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Division I

Cracks in concrete containment – Comparing the self-healing potential under variable air and steam loads between real scale and laboratory scale

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ABSTRACT

The leak tightness of nuclear power plant containments under accidental conditions is a crucial topic in the safety discussion about the operating lives of nuclear power plants. The self-healing potential of cracks in concrete thereby might contribute an important share to avoid possible contamination of the environment. Small scale flow-through experiments with air and steam, conducted under variable pressure and temperature conditions at the MPA Karlsruhe shall gap the bridge between previously performed large scale leakage tests and the sole description of the relevant and already well known chemical and physical processes. The tests were conducted on specimens of cementitious construction materials, i.e. concrete and typical concrete ingredients (sandstone, limestone, granite) to separate possible effects by the material. Experimental conditions were chosen according to realistic temperature and pressure conditions of an accidental scenario. The experimental setup shall first of all measure the amount of flow through the crack and subsequently its reduction accompanying the different crack healing processes. For the assessment and quantification of the mineralogical crack healing processes, the crack surfaces were investigated by ESEM, µ-RFA and microscope. Within this paper, the first results were summarized and compared with results from the large scale experiments. A detailed description of the experimental setup is given as well.

INTRODUCTION

In the case of severe accidents in a nuclear power plant, the containment is the last barrier to prevent the environment from hazardous substances. Therefore the leak-tightness of the containment is of crucial importance to avoid a pass out of radioactive substances to the environment (Niklasch & Herrmann 2009). A core meltdown followed by a malfunction of the reactor pressure vessel might lead to huge amounts of steam combined with high pressures and temperatures. In the case of a double-layer concrete containment, the inner potentially pre-stressed layer should only allow the leaking of a defined amount of steam which might be collected and treated between the two containments (Niklasch 2007). Against this background, estimations of leak rates and their reduction by self healing processes are of enormeous importance.

While the overall mechanisms of self-healing in concrete are known and well described (van Tittelboom & Belie 2013; Edvardsen 1996; Hager et al. 2016), the main healing process for cracks flown with water was already determined with the formation of calcite (Edvardsen 1999). Several large scale experiments already investigated the self-healing behavior of pre-stressed containments pieces under steam conditions with a focus on the prediction of leaking rates (Granger et al. 2001; Riva et al. 1999; Niklasch & Herrmann 2009). Tests on a pre-stressed concrete containment specimens were conducted at the MPA Karlsruhe under most realistic accident conditions (concerning temperature and pressure) to assess the amount of steam and condensed water. Most interestingly, the leakage rates decreased over the test run, whereas the doubling of the cracks did not result in the expected increase of the effluent amount. Therefore it was assumed, that the cracks get internally blocked over time (Stegemann 2012).

While such experiment gather valuable information for accident scenarios, information on self-healing mechanisms inside cracks flown with steam is still lacking, especially under high pressure and temperature conditions. Hence, the aim of this work is to identify the main driving processes in crack healing under steam flow and evaluate their potential contribution related to the above mentioned accident scenario.

METHODOLOGY AND EXPERIMENTAL SETUP

Preparations

The investigation of self-healing processes in cementitious materials flown with steam was investigated on small scale specimens (4 cm * 4 cm * 11 cm) made from CEM I and CEM III and typical concrete ingredients (sandstone, limestone, granite). Both cements where mixed with w/c ratios of 0.3 and 0.4 to investigate the availability of calcium for water saturated and under saturated conditions. The specimens were fixed in a stainless steel reactor (see Figure 1) which followed the function of a glad nut. The reactor was filled with forming silicon to avoid flow around of the specimens. Thereby the in- and outflow to the specimens was kept clear by two tubes connected with oil seals. After the curing of the silicon a crack was introduces into the specimens via the in- and outflow tubes by a MTS 100 pressure testing facility. Finally the crack was opened to a defined width via the threaded rods. The width was measured by a standard of comparison that was mounted at the end of an endoscope. The crack widths of the experiments varied between 0 to 0.6 mm. Finally, the glad nut was uptight to gain additional tightness of the silicon.

