Summary of the Core Degradation Experiments CODEX

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Abstract: An experimental series has been carried out in the integral high temperature test facility CODEX (COre Degradation Experiment) between 1995-2002 with electrically heated UO_2 fuel rod bundles. The test matrix included the first VVER-440 type integral severe accident experiment. The results of a quench test with pre-oxidised bundle indicated the protective role of the external oxide scale. Unique experiments were performed with PWR bundles under air ingress conditions. The last test of the current series helped to resolve the methane production issue during the oxidaton of a boron-carbide control rod in a severe accident. Some experiments were related to the preparation of PHEBUS tests, and some others were performed parallel with similar QUENCH tests. The experimental results contributed to the general understanding of severe accident progression in the loss of rod-like geometry phase and the test data have been used and are available for model development and code validation purposes.

1. INTRODUCTION

The loss or reduction of heat removal from the reactor core during a severe accident results in the heat-up and degradation of fuel rods. The high temperature leads to chemical interactions between the core components, hydrogen is produced and fission products are released from the damaged core.

The TMI-2 accident and the investigation of the damaged core provided a general picture on the main phenomena taking place in a light water reactor during a severe accident. The prediction of the consequences of severe accidents needs detailed description of the degradation process and computational tools. Large part of the model development is based on so called separate effect tests, which investigate some selected phenomena and can establish correlations between the measured parameters (e.g. oxidation of zirconium or dissolution of uranium-dioxide by molten materials) The complex phenomena of core degradation can be studied in integral tests facilities, where the different processes take place simultaneously and the experiments can indicate the importance of different aspects of the accident. Experimental programmes with integral severe accident tests have been carried out in in-pile (e.g. LOFT, PBF, FR2, PHEBUS) and out-of-pile (e.g. NIELS, CORA, QUENCH) facilities and significantly improved our knowledge on severe accidents.

The CODEX out-of-pile integral test facility was built and put into operation in 1995 at the KFKI Atomic Energy Research Institute, Hungary in order to investigate some specific aspects of core degradation and to extend the experimental database for code valiadation and development. Some of the experiments were VVER specific, while others were of general interest for any light water reactor. The comparison of CODEX exepriments with CORA and QUENCH tests can help to sift out the effects related to the specific features or scaling of the facilities. Some new techniques (e.g. aerosol measurements) applied in the test facility provide additional information on the high temperature behaviour of core materials. The CODEX test results are condsidered in the preparation of in-pile PHEBUS tests as well.

2. THE CODEX FACILITY

The CODEX facility was built for the investigation of the behaviour of small fuel bundles under severe accident conditions. The first test series was devoted to the experimental study of VVER type fuel assemblies, later PWR bundles were applied as well.

The VVER type assembly is constructed of seven fuel rods arranged on a hexagonal lattice. The PWR bundle includes nine rods in square arrangement. The peripheral rods are electrically heated with tungsten bars. The central rod is not heated, it is used for instrumentation. Two or three spacer grids are applied to fix the bundle. The heated length of the bundle is 600 mm. The cladding material is Zr1%Nb (VVER) or Zircaloy (PWR), the shroud part is made of a 2 mm thick Zr2%Nb alloy. Inside of the fuel rods annular UO_2 pellets are placed between the heater bars and the cladding. Around the shroud ZrO_2 thermal insulation is added. The test section is connected to the preheater and to the cooler sections as coolant inlet and outlet respectively (Fig. 1.). An additional junction is connected to the bottom part of the bundle to inject cold gas or water. The preheater unit is able to supply either hot gases or steam to the test section. The off-gas streaming out of the test section is cooled down by the cooler/condenser unit and before releasing it into the atmosphere it was conducted through an aerosol trap and filtered by a special system. For the investigation of the aerosol release a cascade impactor system is connected to the upper plenum of the cooler and two pipelines allowes the continuous measurement of aerosols by means of laser particle counters. The gas concentration in the off-gas system is measured using a quadropole mass spectrometer.

