to the actual fuel rod geometry and to actual irradiation conditions. For instance, the thresholds of secondary hydriding, as derived from the research-reactor experiments [3,4] with short fuel rodlets, are distinct from the criteria in Ref.[2] and come into conflict with findings of the out-of-pile tests [5-7]. A comprehensive approach to the problem should include development and application of numerical codes. In particular, the mechanistic codes are capable of calculating the changes in the criteria caused by fuel design innovations.

The present paper gives a review and analysis of the available data on massive hydriding of zirconium claddings. Physical factors are discussed which may be responsible for substantial differences between the in-pile criteria and the out-of-pile thresholds for the onset of massive hydriding. Processing of the test-reactor experiments [3,4] shows that under irradiation massive hydriding is initiated at significantly lower hydrogen-to-steam ratios than under conditions without irradiation. The IFA-631 test [3] was carried out in Halden research reactor (Norway). The experiments [4] were performed in R2 research reactor at Studsvik (Sweden). With these data it is possible to determine the hydrogen content in gas mixture inside the test fuel rodlets with a simulated primary failure. Conditions which led to formation of secondary hydrides can be evaluated as well. The data analysis is accomplished by deriving the parametric dependency of the hydriding threshold on hydrogen-to-steam ratio, irradiation dose rate and temperature.

2. ANALYSIS OF DATA ON THE THRESHOLDS OF MASSIVE HYDRIDING

Data of several studies concerning the thresholds of massive hydriding and secondary failures can be found in literature. At first sight, tests carried out with different techniques and under different conditions lead to conflicting results. Let us take a look what problems arise when different data are attempted to be brought into agreement.

Operation experience with secondary degradation of commercial BWR and PWR fuel (Zircaloy-2 and Zircaloy-4 claddings) has been summarized by D. Locke, [2]. The data summary is known as the Locke diagram (Fig.1). The key conclusion derived from the analysis by Locke is that defective fuel may be operated successfully at low heat rates. The higher is the heat rate the shorter is the time for development of secondary failure. For heat fluxes above certain value (about 1600-1700 kW/m^2) time to failure decreases drastically – to several hours. This actually constrains the range of heat rates for safe operation of fuel rods with a primary defect.

In the DEFEX tests [4] with short rodlets (see Section 4 for details) secondary defects formed rapidly – in a few days – at heat fluxes much lower than it follows from the Locke curve (line I in

Fig.1). Relying on the DEFEX data the failure boundary should be displaced into the range of lower heat fluxes (line II in Fig.1). Thus, the universality of the Locke curve is put in question.

In the out-of-pile experiments [5-7] it was found that massive hydriding occurs if some critical ratio of hydrogen to steam partial pressure in gas mixture is exceeded. Two types of BWR fuel claddings were studied: standard Zircaloy-2 and Zircaloy-2 + unalloyed Zr liner. Several tests were performed with crystal bar Zr specimens. Total pressure of steam-hydrogen mixture in the tests [5-7] varied from 1 to 70 atm. Regardless of specimen type and total pressure the threshold hydrogen-to-steam ratio, $\beta = P_{H_2}/P_{H_2O}$, leading to the onset of massive hydrogen pick-up was governed by identical dependence on temperature

$$\beta_{cr} \approx 6\cdot 10^{-16}\exp\left(26670/T\right). \qquad (1)$$

The value of β_{cr} varies from 10^5 at 300 °C to ~ 200 at 400 °C, Fig.2*a* (for more details in analysis of the tests [5-7] see Ref.[8]). These conditions of excessive hydrogen in steam-hydrogen mixture are commonly called "oxygen starvation".

It is believed that oxygen starvation in failed fuel rods may be achieved away from location of primary defect if mass transfer is limited, e.g. due to local gap blockage, [9]. In this case there may be no (or very low) removal of generated hydrogen and no additional supply of steam. Such conditions can be supposed for BWR and PWR but not for WWER fuel with a central hole in pellets. The central hole is effectively connected to gap by pellet cracks providing good ventilation along the full length of fuel stack. Calculations [8] demonstrate that hydrogen-to-steam pressure ratio in operating defective WWER rods is less than, or about, 1 (typical value $\beta \sim 0.1$) while the temperature of the inner cladding surface is below 400 °C. So, the threshold for massive hydriding, found in experiments [5-7], seems never to be reached. It is remarkable that discrepancy between the calculated β values and the out-of-pile criterion (1) is 3 to 6 orders of magnitude. Nevertheless, hot-cell examinations regularly reveal secondary hydriding in failed WWER fuel rods, [10]. The analysis in Sections 3, 4 will show that the out-of-pile criterion [5-7] is also in conflict with the in-pile tests [3,4].

According to Ref.[8] the differences between the in-pile criterion and the out-of-pile threshold for the onset of massive hydriding may be attributed to irradiation damage of the oxide film at cladding inner surface by fission fragments leaving the periphery of fuel pellets. Substantial increase in concentration of oxygen vacancies in ZrO_2 under irradiation by fission fragments leads to breakdown of the oxide protective properties at much lower hydrogen content in the gas phase in comparison with the out-of-pile conditions. The change in the threshold of massive hydriding may be estimated as the ratio of thermal (K_c) to irradiation-induced ($\alpha_c \dot{F}$) defect generation rate in the oxygen sublattice of ZrO_2, [8]:

$$\beta^{rad}/\beta^{therm} \approx K_c/\alpha_c \dot{F}\ . \qquad (2)$$

Here $\dot{F}$ is the fission rate (at pellet periphery), $\alpha_c \sim \phi V_{tr}$, where V_{tr} is the volume of the fission fragment track and factor $\phi \sim 0.1$ takes into account partial defect recombination within the track.