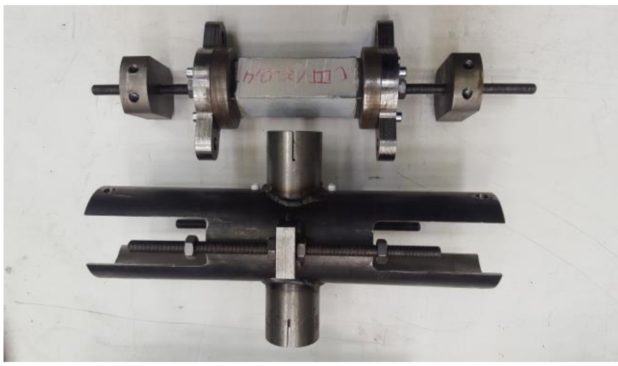

Figure 1: Reactor construction with specimen.

Figure 2: Experimental test facility.

Experimental Facility

The experimental conditions for the tests were chosen according to an accidental scenario for a double layered pre-stressed concrete containment of French nuclear power plants which are designed for a maximum pressure of 5.2 bar and a maximum temperature of 140 °C (Granger et al. 2001). The experimental setup at the MPA Karlsruhe has originally been developed for air steam leakage test at concrete wall segments with 1.2 m in thickness (Niklasch & Herrmann 2009) and is described in detail in Stegemann (2012). This overall setup enables to produce a continuous flow of steam and air with defined pressures (up to 5.2 bar abs.), defined temperatures (up to 140 °C) and defined shares of steam and air. These maximum values where as well the experimental conditions for all steam flow experiments. The steam for all experiments was produced from demineralized water. The setup was modified with six diversions from a reservoir at which the reactors could be connected (Figure 2). Each diversion was equipped with a valve and a manometer before and after the reactor. Therefore a pressure gradient could be adjusted following the assumption that the pressure decrease through a 1.2 m thick containment will only result in a small increment of the total pressure along a 4 cm long crack. For the experiments, this pressure increment was chosen mostly with 1 bar whereas some tests were conducted with 4.2 bar gradient. For volume flow measurements, the outflowing steam was lead over a cooling system where the condensed water was weighted and the remaining air flow was measured. As only one measurement device existed, the flow through each of the six specimens was monitored separately one after each other. During each test run, all reactors where monitored multiple times.

For all experiments, a flow time of approx. 5 h was aimed. However, the real exposure times varied between 4 h up to 6 h. A long term test run spreading measurements over 30 h was performed as well. After each run, the crack width was measured again, as changes occurred due to thermal expansion during the test runs. After steaming, all specimens were stored under nitrogen atmosphere to avoid further carbonation of the crack surfaces.

Optical Examinations Of The Crack Surfaces

All specimens were analyzed by µ-RFA (X-ray fluorescence spectroscopy) for the chemical composition of the crack surfaces, especially their share of calcium compared to blank samples. During the data evaluation, all results were automatically corrected for oxygen according to their mineral phase, which means calcium is further on mentioned as lime (calcium oxide). Due to the problem of analyzing the specimens before and after they were flown by steam, the results were compared to reference samples prepared under the same conditions.

Concerning the µ-RFA results it must me mentioned, that only a certain depth is penetrated by this method. Within this study, the coatings at the surfaces where never thick enough to measure them solely. Therefore all results feature a combination of calcium oxide and the underlying hydrated cement. Any coating of the surface will therefore only displace the share between cement and lime towards the lime.

As the µ-RFA method is only capable to detect elements heavier than neon (sodium and above), selected samples were additionally analyzed by ESEM (Environmental scanning electron microscope) to quantify the elements lighter than sodium. Thin sections through the crack surface were prepared as well from several specimens and investigated by transmitted-light microscope.

RESULTS AND DISKUSION

Flow Results

Comparing the condensed water amount with the remaining air mass flow after the cracks, the expected steam share of 60 % was not reflected in the measurements. As the air measurements showed some inconsistencies for themselves, the following numbers are based on the scaled water volume corrected for the 40 % of the remaining air.

The dependency of mass flow and crack width is pictured in Figure 3 and follows an exponential function. Maximum flows were recorded with up to 18.7 kg/h at a mean crack width of 0.58 mm. The widths were calculated as dominating average, considering the smaller mean crack width of the upper and lower crack profile.