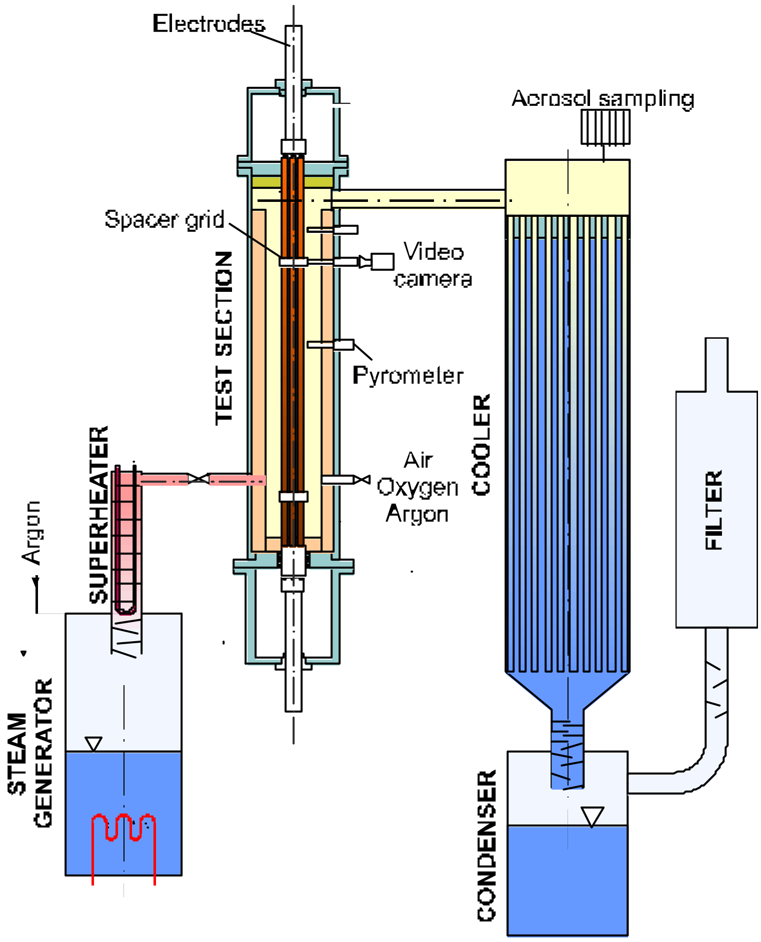

Fig. 1. Main components of the CODEX facility

The instrumentation of the facility consists of the measurements of the operational parameters as heating power, flowrates, temperatures, levels and pressures. Thermocouples are placed in several positions in the heat insulation material, on the heat shield, on the external surface of the shroud, on the fuel rods and inside of the central (unheated) rod. Two pyrometers and a video camera are located at three windows in the upper part of the bundle. After the experiments the post-test examination of the bundle and aerosol samples is carried out with several techniques, including metallography, SEM, microprobe analysis, X-ray radiography and mass spectrometry.

The main advantages of the CODEX facility are the use of real UO_2 pellets, the sophisticated data acquisition technique including aerosol measurements and the large flexibility in the selection of test conditions. The most imporant limitations are the use of fresh – non-irradited - fuel pellets and the application of electrical heating burdened with positive temperature feed-back effect.

3. THE EXPERIMENTAL PROGRAMME

At the AEKI an experimental programme was initiated focusing on the high temperature behaviour of VVER fuel and core materials. The interactions of Zr1%Nb cladding with UO_2 pellet, stainless steel spacer and boron steel absorber were studied in small scale separate effect tests [1]. On the basis of the experience gained in those tests the CODEX integral test facility was constructed to continue this work under more prototypic conditions.

First the capabilities of the facility were demonstrated carrying out the CODEX-1 experiment with Al_2O_3 pellets. The test section was heated up with argon, then the electric power was increased. When the rod bundle degradation was indicated by temperature measurements, the power was switched off and the section was cooled down by argon. The post-test examination showed that the rod bundle partially damaged, the further melting was stopped in time. So the facility proved to be applicable to the experimental analysis of controlled core degradation processes.

In the second experiment similar procedures were taken, but the Al_2O_3 pellets were replaced with UO_2 [4,5]. The CODEX-3/1 and CODEX-3/2 experiments were performed with quick water cooling [6,7]. Air ingress conditions were simulated in the AIT-1 and AIT-2 tests with PWR fuel rods [2]. The CODEX-B4C test was devoted to the examination of control rod degrdadation and gas production during a severe reactor accident. The main parameters of the test matrix are given in Table 1.