The shift of the massive hydriding threshold estimated according to Eq.(2) is shown in Fig.2*b*. If we "multiply" together the two dependencies shown in Fig.2*a* and Fig.2*b* we come to a conclusion that under irradiation conditions the threshold of massive hydriding is a weak function of temperature and about 10% of hydrogen in steam-hydrogen mixture is sufficient to initiate the excessive cladding hydriding. This finding is in agreement with composition of the gas phase calculated in Ref.[8] for WWER fuel rods with a primary defect. Also it explains why despite violation of the threshold criterion (1) massive hydriding and secondary failures still occur in defective WWER fuel.

The hypothesis on the irradiation-induced shift of the massive hydriding threshold and parameters of the model [8] may be verified using the tests [3,4] made in research reactors.

3. THE IFA-631 TEST

The IFA-631 test in Halden research reactor, [3], was carried out to study the phenomena involved in secondary degradation of BWR fuel after simulation of a primary failure. Four short rodlets were tested with liner claddings consisted of an outer Zircaloy-2 component and an inner ZrSn-liner[a]. The liner was about 10% of the cladding's wall thickness. In two rodlets the inner cladding surface was preoxidized (zirconia layer ~ 2 μm) to create an additional barrier towards hydrogen pick-up after simulation of the primary failure. Solid fuel pellets of two different diameters were used to ensure different pellet-to-cladding gaps.

Fuel stack length in the rodlets was 1200 mm. Each rod was instrumented with two fuel center thermocouples (TCs) – in the lower and in the upper end. To insert the TCs two central holes 2.2 mm in diameter were drilled to a depth of 100 mm (TC position) in the fuel column from each rod end. The as-fabricated rodlets were filled with 30% Ar + 70% He gas mixture to a pressure of 2.9 bar at 20 ^{o}C. With the chosen composition a thermal conductivity of the fill-gas was distinctly higher than that of steam and markedly lower than that of hydrogen. The main features of the four test rodlets are presented in Table 1.

Table 1. The main parameters of the IFA-631 test rodlets, [3].

Parameter/Rodlet	631-1	631-2	631-3	631-4
Cladding outer diameter, mm	9.62	9.62	9.62	9.62
Cladding inner diameter, mm	8.36	8.36	8.36	8.36
Pellet outer diameter, mm	8.19	8.25	8.19	8.25
Radial pellet-to-cladding gap (μm)	85	55	85	55
Preoxidized inner cladding surface	no	no	yes	yes
Labeling of TCs in the lower/upper part of fuel stack	TC1/TC5	TC2/TC6	TC3/TC7	TC4/TC8

To simulate the primary failure the test rods were equipped with a special water-ingress device (WID) connected to the upper end plug. After opening a valve in the device the water passed through a 110 mm long tube with an outer diameter of 1 mm and a wall thickness of 0.2 mm before entering the rod. A scheme of the fuel rodlets is shown in Fig.3.

The IFA-631 test was performed in the Halden reactor loop under BWR conditions (286 ^{o}C, 70 atm, normal BWR water chemistry). The axial power profile during irradiation was asymmetric with the maximum in the lower part of the rodlets (Fig.3). The bottom-skewed power profiles are typical for a considerable part of the operating cycle in commercial BWRs.

Prior to opening of the WID valves the test rodlets were irradiated under steady-state conditions for 144 days, reaching an average burnup of 5.3 MWd/kgUO_2. After simulation of the primary failure the valves remained open for additional 118 days of irradiation. The final fuel burnup was close to 9 MWd/kgUO_2 (see Fig.4). The PIE metallography revealed massive cladding hydriding of sunburst type and crack initiation in the lower part of all four rods. The rod 631-4 had a through-wall circumferential crack approximately 70 mm above the bottom end of the fuel stack. According to Ref.[3] massive hydrides formed in a few days after simulation of the primary failure.

The fuel centerline temperature history was recorded by the TCs in the course of irradiation. Fig.5 shows the detailed TC data provided in Ref.[3] for the first 15 days of irradiation with the opened WID valves. Fig.5 also shows fuel centerline temperatures calculated in Ref.[3] using the FTEMP code under the assumption that the gas in the pellet-to-cladding gap is either pure hydrogen or pure steam. It is evident from Fig.5 that during several days after the "primary failure" the gas phase in the lower part of the rodlets was enriched with hydrogen, then steam fraction progressively increased. In the upper end of the rodlets steam dominated continuously.

The data presented in Fig.5 can be used to make qualitative assessments of the hydrogen-to-steam ratio in gas mixture in the lower and upper parts of the test rodlets. For this purpose we must have information on fuel thermal mechanics in the course of irradiation. Namely we should know

[a] The ZrSn-liner was developed to improve the corrosion resistance of cladding inner surface in defective fuel rods. Its PCI resistance is close to that of the unalloyed Zr-liner.

the dimensions of pellets and pellet-to-cladding gap under power load. Then if thermal conductivity of the fill-gas in the gap is known as a function of gas composition we can calculate the evolution of the hydrogen-to-steam ratio inside the test rodlets after simulation of the failure.

