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Originally published:

August 2018

Nuclear Technology 205(2019)1-2, 248-261

DOI: https://doi.org/10.1080/00295450.2018.1499324

Multiscaled experimental investigations of corrosion and precipitation processes after loss-of-coolant accidents in pressurized water reactors

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Number of pages: 23

Number of tables: 2

Number of figures: 17

Abstract

In a joint research project of the Zittau/Goerlitz University of Applied Sciences (HSZG), the Technische Universität Dresden (TUD) and the Helmholtz-Zentrum Dresden-Rossendorf (HZDR), the main emphasis is the time-related assignment of simultaneous and interacting mechanisms at zinc sources and zinc sinks at boundary conditions of a loss of coolant accident (LOCA) in German pressurized water reactors (PWR). The according experiments are carried out at semi-technical and at laboratory scale.

Zinc is used as a protective coating, e.g. for gratings in the containment, showing high corrosion resistance due to a gradual formation of passivating layers. In contrast, its long-term behaviour during LOCA changes significantly under the influence of the coolant chemistry of German PWR. As a consequence, according installations in the containment act as zinc sources. Released zinc ions change the chemical properties of the coolant and could e.g. lead to layer-forming depositions of zinc borates in the core, which increase the possibility of a hindered heat dissipation. For experimental and methodical investigations of these phenomena, the test rig “Zittau Flow Tray” (ZFT), a scaled sump model of a German PWR, was equipped with a full-length 3×3 fuel assembly (FA) dummy acting as core model, a preheater and a cooler component. Nine 4.4 m long fuel rod dummies simulate the decay heat by internal heating cartridges. This rig design enables experimental investigation of physico-chemical mechanisms considering coolant containing boric acid and zinc and their influence on the thermo-hydraulic processes in the reactor core at post-LOCA boundary conditions. Additional zinc corrosion and zinc borate precipitation studies to elucidate chemical zinc corrosion mechanisms and dependencies of those processes on typical LOCA parameters were carried out using lab-scale corrosion / precipitation test facilities.

The time depending zinc release at hot-dip galvanized gratings (HGG) was investigated regarding their position (e.g. inside or near the leaking jet, freely suspended or submerged in the coolant) and their surface area as well as temperature and flow rate of the coolant. The experimental database allows the approximation of corrosion rates in dependence of HGG position and the accident-specific coolant leakage rate as well as first mathematical approaches for the modelling of zinc sources.

Keywords - corrosion; zinc release; deposition; precipitation; particle formation

I. INTRODUCTION

During a LOCA in PWR the outflow leaking coolant is accumulated in the containment sump. Following the injection of the accumulators and from the emergency core cooling system (ECCS)-tanks the coolant recirculates in the long-term phase of a LOCA from the sump to the reactor and the leak driven by the low pressure ECCS pumps to remove decay heat from the core. In comparison to other reactor designs, German PWR are not equipped with containment spray systems. No alkaline substances are added during LOCA. The pH(25 °C) value of the coolant remains in the neutral to slightly acidic range, the latter due to the presence of boric acid [1]. During the flow from the leakage to the sump suction tubes in the containment, coolant gets in contact with containment installations. Parts of these installations are made of hot-dip galvanized steel, e.g. gratings, flight of stairs, inspection platforms, room divider and support grids of sump strainers (Fig. 1). Vertical cross sections of German PWR-Containments can be found in [2].

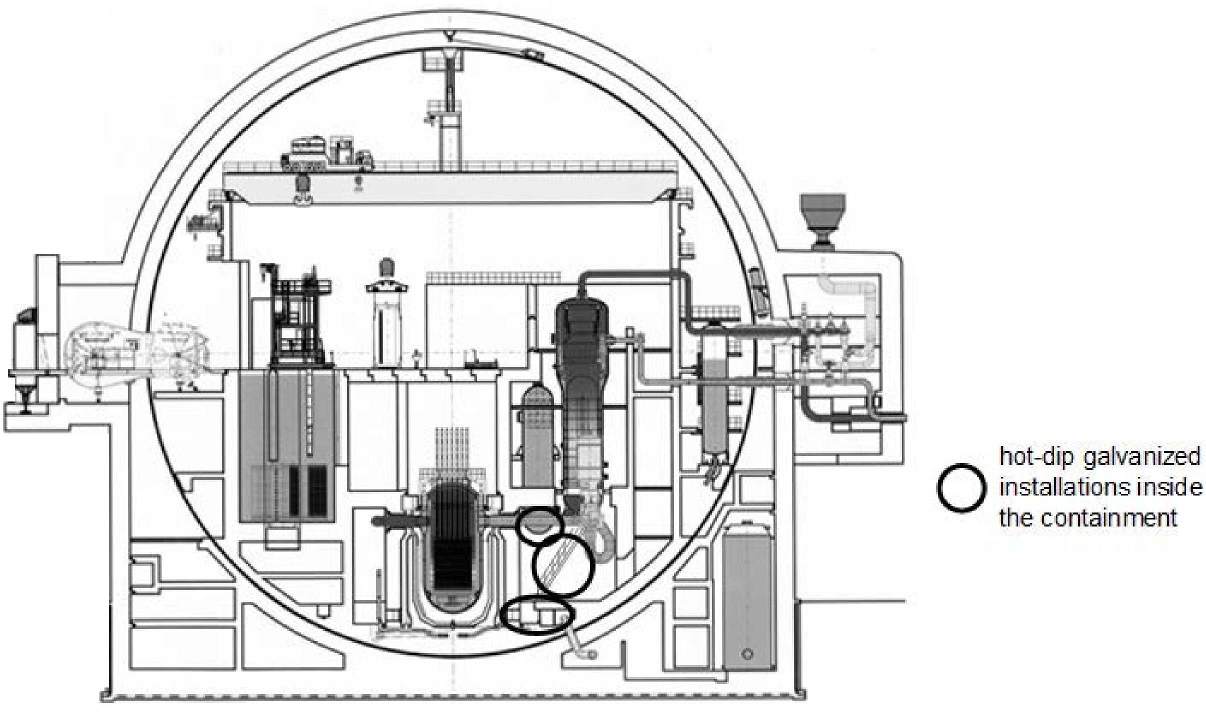


Fig. 1. Vertical cross section of a German PWR-Containment

Hence, corrosion occurs, a process causing a gradual destruction of surfaces in contact with fluids (e.g. air, water) over a longer period. Zinc is commonly used as protective coating for steel due to its relatively high corrosion resistance by the formation of passivating layers, but the long-term behaviour changes significantly by additional contact with several chemicals [3]. Looking into the containment of a German PWR, these corrosion effects affect this reactor type in particular, because the coolant can contain up to 2500 ppm boron in form of boric acid. The time frame, which has to be considered here, is significantly shorter until relevant effects can be observed because borated coolant supports corrosion processes at zinc-coated surfaces of internal PWR installations and leads to higher corrosion rates.

First investigations of zinc corrosion and its influence during a LOCA focused on the formation and deposition of solid corrosion products as one result of zinc release in boric acid solutions [4]. Furthermore, their behaviour in downstream components has been analysed. The experiments led to formation of dissolved zinc by corrosion and deposition of corrosion products at heatable zircaloy cladding tubes (3×3 design) of the test rig “Ring Line 2” (RL2) respectively at 56 heatable zircaloy cladding tubes of a shortened FA dummy. Both were connected to the ZFT as downstream components. Significant amounts of solid corrosion products were found in form of layer-forming depositions on the hot surface of the cladding tubes and the spacers as well as in form of sediments in pipes, at bottlenecks and other passive components. Chemical analyses at the TUD/HZDR identified these products as several types or even combinations of zinc borates. [5] summarizes the experimental results as a postulated incident scenario, which says that the deposits of zinc borates can led to

- blockage of the free mass flow at the strainers,
- reduction of the flow area between the fuel rods ($\leq$ 19 % reduction in the flow area of a cooling duct in the event of a leak size of 30 cm^2),
- deterioration of the heat transfer from the rod surfaces to the coolant (indicated by increasing temperatures inside the heating rods acting as fuel rod simulators during the experiments) as well as
- increase of the coolant temperature along the reduced flow channel in comparison to the unblocked channel.