Table 1. Main parameters of CODEX test matrix

Test	Bundle type	Pellet	Year	Test type	Ref.
CODEX-1	7-rod VVER	Al_2O_3	1995	scoping test	
CODEX-2	7-rod VVER	UO_2	1995	escalation and slow cooling down	[2,3]
CODEX-3/1	7-rod VVER	UO_2	1996	water quench at 1150 ^oC	[4,5]
CODEX-3/2	7-rod VVER	UO_2	1997	water quench at 1500 ^oC	[4,5]
CODEX-AIT-1	9-rod PWR	UO_2	1998	air ingress	[6,7,8]
CODEX-AIT-2	9-rod PWR	UO_2	1999	steam oxidation and air ingress	[7,8,9]
CODEX-B4C	7-rod VVER	UO_2,B_4C	2001	control rod degradation	[10]

4. RESULTS AND CONCLUSIONS

In the following chapters the main results and conlcusions of CODEX tests with UO_2 pellets are summarised and short descriptions of the tests are given.

4.1 The first VVER-440 integral test

The first experiment with UO_2 pellets - named CODEX-2 - was performed in December 1995. The heat-up of the core after loss of heat removal in steam atmosphere was simulated with a VVER-440 type fuel rod bundle. The experiment consisted of three main phases:

- First the bundle was preheated up to 500 ^oC by a constant inlet flow of argon (1.4 g/s) with an inlet temperature equal to 600 ^oC and a stable temperature distribution was established.
- The test phase started with switching on electrical rod heating: the electrical power was linearly increased with a speed of 2 W/s and steam was added to the argon flow. The steam flow rate was ~1.0 g/s and its temperature ~600 ^oC. When the clad temperature reached 1200 ^oC rod temperature escalation was observed (Fig.2.). The pyrometer measurements showed that the highest temperature (2400 ^oC) was reached close to the top of the bundle. In the lower part of the

bundle - 50 mm from the bottom of the heated zone - the maximum temperature remained below 750 °C. The maximum value of measured hydrogen concentration reached 36 mg/s, the total hydrogen production during this test was 26.6 g. The presence of hydrogen indicated the chemical reaction between cladding and steam, which contributed to the heat up and was the cause of the escalation of rod temperatures.

- In the final phase the cooling down was initiated. The electrical heating was switched off, the steam injection was stopped and the facility was cooled down slowly in argon.