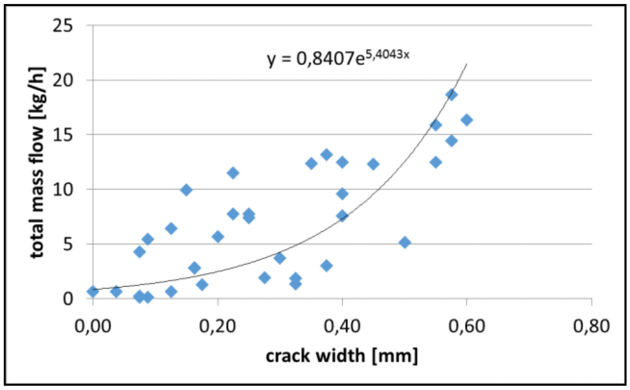

Figure 3: Measured flow amount and their associated crack width. Crack length = 4 cm, wideness = 3 cm.

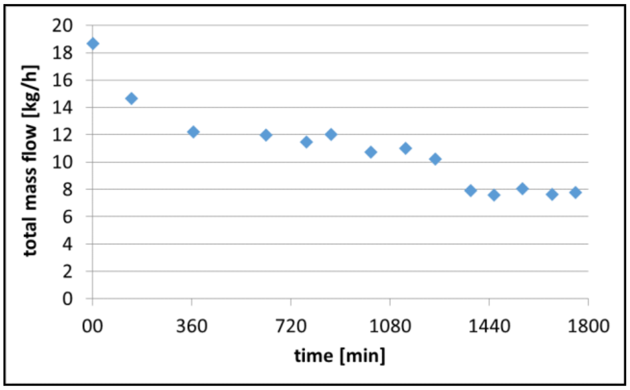

Figure 4: Decrease in the mass flow over the experiment time of a long term test run

These numbers were much higher than the ones for similar experiments with large scale pre-stressed specimens performed at the MPA Karlsruhe before (Herrmann et al. 2015). The main reason here is the separating crack of the specimens which did not occur in the large scale experiments.

A decrease of the flow over the experiment could be recognized as well. Within the long term test runs the flow decreased from 18 kg/h down to 8 kg/h (Figure 4) which might be partly attributed to the formation of calcite. Here, it must be mentioned that the initial drop in the first hour was usually due to thermal extension of the reactor.