Generic experiments at the FA dummy showed that the deposits could form a porous body inside flow channels. Hence, further heating can increase its dehydration. The investigation show that zinc borate precipitations (ZBP) can be an impending risk because it could lead to the loss of integrity of the fuel rods.

In [4] an outlook of upcoming experiments with boundary conditions and relations according to specific LOCA scenarios in a generic PWR is given. This paper is focused on these evaluations of boundary conditions and processes. Furthermore, first experiments considering zinc release and zinc borate precipitations for selected scenarios are presented.

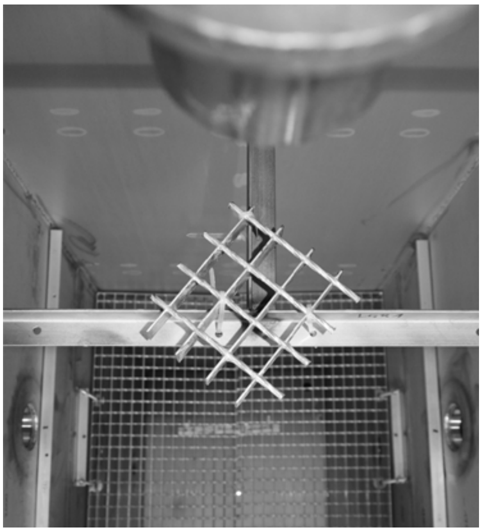
II. EXPERIMENTAL SETUPS

II.A. Experimental Setup at semi-technical scale (HSZG)

The experimental setup consists of a test facility combination representing both, the zinc source in the containment sump and the zinc sink in the reactor core. Therefore, the zinc release was realized in the "Zittau Flow Tray" (ZFT; Fig. 2-a), a scaled German PWR sump model with dimensions of 6.0 m×3.0 m×1.0 m (L×W×H). It allows a wide variety of positions and sizes of HGG as zinc sources (Fig. 2-b). The leakage jet can be simulated in a free-fall section, which also allows variations of leakage position, volume flow and spraying pattern [4, 6].

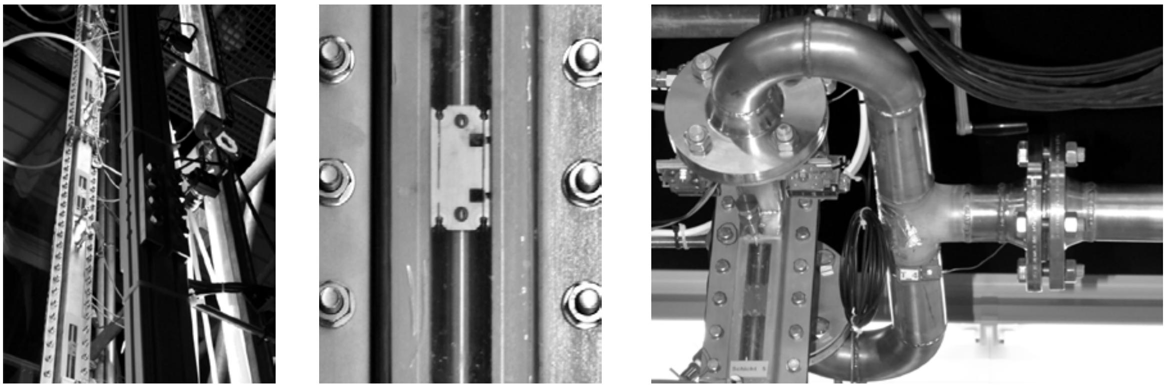
Different downstream components can be connected with the ZFT. Previous experiments were performed at the RL2 with 3×3 heating rod configuration and a shortened heatable FA dummy [5]. Both were equipped with heating cartridges of a constant heating power over a length of 580 mm. For the current investigations, a new core model named CORVUS was built up. This test facility consists of PWR fuel rod cladding tubes with original length of 4.4 m in a 3×3 design (Fig. 3-a).

(a)



(b)

Fig. 2. Sump model “Zittau Flow Tray” (a) in exterior view (b) with HGG placed under a nozzle



(a) (b) (c)

Fig. 3. Test rig CORVUS (a) view from bottom to top (b) observation window with a visible spacer segment and (c) two-way outlet at the upper end

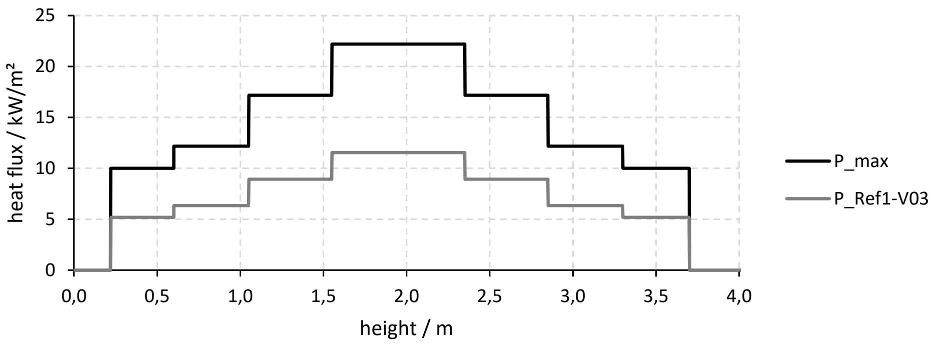


Fig. 4. Power profiles (maximum profile and profile applied at experiment no. CORVUS-Ref1-V03) of the fuel rod dummies of the test facility CORVUS

Hence, the original height allows the installation of nine spacer segments as an original fuel assembly of German PWR (Fig. 3-b). Inserted heating cartridges simulate the decay heat over the active length of fuel rods. The fuel rod cladding tubes of CORVUS have a stepwise power profile to emulate a cosine axial power distribution of real fuel rods. The maximum values of the power profile over the height are shown in Fig. 4. The components of the test facility are semi-technical scaled or rather given in a ratio that allows conclusions of experimental results that meet the conditions in real plants. An upstream preheater and a downstream cooler allow the adjustment of the coolant temperatures due to a sump-core recirculation during a LOCA.

II.B. Experimental Setup at laboratory scale (TUD/HZDR)

Long-term lab scale experiments, which included formation and deposition of corrosion products at heated PWR cladding tubes, were carried out in a modular test facility named PETrA (see Fig. 5). This facility represents the ECCS operation during sump recirculation after a PWR LOCA in a very simplified manner. It mainly consists of a spray section (representing the leak), a bath section as coolant reservoir (representing the sump) with a liquid volume of about 60 litres including a heat exchanger with thermostat to heat up the coolant to a defined temperature and finally a heating module (HEM), representing the fuel rods. One loop transfers the coolant from the sump through the HEM, consisting of an electrically heated zircaloy-4 tube (diameter: 10.75 mm, heated length: 1000 mm) with a surrounding glass tube, where the annular gap between the heating rod and the glass tube represents a single channel of a fuel element. For pure zinc dissolution experiments (without deposition of corrosion products), the lab-scale corrosion test facility KorrVA was used (PETrA facility without HEM, see Fig. 5).