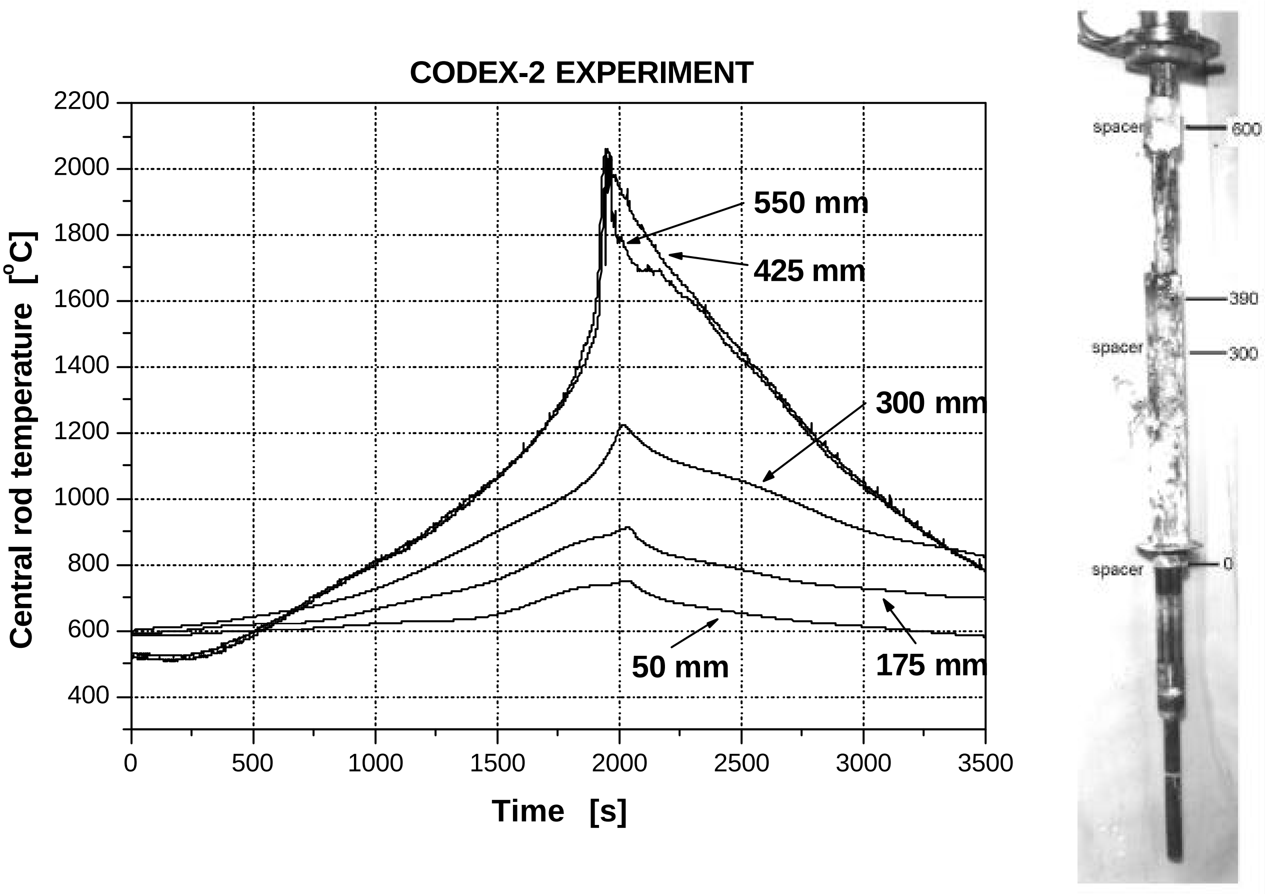

Fig. 2. Central rod temperatures in the CODEX-2 experiment (left)
and view of the bundle after the test (right)

For post test examination of the rods, the bundle was filled up with epoxy and afterwards cut into cross sections in order to facilitate the further investigation. The visual analysis of the polished cross sections showed that during this experiment the early phase of core degradation was reached: the upper part of the bundle melted down, the lower part had no damage and in the central part the stages of partial loss of geometry were observed. In the photograph of the bundle it can be seen, that the shroud and large part of the cladding materials were lost between 400-550 mm elevations. Some part of these materials were melted, but other broken parts fell down and were found as heavily oxidised fragments in the lower part of the section.

The use of VVER materials and fuel assembly geometry made possible to compare the behaviour of VVER reactors under core damage conditions to the better investigated western type reactors. On the basis of comparisons between CODEX-2 and CORA tests it was concluded that during the early phase of core degradation the same phenomena can be expected with VVER materials as with western design reactors. However for numerical simulation VVER specific material properties and correlations must be used.

4.2 Quenching of high temperature VVER bundles

The injection of water into a high temperature damaged core can help to cool down the fuel rods and stop the degradation process. However the experimental investigation of high temperature quenching of Western design PWR and BWR fuel bundles in the CORA facility showed that the flooding of Zircaloy bundles does not decrease the temperature immediately, but results in a preliminary increase and in a peak of hydrogen production before being quenched [11].

The main purpose of the CODEX-3/1 and CODEX-3/2 tests was to investigate the effect of water quenching on the degradation process of a VVER bundle. It was expected to receive similar results as in CORA tests for PWR and BWR bundles. The differences in the bundle geometry and core materials were to be evaluated. In both experiments water quenching was the cooling down process of the high temperature bundle. However the temperatures and the initial bundle states were different in the two cases. These parameters had an important effect on the final bundle states and the degree of degradation (Table 2).