To find composition of gas mixture in the IFA-631 test rodlets the RTOP-CA code was used, [11]. The first series of calculations on fuel thermal mechanics were made for the pre-irradiation stage of 144 days when the rodlets operated as standard intact fuel. The RTOP-CA models for densification/swelling and relocation of BWR fuel were checked at this stage. The fill-gas in the gap was taken as the initial 30% Ar + 70% He mixture. The calculated central temperatures for rods 631-1 and 631-3[a] in comparison with the thermocouple data are show for example in Fig.6*a*. In the lower part of the rodlets (the high temperature domain where secondary hydriding took place) the calculated temperature curve lies between the TC1 and TC3 data. The difference in the TC readings is within ~ 15 ^{o}C and may be attributed, e.g., to some variations in actual power for the two neighboring rodlets in the test rig or to some distinctions in mounting of the two thermocouples in fuel stack. In the upper part of the rodlets (the low temperature domain) deviation between the calculations and the TC data is about 25 ^{o}C while difference in the TC1 and TC3 readings is about 10 ^{o}C.

The second series of the RTOP-CA calculations was carried out for the first two weeks after simulation of the primary failure. Composition of gas mixture in the rodlets was varied in this numerical exercise. The central temperature was determined for either pure steam or pure hydrogen inside the fuel rods. Results were compared with the FTEMP calculations, [3]. The case for rod 631-1 is shown for example in Fig.6*b*. The maximum deviation between predictions by the two codes is for pure steam atmosphere. It is less then 10 ^{o}C for the lower thermocouple (high temperatures) and up to 15 ^{o}C for the upper TC (low temperatures). For pure hydrogen atmosphere difference in the central temperature according to the FTEMP and RTOP-CA codes is within 10 ^{o}C. Remarkably, that the RTOP-CA predictions are more conservative in viewpoint of solution of the inverse problem.

The inverse problem consists in interpretation of the TC data in terms of hydrogen-to-steam ratio in the lower and in the upper part of the rodlets. Since both measurements and calculations are made with some error the composition of the gas phase can be found with accuracy to some range. To assess the boundaries of this range we moved the TC data curves for 10 ^{o}C up and down and the inverse problem was solved for the new arrangement of the "experimental data". The results of data processing are shown in Fig.7.

An additional uncertainty in final results is induced by initial fill-gas in the rodlets – the Ar-He mixture. Estimations show that He and Ar should leave the rod in 1-3 days after occurrence of the primary failure. Nevertheless, the additional series of calculations was carried out with assumption that contents of Ar and He in the rodlets remained unchanged. This case leads to higher hydrogen-to-steam ratios inferred form the TC data. As follows from the analysis, the uncertainty in solution of the inverse problem due to the unknown Ar/He content in the rodlets is noticeably lower than that due to error (~ 10 degrees) in temperature calculations and measurements.

Severe hydriding occurred in all four rodlets. So, the massive hydriding threshold may be conservatively estimated by the minimal hydrogen-to-steam ratio reached in the lower part of the test rods. The actual threshold $\beta = P_{H_2} / P_{H_2O}$ may be even less than this conservative estimation. According to Fig.7 the threshold β value is bounded from above by a value of ~ 10. Temperature of the inner cladding surface in the lower part of the IFA-631 test rodlets was about 310 ^{o}C. Thus, the critical hydrogen-to-steam ratio under the in-pile conditions [3] differs from the out-of-pile threshold [5-7] by more than ~ 4 orders of magnitude.

[a] Rods 631-1 and 631-3 were identical in dimensions and were irradiated under the same conditions. Difference in central temperatures due to thin oxide film at inner cladding surface in rod 631-3 does not exceed 1 ^{o}C.

4. THE DEFEX PROJECT

Experiments [4] in the frame of the DEFEX project were performed in research reactor R2 in Studsvik. The objective of the project was to study the effect of cladding type, pellet-to-cladding gap, heat rate and irradiation time on secondary fuel degradation and occurrence of secondary failure.

In the tests standard Zircaloy-2 cladding was compared to cladding with the unalloyed Zr-liner as well as to rifled cladding. The "rifled cladding" had a very shallow prismatic "rifling" on the inner surface. It has been proposed to facilitate axial mass transfer when pellets come into contact to cladding. The "rifling" was considered as a possible remedy against secondary failure by hydriding. The cladding outer diameter was 12.25 mm, the wall thickness was 0.8 mm. Fresh fuel pellets with different outer diameter were used to provide a variety of pellet-to-cladding gap size. Diametral gap in most rodlets was either 150 or 80 μm, two rods were made with axially profiled gap – 150 μm in the top and bottom zones and 80 μm in the middle zone[a]. Fuel column in the test rods was about 400 mm high. The main fuel parameters as well as the irradiation conditions are shown in Table 2.

The primary failure was simulated with a technique different from that used in the IFA-631 test, [3]. The simulation device consisted of an array with a water reservoir in an extended plenum. It was connected to the rodlet plenum with a thin tube (see Fig.8*a*). After adding water into the reservoir the rodlets were filled with He gas (5 atm at room temperature).