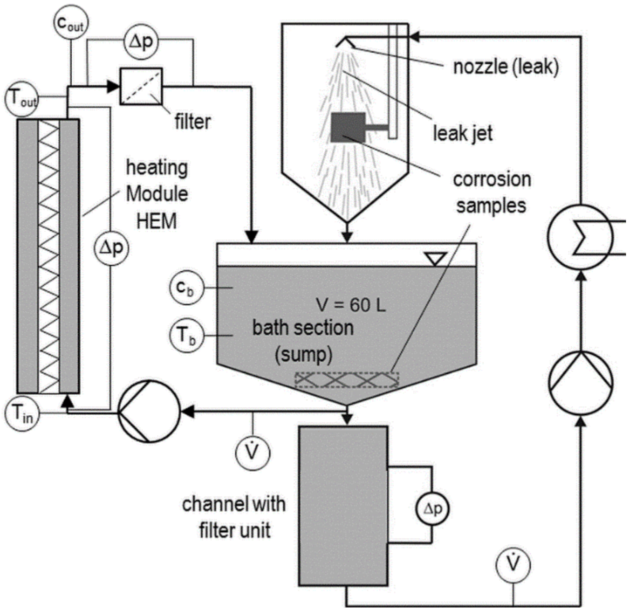


Fig. 5. Scheme of the PETrA facility to simulate the zinc dissolution and subsequent zinc borate precipitation at hot cladding tubes including zinc dissolution unit (KorrVA) as part of the PETrA facility without the heating module HEM [7]

To simulate the zinc dissolution during the sump recirculation period, zinc sheets of suitable sizes were placed inside the spray jet (spray section) or immersed into the liquid (coolant) of the bath section. Online measurements of the coolant temperatures (bath section, inlet and outlet of HEM) and of the electrical conductivities (EC) inside the bath section have been executed during the experiments. More detailed descriptions of the plant components are given in [8] and [9].

III. ZINC RELEASE EXPERIMENTS

III.A. Downscaling

For downscaling, some LOCA scenarios considering a small break (leak size: 30 cm²) or a large break (leak size: 442 cm²) at different points in the cycle as well as various boundary conditions were analysed and evaluated. To reduce the parameter variety, a number of selection criteria for the calculated LOCA scenarios were defined including probability of ZBP in the core, exclusion of LOCA scenarios with cross mixing and transverse flow effects due to their non-presentability by experiments, data redundancy and technical limitations.

The approximation of the resulting coolant chemistry, the classification and determination of involved zinc inventory (see Table I) was done in cooperation with the Gesellschaft für Anlagen- und Reaktorsicherheit (GRS), power plant engineering and operating companies [6]. Based on these results, the scale-down of

- the total area of zinc-coated surfaces,
- the quantity of circulating coolant during LOCA and
- the chemical specifications like boron/lithium concentrations and pH value

for the parameterization of the experiments were realised.

TABLE I

Categories (C) and approximated area (A_{PWR}) of zinc-coated surfaces inside a generic PWR containment

C	**Position**	A_{PWR}
(1)	stairs and platforms above coolant level and in reach of the leakage jet	4 – 25 m²
(2)	submerged stairs and platforms	330 m²
(3)	support grids at sump strainer	450 m²
Overall corrosion surface area		784 – 805 m²

Simulations of different LOCA scenarios with the thermal-hydraulic system code ATHLET (Analysis of thermal-hydraulics of leaks and transients) performed by the GRS provided temperature profiles and flow properties in the PWR core. The flow rates considered for downscaling are

- 400 m³/h for the leak size of 30 cm² and
- 1900 m³/h for the leak size of 0.1 F (442 cm²), where F stands for the open cross-sectional area of the leaking coolant line.

The resulting concentrations of boron and lithium vary due to the considered LOCA scenario. In coordination with the GRS, the coolant chemistry was slightly adapted in comparison to older experiments. Current and future experiments are performed with 2000 ppm boron and 0.2 ppm lithium (instead of 2300 ppm boron and 0.5 ppm lithium as stated in [4, 6]).

A function for the correlation between conductivity $\kappa_{25°C}$ and zinc concentration β_{Zn} was necessary. Eq. (1) is such a correlation for the given coolant chemistry and was empirically determined by TUD/HZDR.

$$\beta_{Zn} = 0{,}00004 \cdot \kappa_{25°C}^2 + 0{,}424 \cdot \kappa_{25°C} - 1{,}7607 \tag{1}$$

The online measurement of the conductivity κ_T at a given temperature has to be converted in an equivalent value at 25 °C by Eq. (2).

$$\kappa_{25°C} = \frac{\kappa_T}{1 + \frac{k}{1413} \cdot (T - 25°C)} \tag{2}$$

To back up the results, the conductivity was measured for samples at 25 °C and the zinc concentration was determined for these samples by chemical analysis at the HZDR. The quotient $k' = {}^{k}/_{1413}$ in Eq. (2) is a correction factor depending on the coolant chemistry. For 2000 ppm boron and 0.2 ppm lithium k' is $\approx$ 0.02467.

III.B. Results at laboratory scale

Series of generic zinc corrosion experiments were carried out in the lab-scale test facility KorrVA (PETrA facility without HEM, see Fig. 5) using zinc sheets as corrosion samples. Zinc corrosion rates were calculated from the time-dependent increase of the zinc concentration in the coolant. Time-related zinc concentrations of the coolant ($ß_{Zn}$ in mg/L) were determined via the online measured electrical conductivities (EC) or via coolant samples using inductively coupled plasma mass spectrometry (ICP-MS).

The aim of these generic KorrVA experiments was to investigate the influences of different boundary conditions (hydrodynamics, coolant chemistry, temperature) on the zinc corrosion rates. The main results of the investigations can be summarized as follows:

- A strong influence of the hydrodynamic impact of the coolant jet onto the corroding surface on the zinc corrosion rates was observed. A zinc dissolution experiment with a zinc sheet (15 × 15 cm²) placed in the spray section of the KorrVA led to a corrosion rate of v_{Zn} ≈ 7900 mg/(m² h) whereas in the most other experiments the typical corrosion rates of zinc sheets dipped in the bath section of the KorrVA (slowly stirred coolant) where in the range between 1500 and 2500 mg/(m² h). Similar effects were observed in the experiments at semi-technical scale (see chapter III.C).
- Particular flow conditions around submerged zinc surfaces had a strong influence on the zinc corrosion rates. Small variations in laminar flow rates parallel to the zinc surface did not lead to significant changes, but an increase of cross flows or turbulences nearby the zinc surfaces led to significant higher corrosion rates.

- Variations of the coolant temperature in the range between 25 °C and 70 °C as well as small changes in boric acid and in lithium hydroxide concentrations (range between 0 – 2 ppm Li, resulting pH range 4.7 to 6) had no significant influence on the corrosion rate during the first stage of the corrosion process.
- After a temperature-dependent period, the corrosion rates showed an abrupt decrease during all generic corrosion experiments. The fact that the decrease of the corrosion rates occurs at lower zinc concentrations with increasing temperatures indicates the formation of corrosion inhibiting surface layers. Probably these layers mainly consist of (different) zinc borates due to their decreasing solubility with increasing temperature (reversed solubility behaviour).

A more detailed description and discussion of these results is given in [9].

Additional zinc release experiments were carried out in the KorrVA lab facility with the aim to reproduce courses of zinc concentration profiles, which correspond to particular LOCA scenarios, in order to define the experimental conditions for the subsequent zinc borate precipitation experiments (see section IV.A). The underlying zinc concentration profiles had been evaluated before by downscaled semi-technical experiments of the HSZG (compare section III.C) to be representative for LOCA scenarios with a small leak of 30 cm^2 (Ref 1) or with a 0.1 F leak (Ref 2). A typical result of such a zinc release experiment is shown in Fig. 6. Two zinc sheets (1.6 mm thickness) placed in the upper part of the bath section were used. The main zinc sheet (8.3 x 15 cm^2) was immersed during the whole testing time whereas the smaller zinc sheet (2.8 x 10 cm^2) was taken out of the bath section after 48 h. As coolant, 74 L boric acid solution (11.3 g/L boric acid corresponding to 2000 ppm boron) and 0.2 ppm lithium (as lithium hydroxide) was used. All experiments were carried out at isothermal conditions at a coolant temperature of 45 °C.

Fig. 6 shows, that the zinc concentration course of LOCA scenario Ref 1 (30 cm^2 leak) obtained at semi-technical scale (curve ZSW_LS_V010 - see also section III.C) has been well reproduced in the laboratory experiment (KorrVA) by using two different zinc sheets (zinc concentrations evaluated via ICP-MS (curve Ref 1 (KorrVA) / ICPMS) and via EC (curve Ref 1 (KorrVA) / EC)). It is also remarkable that, as before in the generic zinc corrosion experiments, the zinc corrosion rates significantly decreased after obtaining higher zinc concentrations (kink in the curve at about 50 h).