Table 2. Main parameters of the CODEX-3/1 and CODEX-3/2 tests

	CODEX-3/1	CODEX-3/2
Argon flowrate	2 g/s	2 g/s
Steam flowrate	1.5 g/s	1.5 g/s
Heat up period	1800 s	2000 s
Maximum electrical power	3.3 kW	4.2 kW
Temperature of quenching	1150 ^oC	1500 ^oC
Period of quenching	90 s	80 s
Maximum temperature	1158 ^oC	1643 ^oC
Bundle state before test	as received	pre-oxidized
Bundle state after test	intact	damaged

The first test showed only very limited temperature excursion, which was in the magnitude of 10 ^oC (Fig. 3). The hydrogen concentration measurement indicated some relative peak, but its value was very low (below 1mg/s). The test results were in good agreement with the widely used peak cladding temperature criterion (1200 ^oC): water quenching took place close to but below this limiting value and the process did not lead to severe temperature excursion, the bundle was cooled down without losing the rod-like geometry. Furthermore the later investigation showed, that during this rapid cooling down process the fuel integrity did not damage. After the first phase the facility was disassembled and the bundle was taken out of the test section for visual investigation. No cladding failure, neither signs of cladding-spacer interaction were observed. The post-test calculation estimated 50 microns oxide layer thickness in the vicinity of highest temperature.

The second test was carried out with the same bundle, which was pre-oxidized in the first experiments. In this test water quenching was initiated, when the maximum temperature reached 1500 ^oC. The water injection initiated some temperature excursion, but the temperature increase was less then 150 ^oC and the maximum temperature remained below 1650 ^oC. Together with the temperature peak a hydrogen production peak was observed, which indicated the exothermic reaction of Zr oxidation, the total hydrogen production was about 1 g. In the previous CODEX-2 test, the excursion took place in steam atmosphere 26.6 g hydrogen was produced, which was much more then in the case of water quenching of the CODEX-3/2 pre-oxidized bundle.

The results of the CODEX-3/2 test were in some contradiction with similar high temperature quench tests performed earlier on the CORA facility. The German experiments with comparable reflooding rates normally lead to higher temperature escalation and resulted in partial melting of the cladding. There is an obvious difference between the CODEX-3/2 and the CORA tests: in the present case the high temperature quenching took place on a preliminary pre-oxidized bundle.

The CODEX-3/1 and CODEX-3/2 tests were carried out in November 1996 and January 1997. In February 1998 the QUENCH-01 test was performed at FZ Karlsruhe with a preliminary pre-oxidized bundle [12]. In this test the bundle appeared to quench steadily during reflood with no evidence of temperature excursion or excess hydrogen production. These facts are in good agreement with CODEX-3/2 test data and confirm that the reason for the unexpected behavior of bundle during quenching was not the application of VVER materials and geometry, but the protective role of oxide layer on the external surface of cladding material.

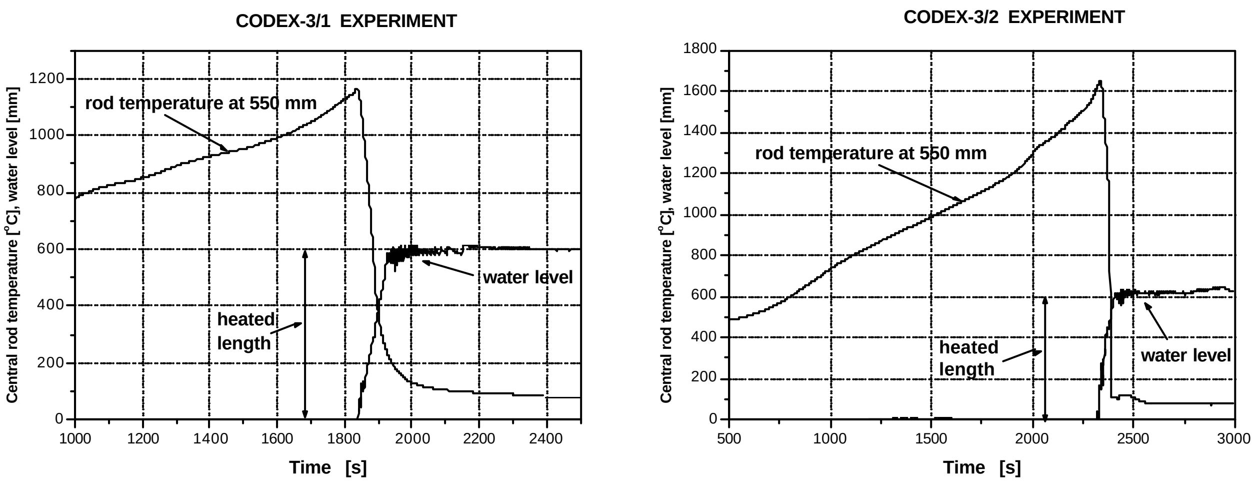

Fig. 3. Central rod temperatures and water levels in the CODEX-3/1 (left) and CODEX-3/2 (right) experiments