When the test rodlets were inserted into the test loop, in which the circulating coolant was kept at around 265 °C, some of the water in the reservoir evaporated. Consequently, at the beginning of the irradiation steam was distributed along the fuel stack. At ~ 265 °C the saturation pressure of steam is about 50 atm. After the rodlets were inserted into the loop the power was increased up to the holding level as rapidly as possible (see Fig.8*b*). In the course of irradiation the test rodlets were constantly supplied with steam from the water reservoir in the extended plenum. As steam was consumed by oxidation of fuel and cladding additional amounts of water evaporated in the reservoir and new portions of steam entered the underlying parts of the rodlets.

The peak linear heat rate in different rods during irradiation was hold in the range of 50 to 25 kW/m. The axial power profile was close to symmetric, so the peak power load was attained in the middle fuel stack zone (Fig.8*c*). The initial pellet-to-cladding gaps were intentionally chosen to provide the gap closure in the middle zone at the holding level of linear heat rate. This hard pellet-cladding mechanical interaction was believed to be an obstacle for steam from the simulated defect in the top to flow and diffuse downwards. This should have promoted the conditions of oxygen starvation in the bottom zone of the rodlets. But the PIE data on fuel/cladding oxidation indicated that the gap could not be considered completely closed and steam had to permeate into the lower part of fuel stack in noticeable amounts, [4].

Table 2. Summary of data and results of the DEFEX tests, [4].

DEFEX test No.	**1**	**3**	**4**	**5**	**5X**	**6**	**7**	**8**	**9**	**10**
Cladding type	Zry-2	liner	rifled	Zry-2	Zry-2	liner	Zry-2	Zry-2	liner	liner
Gap, μm	150	150	150	150	150/80/150	150/80/150	80	150	150	80
Hours of irradiation, hrs	90	281	103	76	12	12	76	167	168	48+1[1)]
Linear heat rate when PCI occurs, kW/m	30	27	25-30	25	20	20	20	25-28	–	20
Holding linear heat rate, kW/m	50	50	50	40	40	40	26	30	30	40/62[*]
Loop pressure, atm	89.9	84.7	84.9	84.8	84.9	84.7	84.7	84.8	85.5	84.5
Coolant inlet temperature, °C	264.3	265.6	264.8	265.5	265.5	266.2	265.7	265.7	265.7	264.8
Massive hydriding	yes	yes	yes	yes	yes	no	no	no	no	no
Failure	yes	yes	yes	yes	no	no	no	no	no	no
Time to failure, hrs	88	216	103	75	–	–	–	–	–	–
[*] Test with power ramp at the end of irradiation										

[a] In the DEFEX database, [4], all rods are divided into 3 axial zones of equal length. Initial dimensions and irradiation history are given for each of the 3 zones.

The major result from the tests was that in the high-power rodlets (peak holding level 50 kW/m) both severe hydriding and "secondary" failure occurred. Massive hydrides and through-wall cracks formed in the bottom zone of the test rodlets – the most distant part from the steam source in the extended plenum. For lower peak power loads of 40 kW/m the experimental results were not so straightforward. One of the rods with standard Zircaloy-2 cladding experienced both hydriding and failure. Another rod with standard cladding was massively hydrided but remained intact. The rodlet with liner cladding successfully operated at 40 kW/m without any abnormal hydriding. In the rodlets operated at peak heat rates below 30 kW/m no excessive hydriding was observed regardless of cladding type.

The post-irradiation examinations for several rods included studies of fuel stoichiometry in two cross-sections – one from the upper and one from the lower axial zone of fuel column (Fig.9*a*). Radial stoichiometry distributions for UO_{2+x} were not specified in Ref.[4]. The fuel oxidation data comprise only the maximum, minimum and average levels of x for each examined cross-section. The initial deviation from stoichiometric UO_2 in pellets was $x \approx 0.003$. For several cases the full content of hydrogen gas remained inside rodlets was measured. These data may be used to estimate the composition of steam-hydrogen mixture in the test rodlets during the irradiation.

At given temperature the equilibrium deviation from stoichiometric UO_2 in fuel is entirely governed by the ratio of hydrogen and steam partial pressures in the ambient gas mixture (Fig.9*b*). If we know fuel temperature (it was calculated with the RTOP-CA code) and take the data [4] on UO_{2+x} stoichiometry then Fig.9*b* will give us the hydrogen-to-steam ratio in the DEFEX rodlets. The final estimations for the bottom zone of fuel column are listed in Table 3: for each test the range of $P_{\mathrm{H_2}} / P_{\mathrm{H_2O}}$ is shown which corresponds to the maximum and minimum values of x measured experimentally. The numbers in brackets are the $P_{\mathrm{H_2}} / P_{\mathrm{H_2O}}$ values which correspond to average stoichiometry of UO_{2+x} in the examined cross-section.

Table 3 also shows the estimated composition of steam-hydrogen mixture inferred from the data on the hydrogen gas remained in the rodlets after the irradiation. Estimations were made assuming that steam partial pressure in the rodlets was equal to the saturation water pressure at 265 $^{\circ}$C.

As follows from Table 3, the hydrogen-to-steam ratio inside fuel rods with the simulated primary defect in the course of the DEFEX experiments did not exceed the values of about 10. Nevertheless, at high linear heat rates these ratios appeared to be sufficient to cause massive hydriding and even lead to "secondary" failure in the bottom part of the test rodlets. The temperature of cladding inner surface in the bottom axial zone of the rodlets was about 310 $^{\circ}$C or less. So, the threshold of massive hydriding resulting from the in-pile experiments [4] is much lower than the out-of-pile criterion, Eq.(1). This is to confirm a substantial decrease of the hydriding threshold under irradiation conditions.