In all lab-scale zinc release experiments, area-related corrosion rates in the range of 1500 2500 mg/(m^2 h) were identified using slowly stirred boric acid solutions in the bath section of KorrVA. Furthermore, the zinc dissolution causes an increase of the pH value of the coolant towards the neutral region (see pH increase from 5.2 to 6.9 in Fig. 6).

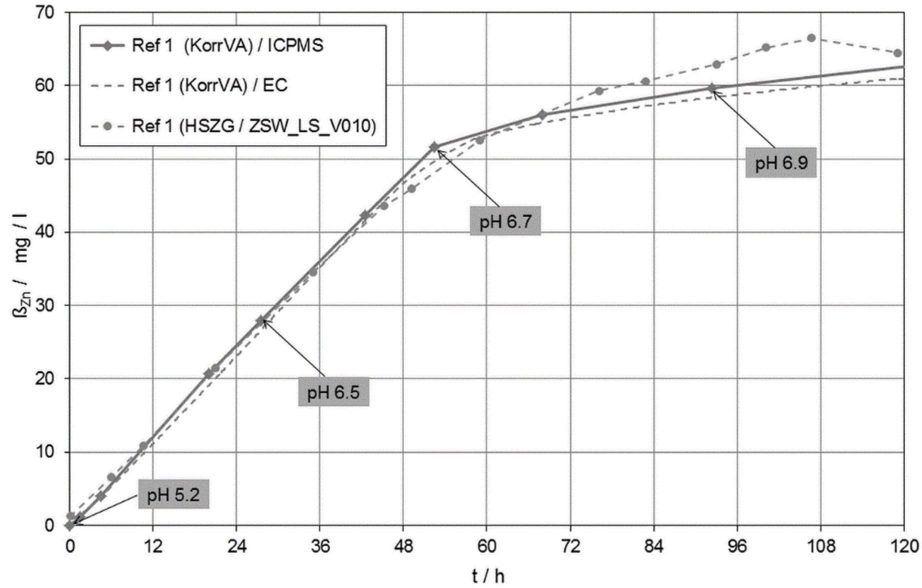


Fig. 6. Results of a zinc corrosion test in the KorrVA facility to reproduce the course of the zinc concentration for the LOCA reference scenario Ref 1 (leak of 30 cm²). The experiment was carried out at 45 °C with two zinc sheets in the bath section

III.C. Results at semi-technical scale

The LOCA boundary conditions were transformed into multiple scales for experimental parameterization. The factor $\tau = V_{PWR}/Q_{leak}$ with V_{PWR} as the amount of circulating coolant inside a generic PWR during LOCA and the presumed leakage rate Q_{leak} in m^3/h specifies the time needed for a complete coolant circulation. With V_{ZSW} as the given coolant volume in the ZFT test rig, the volume flow of the scaled leakage jet results from $Q_{ZFT} = V_{ZFT}/\tau$. The scaled zinc surface A_{ZFT} for each category C of Table I is calculated by Eq. (3).

$$A_{ZFT}(C) = \frac{A_{PWR}(C) \cdot V_{ZFT}}{V_{PWR}} = \frac{[m^2] \cdot [m^3]}{[m^3]} \quad (3)$$

Fig. 7 illustrates the configuration of the HGG in the ZFT for the zinc release experiments ZSW-LS-V09 and ZSW-LS-V10. The HGG of category C1 is 385 mm in V09 respectively 1197 mm in V10 below the simulated leaking jet. The HGG of category C2 is placed 200 mm below the water level and C3 in front of a 2 mm × 2 mm strainer.

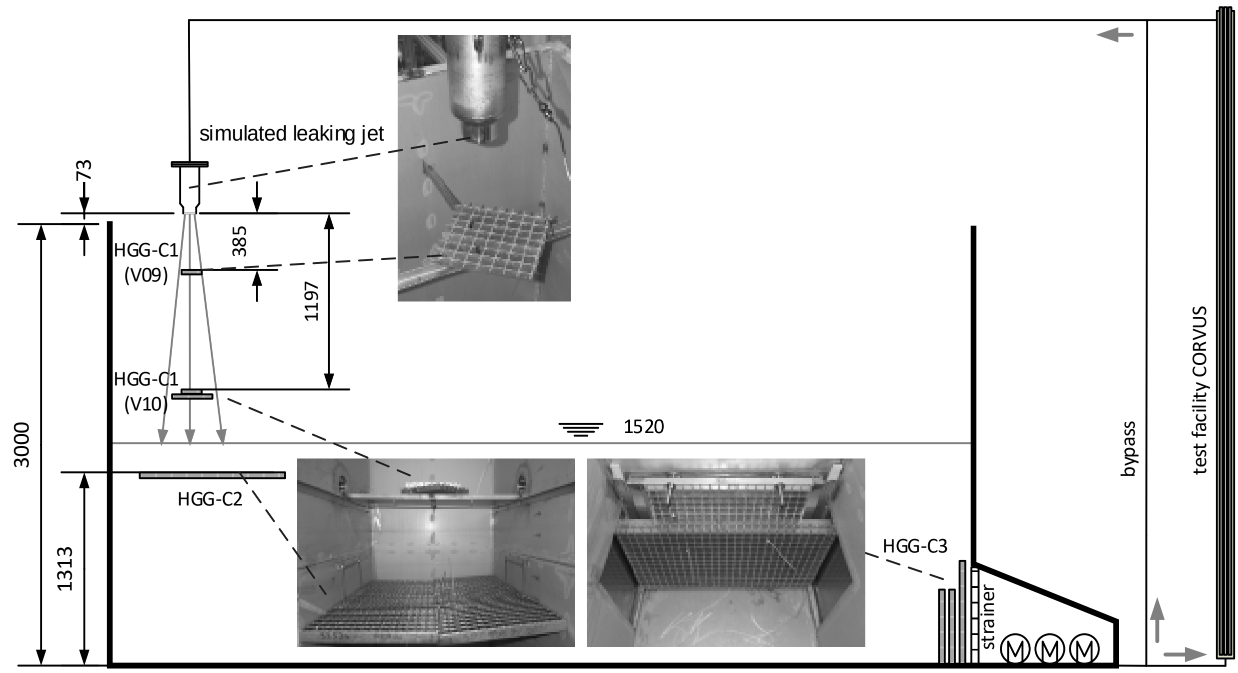


Fig. 7. Scheme of the test facilities ZFT and CORVUS and the positions of HGG in the experiments ZSW-LS-V09 and -V10

The parameters for the experiments ZSW-LS-V09 and -V10, the considered coolant chemistry and zinc inventory are listed in Table II.

TABLE II

Parameter of zinc release experiments ZSW-LS-V09 and -V10

experiment	**zinc inventory (m^2)**			**B conc.**	**Li conc.**	$\boldsymbol{Q}$	$\boldsymbol{T_{ZFT}}$
no	**C1**	**C2**	**C3**	**ppm**	**ppm**	**m^3/h**	**°C**
V09	0.044	3.12	4.51	2000	0.2	3.9	45
V10	0.26	3.12	4.51	2000	0.2	3.9	45

Zinc release was determined over 120 h. The measured conductivity values at 25 °C and pH values are shown in Fig. 8. In both experiments, the conductivity κ increases with a comparable course to value of 160 µS/cm after 120 h of experimental time. During the zinc release the pH values increase from 5.0 respectively 5.25 up to 6.5.