4.3 Air ingress tests

The investigation of the degraded core at TMI-2 revealed that the most extensive damage took place initially along the centreline of the reactor core while the peripheral regions remained intact although they were heavily oxidised. In the late phase of core degradation the core debris can slump to the lower plenum and cause rupture of the vessel. As a result of the chimney effect air can be sucked into the vessel from the reactor cavity and interact with the core materials. The ingress of air into the vessel will result in an extremely high oxidation rate of the remaining zirconium and oxidation of the fuel can be followed by the release of different kinds of fission products [13]. The objective of the CODEX air ingress tests (AIT) – which represent the first integral tests with air atmosphere - was to provide information on the consequences of air oxidation on the core degradation process.

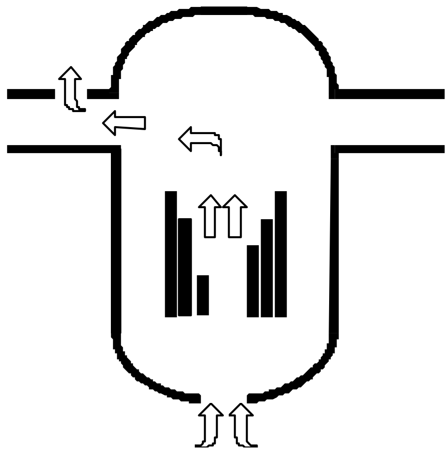

Fig. 4. Scheme of air ingress scenario

The two experiments CODEX-AIT-1 and CODEX-AIT-2 with slightly different conditions and with the same PWR 9-rod bundle design lead to rather different results regarding the extent of fuel rod and spacer grid degradation and relocation, the axial temperature profile, the intensity of nitride formation and U-release and the conditions of temperature excursions (Table 3). The pre-oxidation period was carried out in argon-oxygen mixture in the first test and in steam with limited air content in the second test. Short temperature escalation took place during this period in the first test, while stable temperature plateaus were measured in the second test. The air ingress lead to escalation in both tests, in the first test with constant power and in the second test with linear power increase. The bundle state after the first test showed very limited damage: the fuel rods remained mainly intact. They were heavily oxidized and thick oxide and nitride layers (Fig. 5.) were measured. The second bundle survived more extended damage, the fuel rods were fragmented and melted, part of them relocated to the lower regions. The zirconium-nitride formation was entirely different in the two tests: in AIT-1 thick layers were formed between the oxide and metal phases, while only very thin nitride layers appeared in slices of AIT-2 experiment.

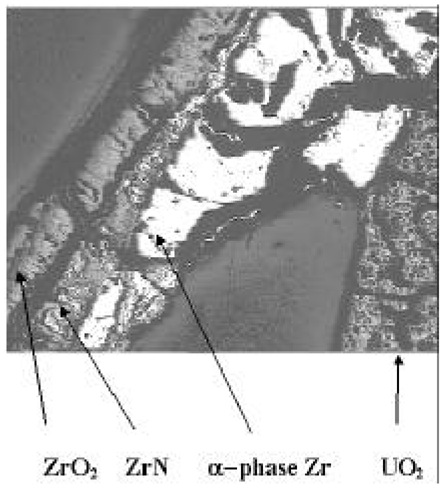

Fig. 5. Fuel segment layers of the CODEX-AIT-1 bundle

Table 3. Summary of CODEX-AIT experiments

	CODEX-AIT-1	CODEX-AIT-2
Pre-oxidation atmosphere	argon-oxygen	steam-air
Pre-oxidation temperature	~950 °C	~750 °C (1st step) ~900 °C (2nd step)
Pre-oxidation period	~100s	~3600s
Temperature excursions	1st in pre-oxidation phase 2nd in air ingress phase	in air ingress phase only
Air flowrate	3.5 g/s	2.5 g/s
Rod temperature before air ingress	~900 °C	~750 °C
Power history in air ingress phase	constant 1.25kW	linear increase from 1.6kW to 3.3kW
Air ingress period	~500s	~1000s
Maximum measured temperature	2000 °C	1880 °C
Max. nitride layer thickness	290 μm	30 μm
Max. oxide layer thickness	520 μm	170 μm
U-release	0.1-0.5 μg	50-100 μg
Final bundle state	mainly intact rod structure	relocation of fragmented and molten fuel