Table 3. Hydrogen-to-steam ratios inferred from the DEFEX data analysis.

			estimated $P(H_2)/P(H_2O)$	
			by data on fuel oxidation	by data on hydrogen gas amount
massive hydriding	yes	DEFEX-1	0.06 – 35 (0.15)	–
		DEFEX-3	0.02 – 0.06 (0.03)	–
		DEFEX-4	0.04 – 0.1 (0.08)	–
		DEFEX-5X	–	0.08
	no	DEFEX-6	–	0.13
		DEFEX-7	–	0.24
		DEFEX-8	0.016 – 4 (0.03)	0.5
		DEFEX-9	0.02 – 4 (0.03)	0.5
		DEFEX-10	–	0.13

5. CONCLUSION

It is shown that under irradiation the onset of massive hydriding occurs well before approaching the conditions of oxygen starvation found in the out-of-pile tests. This conclusion is derived from the data of experiments in research reactors. The key factor governing the change of the hydriding threshold is thought to be the intense generation of oxygen sublattice defects in the oxide film on cladding inner surface. This irradiation damage is due to fission fragments leaving the periphery of fuel pellets. Eqs.(1) and (2) show that the threshold of massive hydriding under irradiation may be written in the following form

$$P_{\mathrm{H_2}} / P_{\mathrm{H_2O}} = A\dot{F}^{-1} \exp\left(B/T\right). \qquad (3)$$

Eq.(3) may be used to determine the threshold of severe secondary hydriding in fuel of different design. For this purpose it is necessary to have a mechanistic code capable of calculating the axial distribution of hydrogen gas inside a fuel rod with a primary defect. Secondary failure criteria for WWER fuel may be developed by means of the certified mechanistic code RTOP-CA.

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Figures

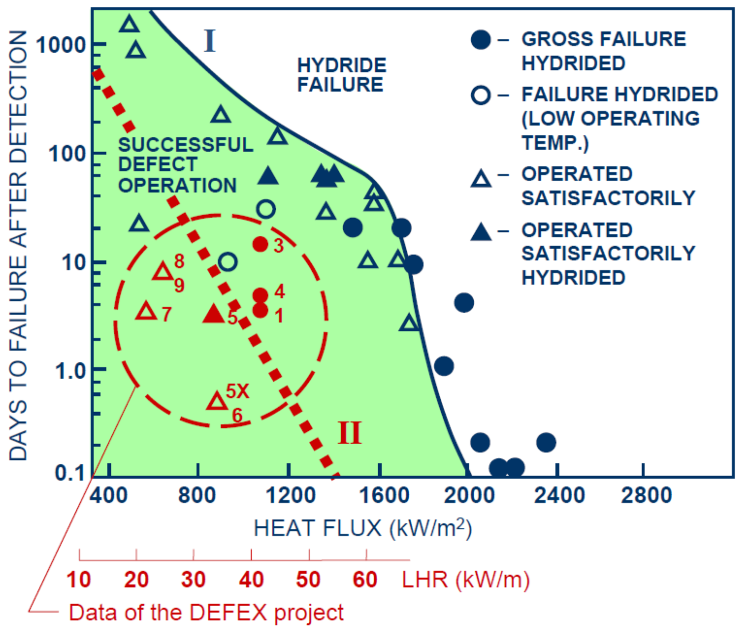


Fig.1. The Locke diagram. Dark-blue markers and line I are the experimental data and the Locke criterion of secondary failure, respectively, [2]. Green area is the range of parameters for successful operation of fuel rods with a primary defect. Red markers are the data of the DEFEX project, [4] (the corresponding linear heat rates are shown below in red color); line II is the rough boundary separating the DEFEX tests with successful fuel operation from that with secondary failures.

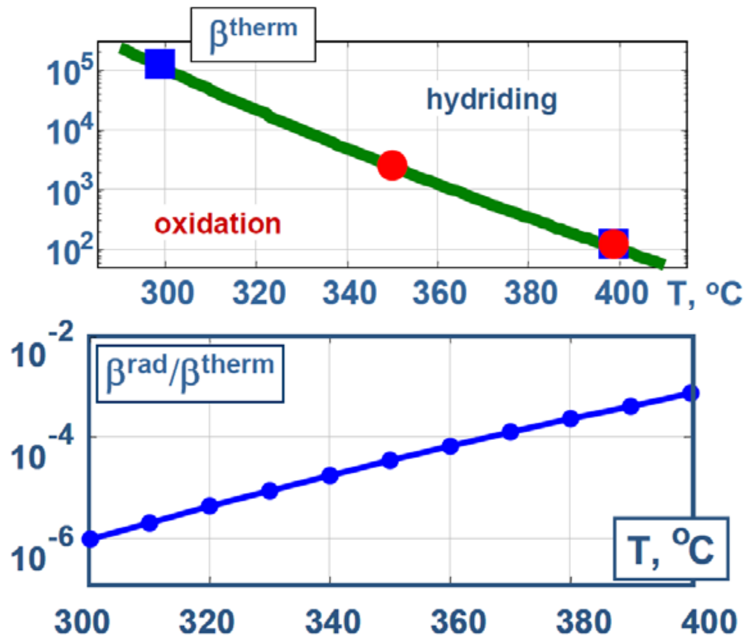


Fig.2. *a* – The out-of-pile data on the threshold of massive hydriding: ● – sponge Zr at total gas pressure of 70 atm [5], ■ – Zircaloy-2, 1 atm [7]. *b* – The change of the threshold under irradiation as estimated in Ref.[8].