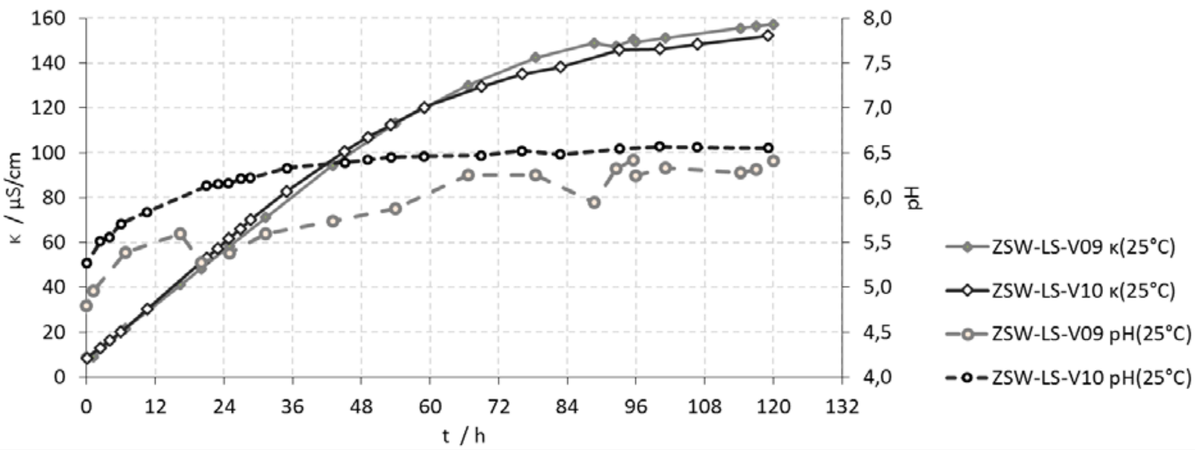


Fig. 8. Course of conductivity κ and pH value in ZSW-LS-V09 and -V10

The pictures of the HGG in the end of experiment ZSW-LS-V09 illustrate the impact on their surfaces (Fig. 9). Corrosion processes affect especially HGGs near the leakage jet due to the flow properties and the presence of air. The higher corrosion rates at these HGG are indicated by their high mass loss and the early onset of iron rust formation, which comes along with brownish discolorations at the HGG surface. Analyses of the layer structure reveal a removal of zinc layers down to the basic steel material. The zinc layer of the HGG above the water level is almost completely removed. The slow flow near the sump strainer results in lower corrosion rates. After 120 h, the hard zinc layer and for the most part the pure zinc as external layer are still present at the HGG in the sump strainer area, leading to a low but constant zinc release over days.

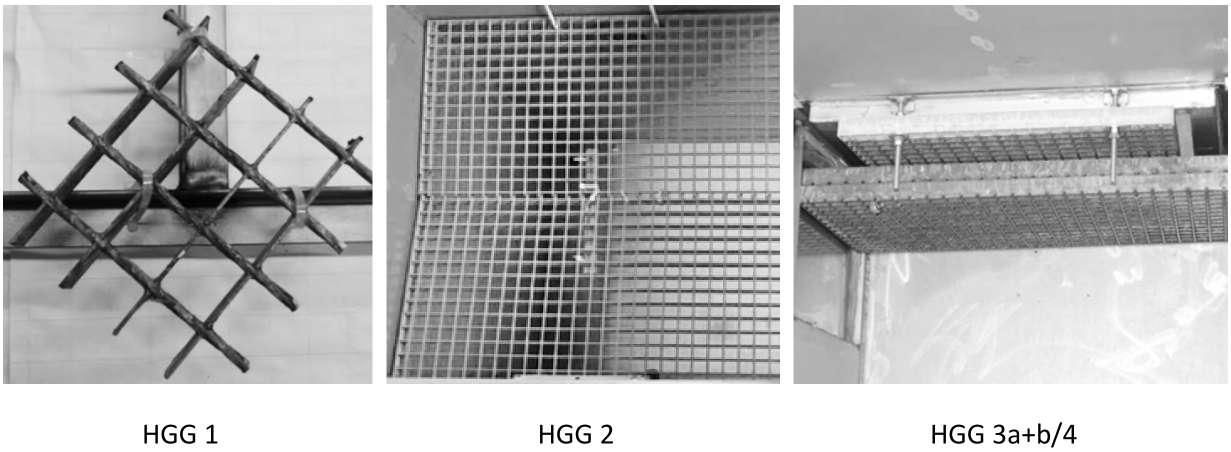


Fig. 9. HGG in the end of experiment no. ZSW-LS-V09

Fig. 10 shows the courses of the zinc concentration in the experiments ZSW-LS-V09 and ZSW-LS-V10. The online results are calculated by Eq. (1). The supporting points result from sample analysis with inductively coupled plasma mass spectrometry and confirm the conversion equation.

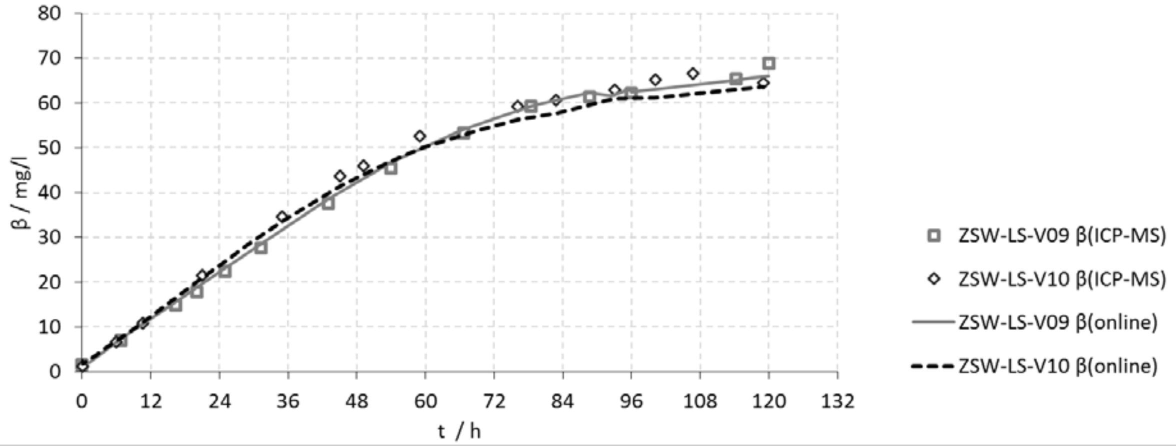


Fig. 10. Course of zinc concentration in the coolant in the experiments no. ZSW-LS-V09 and -V10

The course and the end values indicate a negligible influence of the larger zinc surface in the leakage jet so far. A possible influence caused by the different distances between HGG and leakage jet nozzle cannot be excluded. The final zinc concentration after 120 h was 68.8 mg/L in V09 and 64.5 mg/L in V10. These values correspond well to 0.667 kg respectively 0.626 kg dissolved zinc in the test facility after the zinc release experiments based on HGG mass loss. Based on the measured zinc concentration the corrosion rate v_{Zn} is calculated by

$$v_{Zn}(t) = B(t) \cdot \frac{V_{ZSW}}{A_{corr}} = \left[\frac{mg}{l \cdot h}\right] \cdot \frac{[l]}{[m^2]} \tag{4}$$

with the release rate $B(t) = \left(\beta_{\mathrm{Zn}}(t_2) - \beta_{ZN}(t_1)\right)/\Delta t$.

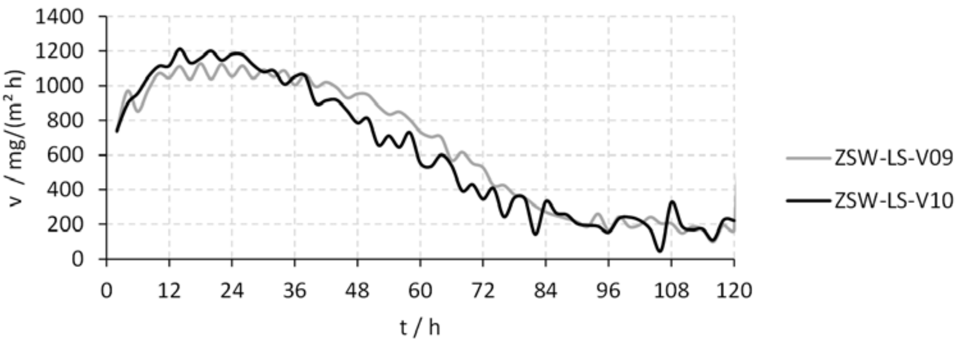


Fig. 11. Course of the corrosion rate in the experiments no. ZSW-LS-V09 and -V10

The course of the corrosion rate (Fig. 11) can be differentiated into three parts: Relatively high release rate in the first 36 h with values around v_{Zn} = 1100 mg/(m² h). After 36 h, the rate decreased down to $v_{Zn} \approx$ 200 mg/(m² h) at around 84 h. From that time, the rate kept nearly constant.