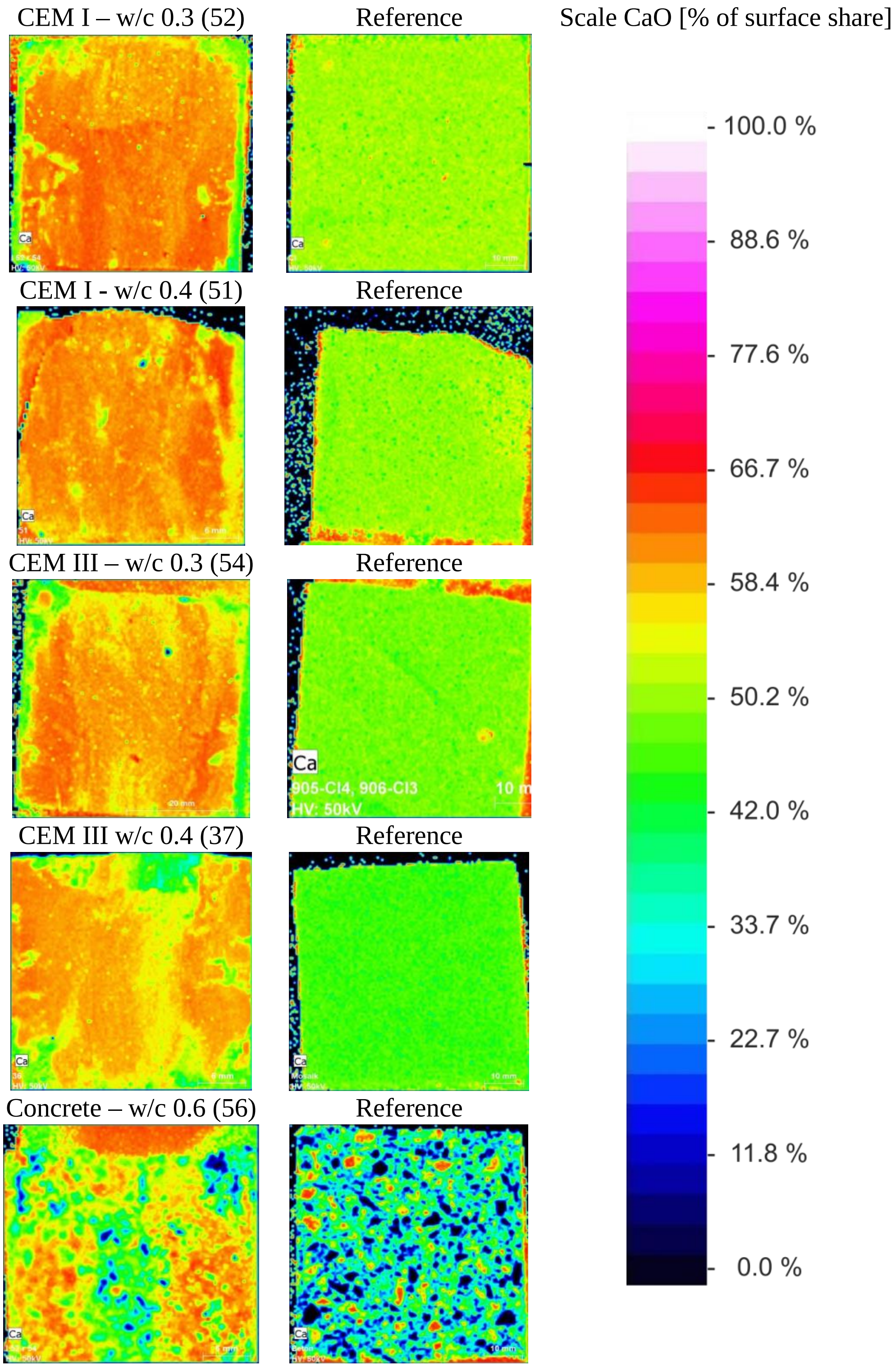

Figure 5: µ-RFA surface scans showing one example from each steam flown material and the related reference. Flow direction from top to bottom.

Mineralogical Investigations

While no changes occurred on the crack surfaces of the granite, calcite and sandstone specimens during the steam flow test, all cement and concrete specimens showed increasing CaO surface concentrations compared to the reference specimens. This approves for the experimental conditions of 140 °C and 5.2 bar but as well already for test performed with 120 °C and 3 bar. Each row in Figure 5 shows one example from the surfaces of all cement and concrete mixtures. The mean increase was 16.9 % for CEM I and 18.9 % for CEM III. A maximum increase thereby was found with 30.5 % for CEM III and with 25.1 % for CEM I for 6 h test runs.

Due to the vast amount of variable parameters like crack width and the pressure gradient, both resulting in the amount of flow, or the influence of different materials, statistical analyses of the data are of rather limited significance. Distinct trends deriving from the choice of material or the influence of the crack width were seriously considered but did not lead to reliable results.

The ESEM analyses prove the CaO on the crack surface to be $CaCO_3$ with a good fit (compare Table 1) to its characteristic mass share of calcium (20 % wt.), carbon (20 % wt.) and oxygen (60 % wt.). The surface pictures underscored this as well, showing the typical structure for CaCO3 minerals (Figure 6) compared to the cement surface of the reference sample (Figure 7).

Table 1: Atomic mass shares for the crack surface of two steam flown specimens and their related reference.

	Ca [% wt.]	C [% wt.]	O [% wt.]	Si [% wt.]
CEM I - w/c 0.4 (51)	20.8	16.6	57.8	0,9
Reference CEM I - w/c 0.4 (51)	13.5	9.8	67.4	5,3
CEM III - w/c 0.4 (55)	19.5	20.1	58.1	0.7
Reference CEM III -w/c 0.4 (55)	15.0	6.7	64.4	7.6

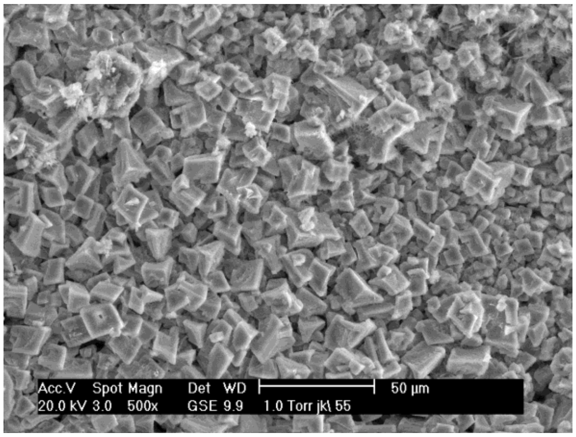

Figure 6: ESEM picture of a CEM III (w/c 0.3) specimen (55) flown by steam.