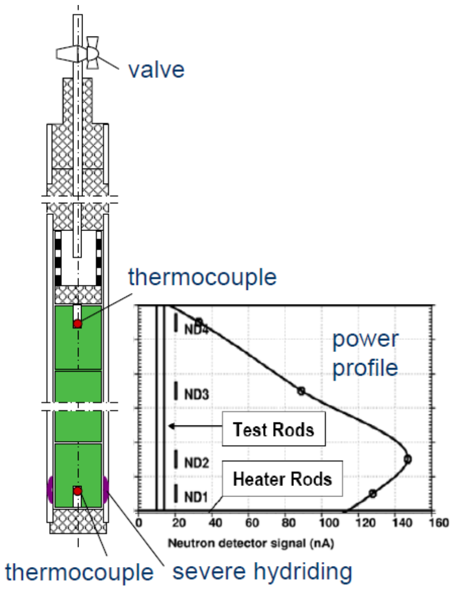


Fig.3. The scheme of the IFA-631 test rodlets and the axial power profile during irradiation.

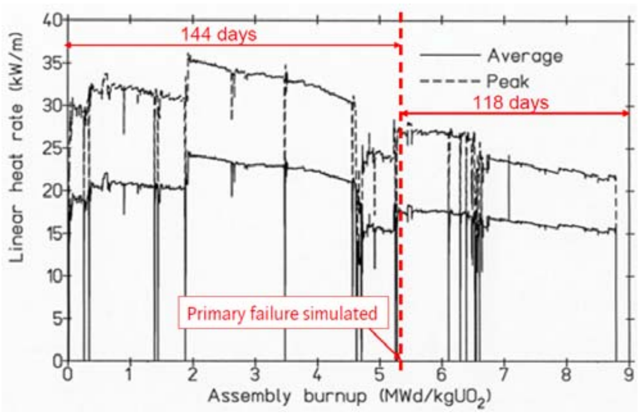
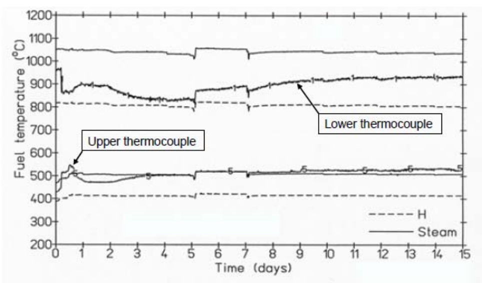
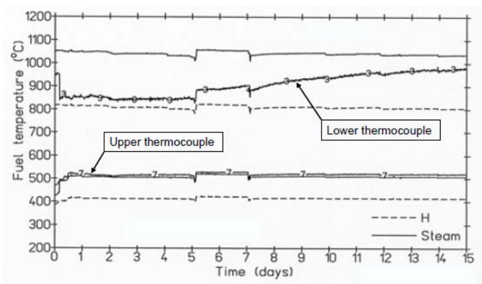


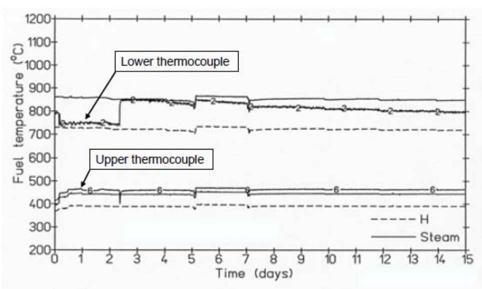
Fig.4. Irradiation history of the IFA-631 fuel rodlets, reproduced from [3].



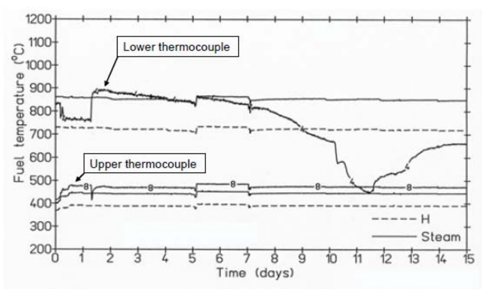
a) Rod 631-1 (Radial as-fabricated pellet-clad gap: 85 μm)



c) Rod 631-3 (Radial as-fabricated pellet-clad gap: 85 μm)



b) Rod 631-2 (Radial as-fabricated pellet-clad gap: 55 μm)



d) Rod 631-4 (Radial as-fabricated pellet-clad gap: 55 μm)

Fig.5. Thick lines – TC readings in the test rodlets during the first 15 days after simulation of the primary failure. Thin lines – centerline temperature calculated for the TC axial positions with the FTEMP code under the assumption that the gas in the pellet-to-cladding gap is pure hydrogen (dashed line) or pure steam (solid line). Reproduced from [3].