IV. ZINC BORATE PRECIPITATION EXPERIMENTS

IV.A. Results at laboratory scale

Simultaneous zinc corrosion / zinc borate (ZnB) precipitation experiments at laboratory scale using the PETrA test facility (see Fig. 5) were carried out at typical LOCA boundary conditions for the 30 cm² leak size (Ref 1) as well as for the 0.1 F leak size (Ref 2). Some main parameters of these experiments (e.g. temperature courses for the bath section / sump and of the spray section) were chosen according to the ATHLET data for the corresponding PWR LOCA situations (see temperature courses in Fig. 12), other parameters (e.g. coolant chemistry) were determined according to the described estimations for the downscaling from the PWR to smaller dimensions (see chapter III.A).

The results indicated significant formation of solid zinc borates as deposits onto hot surfaces of the heating unit (HEM) and simultaneously as particles inside the boric acid solutions. The boundary conditions related to a LOCA scenario with a small leak (30 cm² / Ref 1) led to an earlier start of zinc borate precipitation as well as to higher ZnB precipitation rates compared to those of a scenario with a larger 0.1 F leak (Ref 2).

Time-averaged ZnB precipitation rates (relating to the deposited mass of zinc) have been calculated by the mass losses of the used zinc sheets minus the amount of dissolved zinc at the end of the experiments. Time dependent ZnB precipitation rates have additionally been calculated based on the difference of the zinc concentrations between the sump (inlet HEM) and the outlet of the HEM.

Furthermore, ZnB precipitations inside the coolant (flocks and particles) were collected in a filter unit behind the outlet of the HEM. The amounts of these mobile ZnB precipitations were evaluated by weighing, and additionally their zinc content was determined using ICP-MS after dissolution of the ZnB in hot citric acid. After each experiment, also the zinc mass of the immobile ZnB inside the heater unit (layer onto cladding tube of HEM) was quantified using ICP-MS.

In Fig. 12 and Fig. 13 essential boundary conditions respectively results of two ZnB precipitation experiments are shown, which were related to the LOCA scenario Ref 1 (30 cm² leak). Fig. 12 shows the coolant temperature courses (sump and outlet HEM) for two ZnB precipitation experiments in comparison to the corresponding calculated ATHLET data. The temperature course of Ref 1+ was used in one experiment with temperatures ($T_{outlet,HEM}$) about 5 K higher than the calculated coolant temperature course inside the hot bundle using ATHLET, and the temperature course of Ref 1- was used in another experiment with temperatures ($T_{outlet,HEM}$) about 5 K lower than calculated ATHLET data. The shown course of the sump temperature was used for both experiments (see Fig. 12), and the simultaneous zinc dissolution using two corroding zinc sheets in the bath section of the KorrVA (sump) was similar as described in Fig. 6 (section III.B).

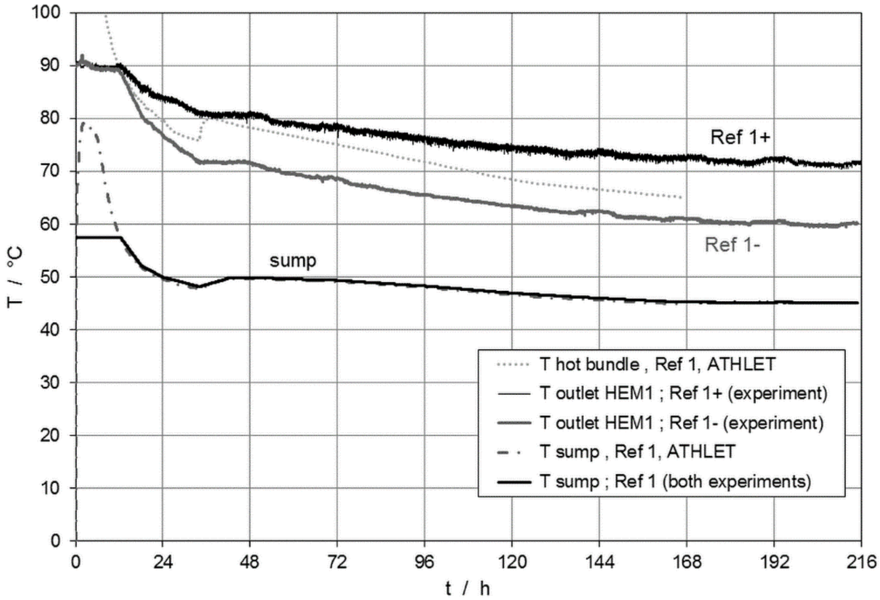


Fig. 12. Temperature courses (sump and outlet HEM1) for two ZnB precipitation experiments with boundary conditions related to the LOCA scenario Ref 1 (30 cm² leak)

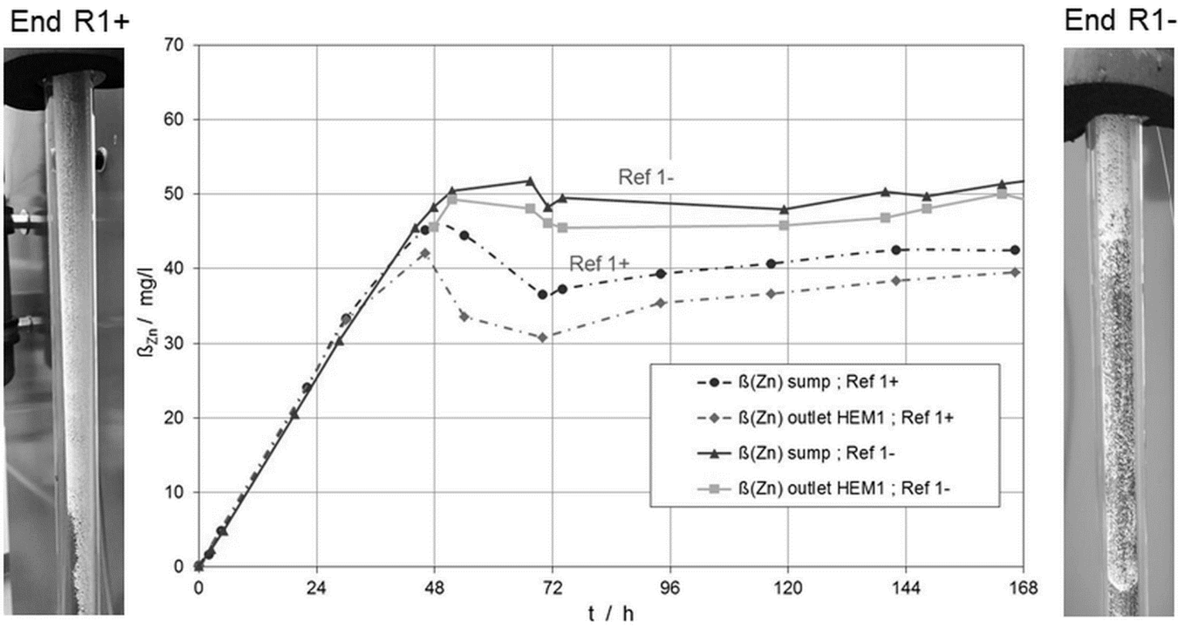


Fig. 13. Courses of the zinc concentrations in the sump and at the outlet of the HEM for experiments related to the Ref 1 scenario and photographs of ZnB depositions on the surfaces of hot cladding tubes of HEM after the experiments

The main results of these two experiments can be described as follows (see Fig. 13): As seen from the photographs, significant ZnB precipitations inside the HEM have been observed in both experiments, where the precipitation rate in case of Ref 1+ seems to be significantly higher compared to Ref 1-. Also, the difference of the zinc concentrations between the inlet ($\beta_{Zn,sump}$) and the outlet ($\beta_{Zn,outlet}$) of the HEM indicated precipitation rates in the Ref 1+ experiment nearly twice as much compared to those of the Ref 1- experiment. In both experiments, the ZnB precipitation started after about 48 h, visible in the kink of the zinc concentration courses (beginning decrease of concentrations of dissolved zinc ions) in Fig. 13.