The CODEX-AIT-1 and AIT-2 air ingress tests indicated the acceleration of oxidation phenomena and core degradation processes in air compared to steam oxidation conditions. Heavy oxidation and nitride formation on the Zr surfaces lead to the mechanical degradation of the cladding. The high temperature oxidation resulted in the release of large number of aerosol particles, some of which contained uranium.

4.4 The role of B_4C control rod

During a severe accident the degradation of the core materials starts with the degradation of control rod materials, which have lower melting points than the fuel rods with zirconium cladding. Boron-carbide (B_4C) control rods or blades are used in different BWR and PWR type reactors, including the Russian design VVER-1000 The CODEX-B4C test was characterised by a VVER-1000 bundle with a central B_4C control rod in stainless steel cladding and Zr guide tube. The objectives were to provide experimental data on the impact of B_4C on the gas production (in particular H_2, CO, CO_2 and CH_4) and aerosols in conditions as representative as possible for a VVER-1000 reactor core under severe accident and on the impact of B_4C on the degradation of surrounding fuel rods and structures.

The test conditions of CODEX-B4C experiment were selected to simulate a severe accident scenario and emphasize the role of the control rod in the degradation process. The preheating period served to establish a stable temperature distribution in the bundle and in the test section without chemical interactions and changes in the geometry of the rods. The power ramp simulated the heat-up of the bundle as a consequence of loss of heat removal from the reactor core and the power production due to decay heat. After the power ramp the further temperature increase was artificially delayed with the regulation of steam flowrate and so an extended time window was available for the degradation of control rod before the beginning of fuel rod degradation. The steam starving conditions established in this period were supposed to create hydrogen rich atmosphere and in this manner to facilitate methane formation. The escalation period was initiated without further power increase, only the coolant parameters were changed. After reaching the desired degree of bundle degradation slow cooling down was applied in steam atmosphere. This last phase covered wide temperature range and provided possibility for the investigation of gas production. The maximum temperature ~2300-2400 ^oC was reached on the guide tube at 495 mm elevation and on the heated rods at 450 mm, but 2000 ^oC was seen at 300 mm elevation as well

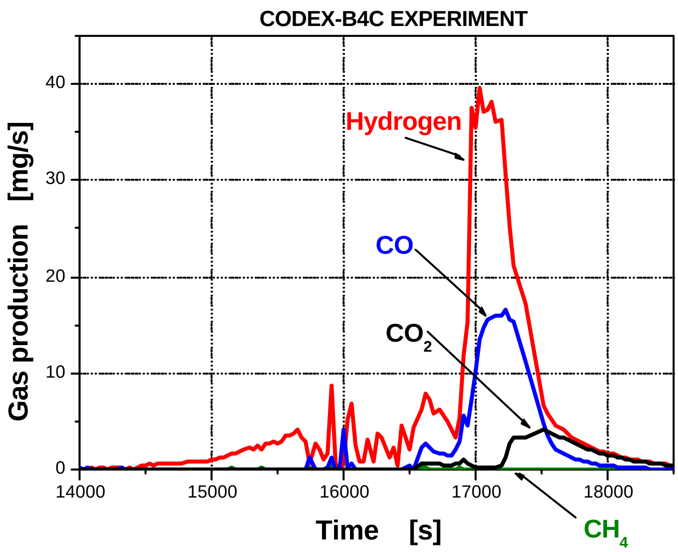

Fig. 6. Gas prdoction in the CODEX-B4C experiment

The gas measurement with mass spectrometer indicated hydrogen, methane, carbon-monoxide and carbon-dioxide. Significant hydrogen production was seen as a common result of Zr and B_4C oxidation in steam (Fig. 6.). The peak of hydrogen generation was related to the temperature escalation, the maximum H_2 production was ~40 mg/s and the total integrated H mass was about 25 g. The methane measurement showed very low values, practically below the detection limit. Both CO and CO_2 production was detected, the appearance of CO_2 was delayed compared to CO. The maximum production rate was 16 mg/s for CO and 4 mg/s for CO_2. The integrated mass of CO and CO_2 showed that 39% of the original B_4C was oxidized during the test.