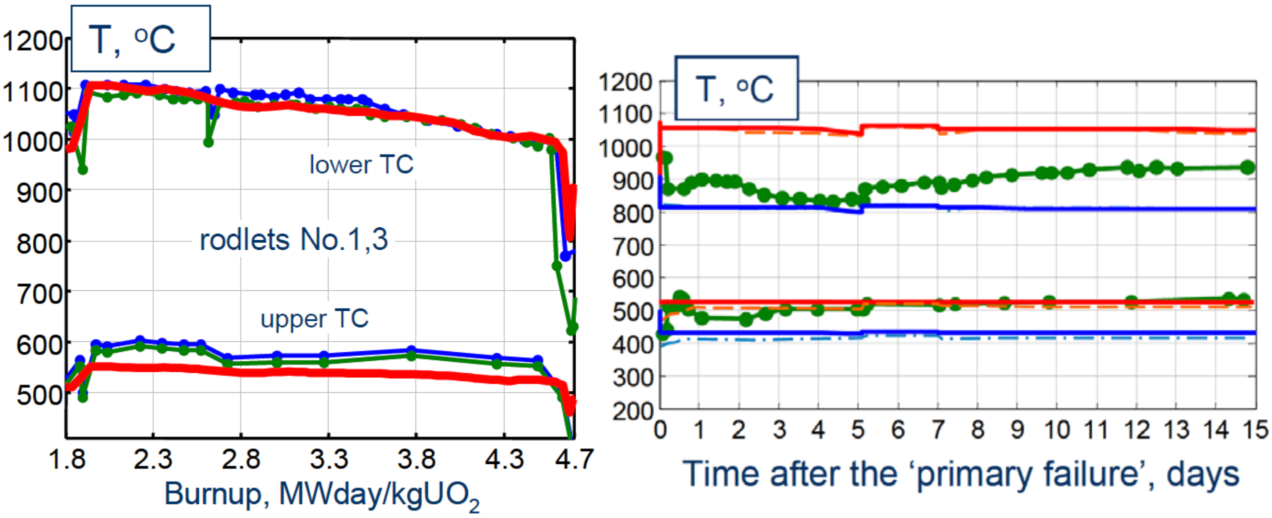


Fig.6. The RTOP-CA calculations for rodlet 631-1 compared to the TC data.

a – The pre-irradiation stage: ▬▬ – calculated central temperature, ▬■▬, ▬■▬ – TC readings in rodlets 631-1 and 631-3.

b – The stage after occurrence of the “primary failure”: ▬●▬ – TC data for rodlet 631-1; ▬▬ – central temperature calculated with the RTOP-CA code in assumption of pure steam inside the rodlet, ▬▬ – central temperature in assumption of pure hydrogen; pure steam (— — —) and pure hydrogen (– · – · –) calculation with the FTEMP code, [3].

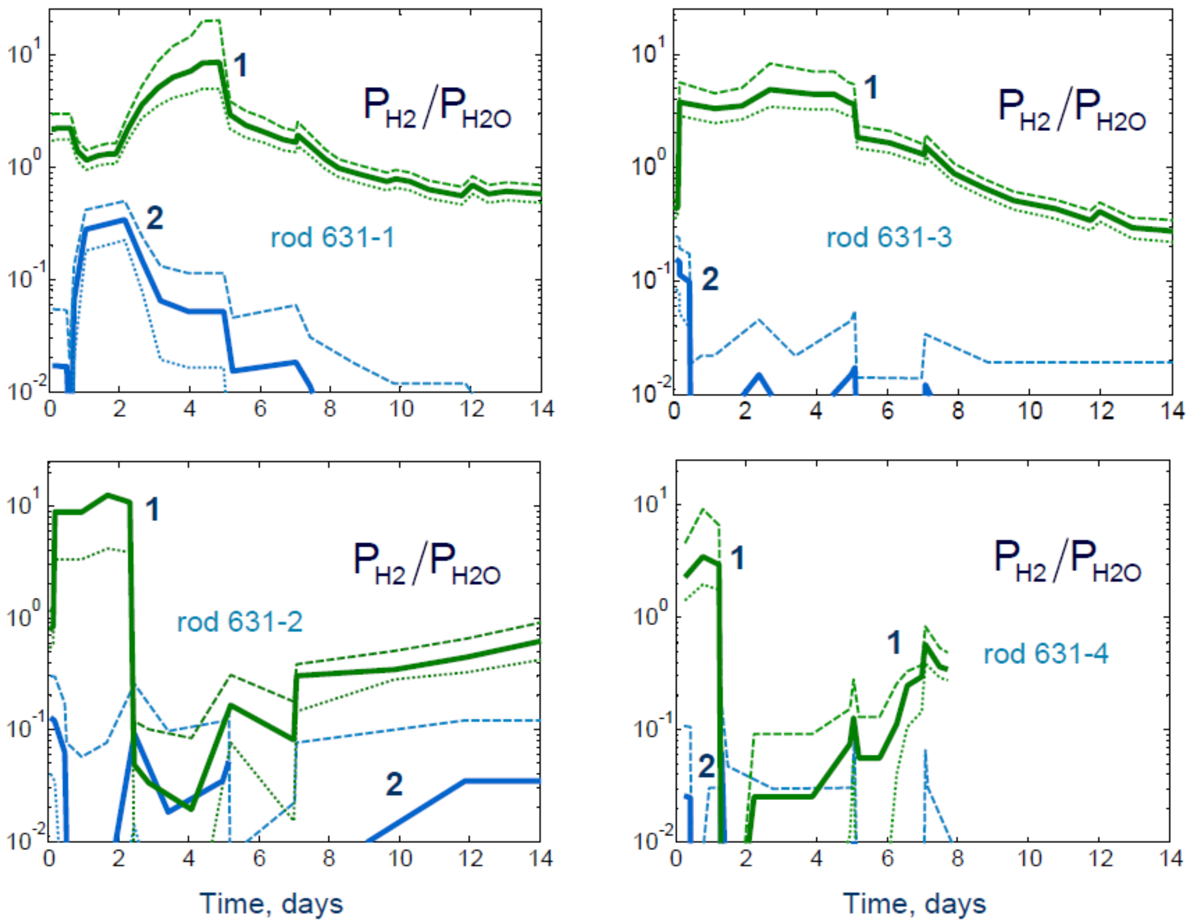


Fig.7. Evolution of hydrogen-to-steam ratio after simulation of the primary failure – as inferred from the data [3]. Lines in green color (1) are for the lower part of the rodlets with higher heat rate (area of secondary hydriding), lines in blue (2) are for the upper part of the rodlets (the low temperature domain). Dashed lines show the uncertainty boundaries for solution of the inverse problem.