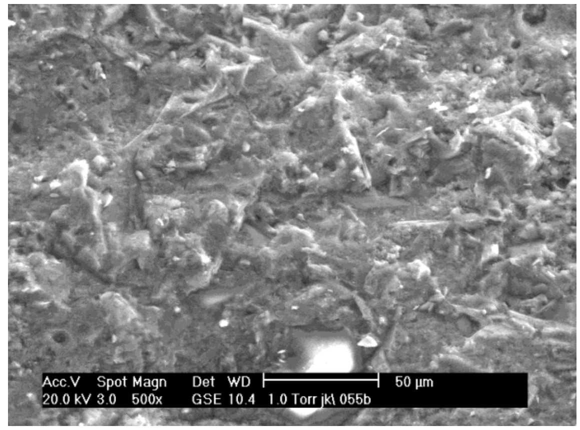

Figure 7: Reference sample of CEM III (w/c 0.3)

The thickness of the calcite layers was measured in thin sections of selected samples and for the long term test run. They varied from 10 - 40 nm after approx. 5 h of steaming up to a maximum of 300 nm after 20h. Within a long term test run, 6 specimens were investigated under similar conditions (initial crack width of 0,6 mm, pressure gradient of 1 bar, CEM III) to assess the temporal resolution of the calcite mineralization. After each 6, 12, 18, 24 and 30 hours, one reactor was disconnected and the specimen analyzed. The CaO concentrations on the crack surfaces thereby show in general an increasing trend over the exposure time (Figure 8). This goes as well along with the thickness of the calcite layer in the thin sections.

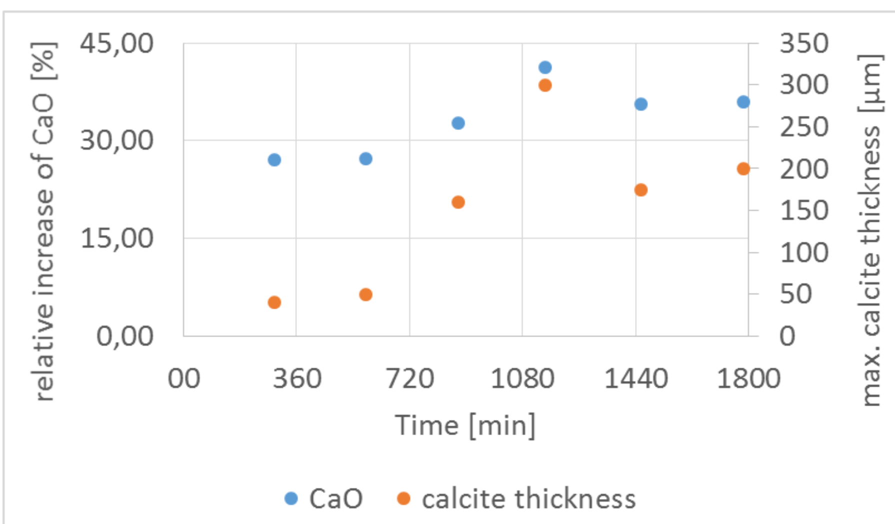

Figure 8: Relative increase of the CaO concentration of the crack surfaces (left) and maximum thickness of the calcite layer (right) related to the time the specimens were flown by steam.

CONCLUSIONS

Within the research framework of self healing mechanisms in the containment of nuclear power plants, small scale flow experiments were performed with specimens of different cement and concrete mixtures under accidental scenario conditions of a steam flow with 140 °C and 5.2 bar. Crack healing processes could thereby be investigated via different mechanisms.

The flow measurements showed a decrease of the flow amount over the operation time of the experiments therefore indicating self healing of the cracks. The formation of calcite contributes here along with other mechanisms like thermal expansion which makes it difficult here to differentiate between them.

The combination of µ-RFA and ESEM analysis feature the formation of a calcite layer during the experiment on the crack surfaces of all specimens. This proves that the in situ mineralization of calcite under flow conditions exceeds counteractive processes e.g. abrasion o the surface or restraint to reactive kinetics due to the fast transport of participating ions in the crack. Beside, the thickness of this calcite layers derived from selected thin sections with up to 300 nm allow first estimation to what share these layers may contribute to the total crack healing process.

Long term experiments up to 30 h showed the increase of the calcite concentrations over time. Though the interaction between the different processes like abrasion, the reduction of mineralization due to the flow amount, the reduction of the flow due to the formation of calcite could not be described satisfying based on the experimental data. Here, further research might help to differentiate such opposing effects.

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