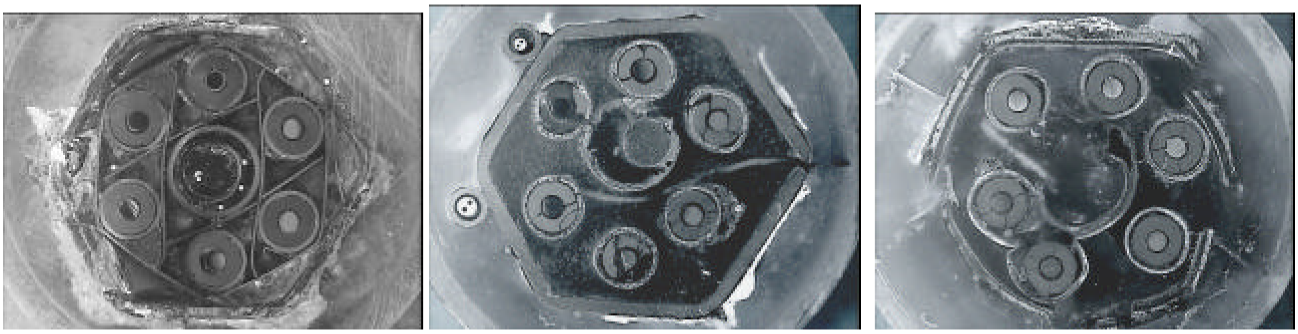

Fig. 7. Cross sections of CODEX-B4C bundle showing different degrees of control rod and fuel rod degradation

The cross sections of the rod bundle at different elevations showed the steps of control rod and fuel rod degradation. In the lower part of the bundle the rod remained intact, as the temperature was low. In the central part the stainless steel cladding melted down and the oxidation of B_4C started. The control rod completely disappeared from the upper part of the bundle (Fig.7.) due to high temperature oxidation and melting.

The two particle counters indicated high release of aerosols when steam was available in the coolant Dramatic increase was observed when the bundle temperature reached 1500 ^oC, the typical number of particles in each measured range was close to 10^6. The oscillating behaviour of the measured values was in direct correlation with the steam supply, which was in manual regulation mode. With no steam flow the number of particles decreased, while with available steam flow the sharply increased. It indicates that there was a strong correlation between the oxidation phenomena and the production of particles.

4.5 Use of CODEX experimental data

The experience gained in the CODEX tests can be applied to other integral facilities:

- One of the objectives of CODEX-B4C test was to support the preparation of in-pile PHEBUS-FPT3 experiments. The execution of air ingress tests CODEX-AIT-1 and CODEX-AIT-2 aimed to gain experience for the later PHEBUS-FPT5 experiment.
- Similar test conditions in QUENCH and CODEX tests (e.g. quenching of pre-oxidized high temperature bundle in QUENCH-01 and CODEX-3/2, or the B_4C tests QUENCH-07, QUENCH-09 and CODEX-B4C) and the differences in the facility desings make possible the better understanding of core degradation phenomena and the evaluation of facility specific or scaling effects.

The CODEX experiments not only extended our knowledge on severe accident related phenomane, but provided important data for code validation and model development purposes:

- The pre-test and post-test analysis of CODEX experiments is carried out in close international co-operation with European partner institutions. The severe accident codes ICARE/CATHARE, MELCOR and ATHLET-CD are used to assist in the selection of optimum experimental conditions before the test and to evaluate the experimental data after the tests. CODEX data are used for model development (e.g. air ingress model, B_4C oxidation and gas release) and code validation purposes as well.
- The CODEX experimental data were collected into an experimental database, which is available for code developers and evaluation teams. The database includes all on-line measurements in a time-dependent manner and the results of post-test examination procedure of bundle and aerosol samples. The CODEX facility and experiments are listed in the OECD in-vessel core degradation code validation matrix [14].

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