The average precipitation rates (zinc mass from overall ZnB) were 30.2 mg/h for the Ref 1+ experiment and 16.8 mg/h for the Ref 1- experiment. In both cases, about one third of the ZnB precipitated as layer (immobile ZnB) on the cladding tube, whereas the rest precipitated inside the fluid as particles or flocks and were consequently retained in the downstream filter unit.

Summarizing, in most of the ZnB precipitation experiments related to the boundary conditions of the LOCA scenarios Ref 1 (30 cm² leak) and Ref 2 (0.1 F leak), the ZnB precipitations inside the HEM started after about 48 h and 92 h, respectively. In all cases of ZnB precipitations, simultaneous precipitation inside the fluid (particles, flocks) and as layer on the hot surface of the cladding tube (HEM) occurred. The ratio of mobile to immobile ZnB depends on the temperatures of the coolant and of the surface (cladding tube). There was a trend observed towards an increasing ratio of immobile ZnB (layer formation) with increasing temperature inside the HEM in relation to the formation of mobile ZnB in the fluid. Typical overall precipitation rates (HEM) for these experiments were in the range of 20 - 40 mg/h (calculated as zinc mass).

IV.B. Results at semi-technical scale

Due to the need of information to the zinc release rates for plant near configurations the experiments for ZBP were performed subsequently. Hence, the decay heat was calculated for t = 120 h after begin of the LOCA.

The experiment CORVUS-Ref1-V03 continued the zinc release experiment ZSW-LS-V10. The courses of the main parameters are shown in Fig. 14. The flow rate Q_{Gesamt}, the temperature at the Inlet T_{ECV} and the heating power P_{Ges} were constant over time. The power of 8.9 kW is equal to 53 % of the heating rod power profile (see Fig. 4). The mean value of the flow rate Q_{Core} is 0.22 m³/h and corresponds to a speed of 0.05 m/s between the tubes in the core model CORVUS. For comparison, the coolant temperature curves in the DWR core, corresponding to the ATHLET simulation results, are also shown.

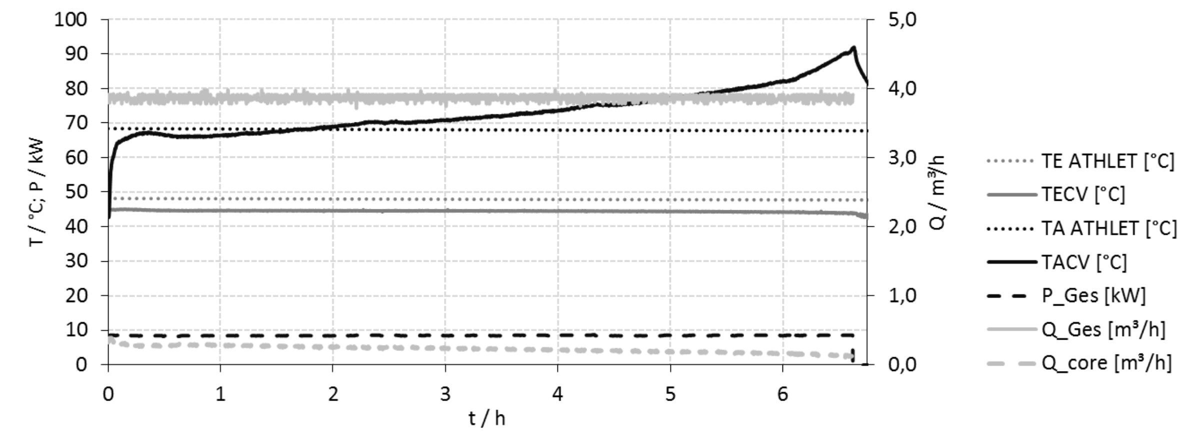


Fig. 14. Coolant temperatures at CORVUS inlet (TECV) and outlet (TACV), volume flows and total heat output during experiment no. CORVUS-Ref1-V03

The temperature at the outlet T_{ACV} increased up to 83 °C despite nearly constant parameters. The mass distribution of the precipitations at spacers (S1-S9) and intermediate rod areas (R1-R10) is shown in Fig. 15. The mass ratio is plotted based on the founded mass (grey - mg) and additionally calculated in relation to the length of the corresponding rod section respectively spacer (black - mg/mm). From the lower rod end up to spacer 4 (S4), no deposits were observed. Above S5 the deposit increases significantly. Nearly 90 % of all ZBP was found in the range between S6 and S8.

Fig. 16 shows the according parts of the heating rod configuration in dried state after the experiment CORVUS-Ref1-V03. The head loss was measured over all nine spacers. The measured value is the total of hydrostatic and dynamic pressure. The hydrostatic head loss is the result of the temperature difference between the water in the flow channel and in the impulse line of the measurement system.

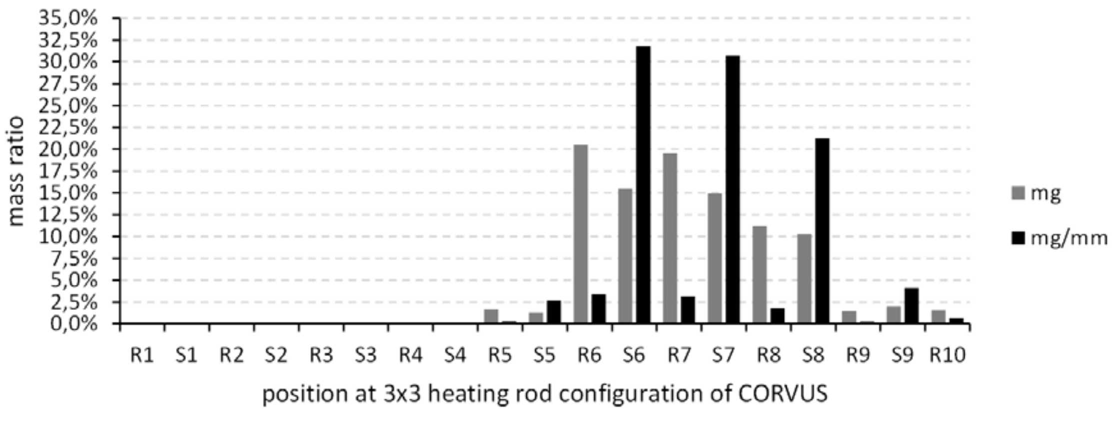


Fig. 15. Mass distribution of ZBP in upward flow direction at rod segments (R) and spacers (S) of CORVUS in experiment no. CORVUS-Ref1-V03