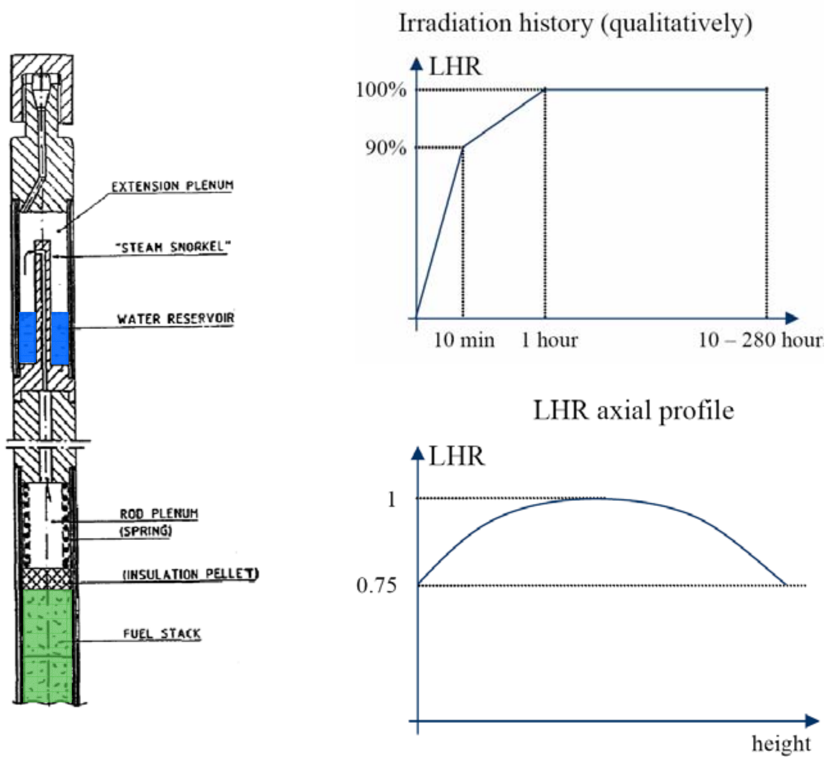


Fig.8. Data to the DEFEX project, [4]: *a* – a scheme of the test rodlets; *b*, *c* – sketches for linear heat rate history and axial power profile, respectively.

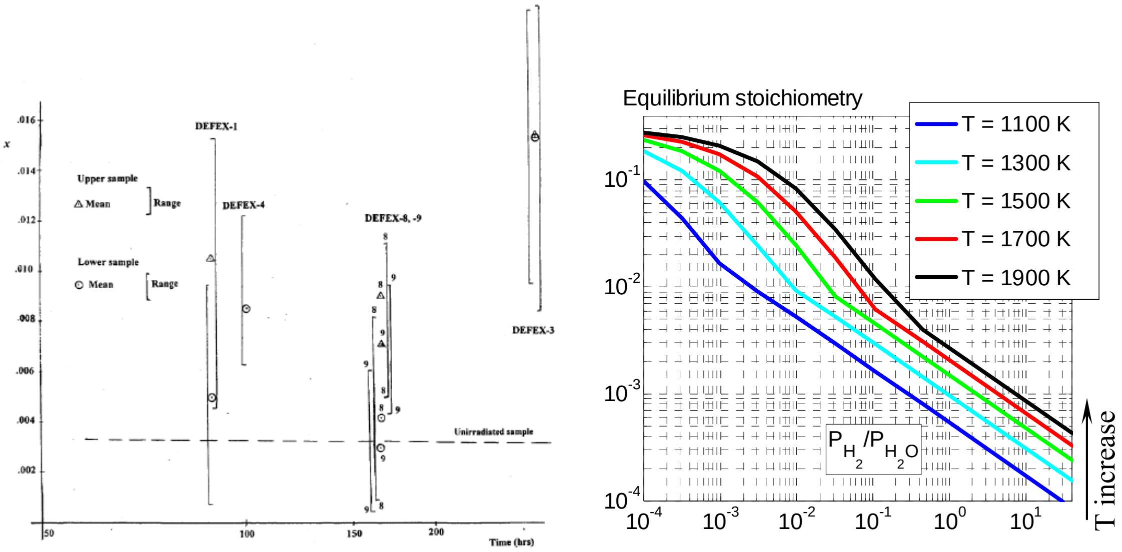


Fig.9. *a* – The DEFEX PIE data on fuel stoichiometry, [4]. *b* – Equilibrium deviation (x) from the stoichiometric composition of UO_{2+x} at different temperatures as a function of hydrogen-to-steam ratio in the gas phase.