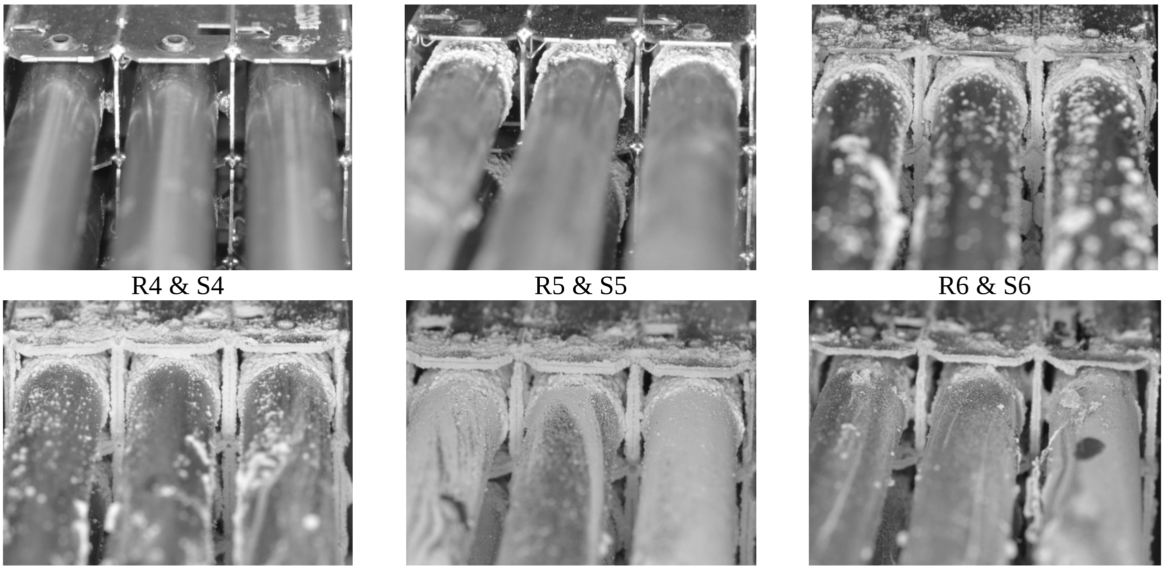


Fig. 16. Deposits at the spacers S4 to S9 after experiment no. CORVUS-Ref1-V03

Hence, the head loss decreases after experiment start due to the increasing temperature over the height of the test section. To get information on the head loss change due to the deposits at tubes and strainers the head loss of the free test section had to be calculated for expected range of boundary conditions (temperature, volume flow). This included the calculation of the head loss proportion as a static and as a dynamic variable, considering pipe friction over pipe length ($lambda$) and local head losses at spacers ($lokal$). Therefore, the main fluid parameter and the flow characteristics (laminar or turbulent) of the test section were determined based on results of preliminary experiments.

Possible influences of the channel in- and outflow patterns were not taken into account. The results were verified by ATHLET simulations of the test section. Based on these information, the pressure loss coefficient of the spacers was calculated with the measured head loss (back calculation).

The courses of the measured and calculated head losses are shown in Fig. 17. $PDGES$ is the measured overall head loss over the test section with the nine spacers. The determined dynamic part of $PDGES$ is $PDGES\_dyn$. The course of the calculated overall head loss for the test section without agglomeration and the given boundary conditions is DP_CGES. Its dynamic ratio DP_CGES_dyn consisting of the pipe friction DP_CGES_lambda and the local head loss at spacers $DP\_CGES\_dyn\_lokal$. This difference between the measured (during zinc borate precipitation) and the calculated (free test section) proportion of the dynamic head loss is the ratio caused by deposits in the test section.

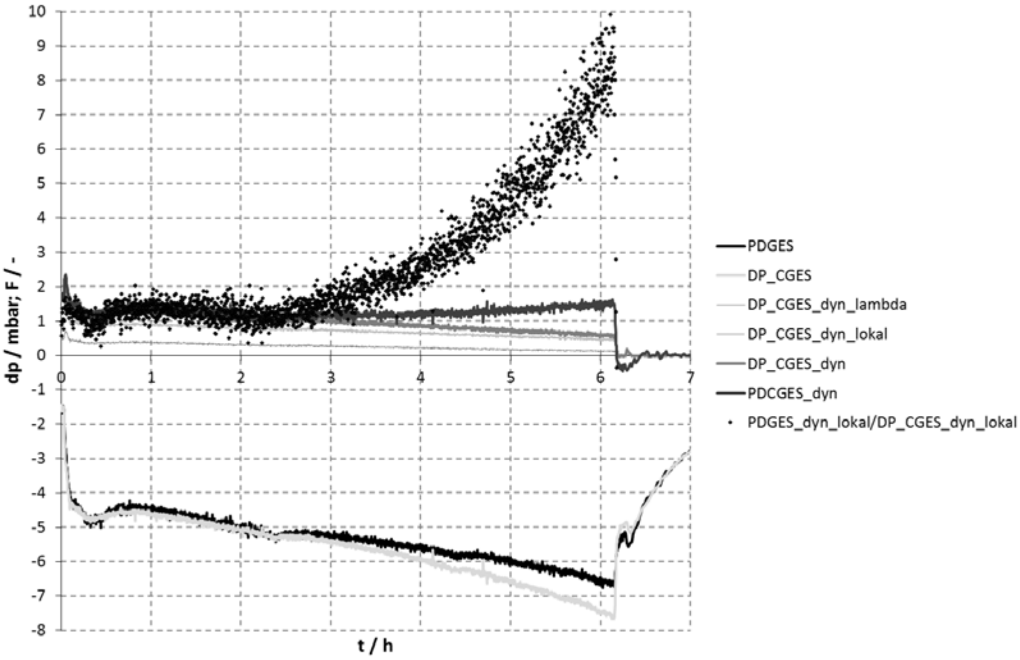


Fig. 17. Measured and calculated head loss in experiment CORVUS-Ref1-V03

A factor F is introduced to specify the accumulation at the spacers and rod. F is the ratio between the measured and calculated (free test section) local head loss at spacers by the assumption that all additional head losses are caused by the accumulation (local dynamic).

The course of F ($PDGES\_dyn\_lokal/DP\_CGES\_dyn\_lokal$) for experiment CORVUS-Ref1-V03 is shown in Fig. 17. At the start of the experiment $F = 1$ as expected for the free channel. After $t_{Exp} = 3$ h F starts to increase, indicating the time of first precipitations in the test section. After $t_{Exp} = 7$ h the head loss of the free flow in the channel increased in experiment CORVUS-Ref1-V03 by the factor 8 to 9 due to the zinc borate agglomeration. Hence, the results show that the back calculation provide a parameter that allows a valuation and time behaviour visualization of the precipitation.

V. Conclusion

Experiments concerning corrosion effects at technical facilities should always be considered to be analyzed at several scales. The analyses of LOCA scenarios and the definition of real plant conditions in cooperation with GRS, power plant engineering and operating companies allow a realistic simulation of the processes of zinc release and the precipitation at hot spots in the reactor core.

The performed experiments led to the conclusion that there is a potential of ZBP in the core under the adjusted LOCA boundary conditions. Furthermore, the general statements on corrosion effects at metal surfaces are proven for both the lab and semi-technical scale with remarkably higher corrosion rates than under atmospheric conditions [9]. The corrosion processes at zinc-coated surfaces of internal PWR installations by borated coolant lead to zinc concentrations up to 70 g/L after 120 h of the sump recirculation operation after LOCA. First experimental results at CORVUS considered near-LOCA core temperature courses. According results show that the occurring amounts of zinc borate influences the flow conditions and local temperatures in the core simulator.

VI. Outlook

An important difference of the upcoming experiments has to be the analysis of the temperature gradient. Up to now, the experiments with CORVUS as a core simulator had started at sump temperature of 45 °C due to the need of realistic release rates without a zinc sink. During the course of the experiments, the temperature in the test section was increased by the heated rods. Hence, the temperature regime was from “cold to hot”. Experiments had shown that the simulation of the high sump temperatures at the start of the recirculation are not crucial for the zinc release. However, the impact of the layer formation of the different kinds of zinc borates and their temperature related precipitation

- Flocculent corrosion products (CP) with the ability to form very sparse and highly porous layers at hot FRS surfaces (formation started in a range of coolant temperature of 60 - 70 °C)
- Crystalline CP with the ability to form very dense layers at hot FRS surfaces (temperature range of 70 - 80 °C) [10]

has to be considered in the temporal sequence of experiments. For the regime “hot to cold” the dense layers of crystalline CP will already be present when flocculent CP will occur. In the past experiments at the RL2 with one spacer, parts of the flocculent CP were transported out of the test section. At the configuration with original length, the possibility of agglomeration at one of the nine spacers has to be evaluated. Furthermore, the crystalline CP at the spacers will limit the removal out of the test section.

Furthermore, combined experiments at CORVUS and the partially heatable FA dummy are planned to investigate the axial and radial precipitation behaviour of zinc borates and the influence of deposits on possible flow changes. Different heated zones in the FA dummy should show if various kinds of zinc borate could precipitate at the same stage of LOCA. For the longer term, measurements of heat fluxes at layer-forming ZnB on the cladding tubes are planned.

ACKNOWLEDGMENTS

The reported investigations are funded by the German Federal Ministry for Economic Affairs and Energy (BMWi) under grant no. 150 1491 and 150 1496 on the basis of a decision by the German Bundestag. The responsibility for the content of this publication lies with the authors.

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