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The COLIMA experiment on aerosol retention in containment leak paths under severe nuclear accidents

Flavio Parozzi[a,*], Eduardo D.J. Caracciolo[a], Christophe Journeau[b], Pascal Piluso[b]

[a] RSE, Power Generation Department, via Rubattino 54, I-20134 Milano, Italy
[b] CEA Cadarache, France

HIGHLIGHTS

- Experiment investigating aerosol retention within concrete containment cracks under nuclear severe accident conditions.
- Provided representative conditions of the aerosols suspended inside the containment of PWRs under a severe accident.
- Prototypical aerosol particles generated with a thermite reaction and transported through the crack sample reproducing surface characteristics, temperature, pressure drop and gas leakage.
- The results indicate the significant retention due to zig-zag path.

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ABSTRACT

CEA and RSE managed an experimental research concerning the investigation of aerosol retention within concrete containment cracks under severe accident conditions. The main experiment was carried out in November 2008 with aerosol generated from the COLIMA facility and a sample of cracked concrete with defined geometric characteristics manufactured by RSE.

The facility provided representative conditions of the aerosols suspended inside the containment of PWRs under a severe accident. Prototypical aerosol particles were generated with a thermite reaction and transported through the crack sample, where surface characteristics, temperature, pressure drop and gas leakage were properly reproduced.

The paper describes the approach adopted for the preparation of the cracked concrete sample and the dimensioning of the experimental apparatus, the test procedure and the measured parameters. The preliminary results, obtained from this single test, are also discussed in the light of the present knowledge about aerosol phenomena and the theoretical analyses of particle behaviour with the crack path.

1. Introduction

Under severe accident conditions, there is the possibility for gases and aerosol particles to pass the reactor containment through cracks and/or failed seals even if a catastrophic containment failure does not occur. For these cases, past safety analyses usually assumed that the aerosol release rates are identical to the gas leak rates.

However, narrow leak paths can trap airborne particles significantly, so that releases of radioactive materials are quite lower than the correspondent gaseous leak rates.

To investigate about this phenomenon, past experimental studies were performed within small leaks and capillaries having diameter ranging from few microns to few millimetres, with pressure differences up to several bars. Large scale experiments were managed during the 60s in the USA, when the Atomic Energy Commission sponsored the Containment System Experiment program (CSE) at Battelle Memorial Institute. With a one-fifth linear scale model of a typical 1000 MWe PWR plant, a series of aerosol leakage tests with artificial pathways gave a decontamination factor 15 for iodine and 100 for caesium in dry conditions, and almost complete retention in wet conditions. Other experiments, on large scale, were performed in Japan by NUPEC on actual containment penetrations of a BWR plant using dry CsI aerosol particles: those experimental tests indicated decontamination factors ranging between 10 and 1000 (Parozzi et al., 2005).

With the target of developing a reliable computational model for predicting the aerosol retention in cracks, easy to be

* Corresponding author.
E-mail addresses: flavio.parozzi@rse-web.it, parozzi@rse-web.it (F. Parozzi), eduardo.caracciolo@rse-web.it (E.D.J. Caracciolo), christophe.journeau@cea.fr (C. Journeau), pascal.piluso@cea.fr (P. Piluso).

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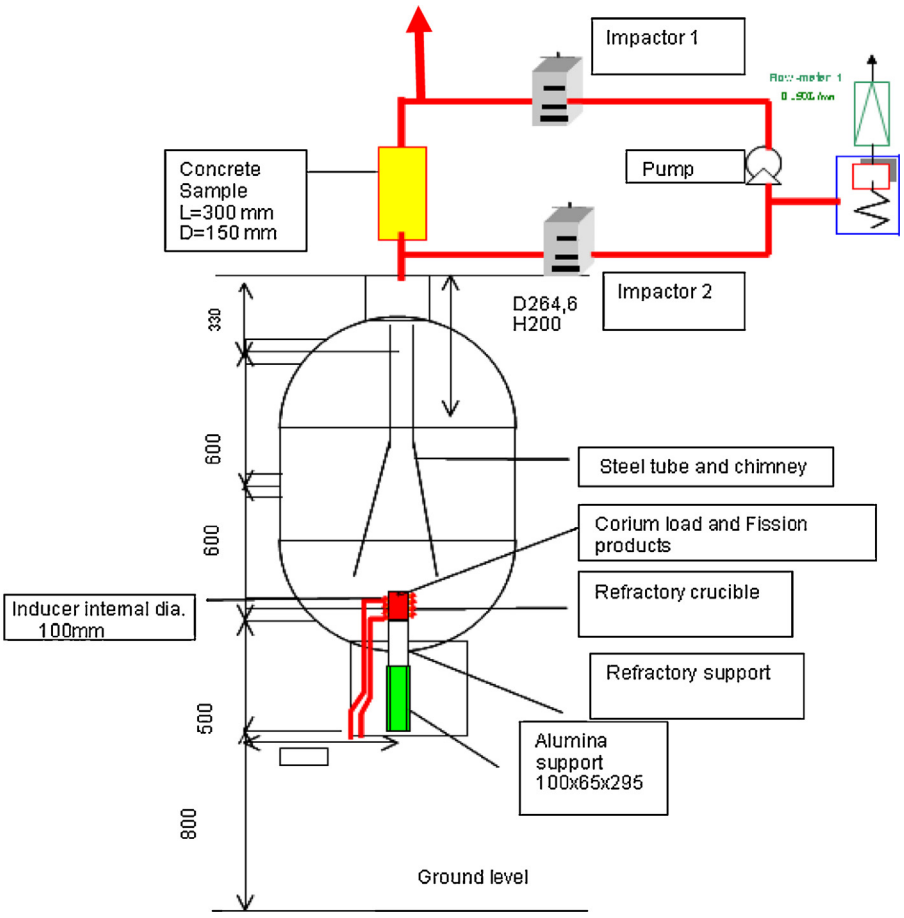

Fig. 1. Schematization of the test COLIMA on the concrete crack sample.

implemented into Source Term Codes like ASTEC, an ad-hoc "circle" was managed in the framework of the Network of Excellence of FP6 SARNET (Severe Accident Research and management NETwork) (Albiol et al., 2010). That circle also promoted the design and execution of experimental activities for the validation of the models themselves (Herranz et al., 2010; Mitrakos et al., 2008).

To this aim, CEA and RSE specialists set-up an experimental equipment for reproducing the aerosol retention within containment cracks under severe accident conditions. The experiment was carried out in November 2008 with aerosol generated from the facility COLIMA at CEA Cadarache, using a sample of cracked concrete with defined geometric characteristics manufactured by RSE (Fig. 1).

2. The COLIMA facility

COLIMA is a facility belonging to the PLINIUS platform, a research installation dedicated to the study of core-melting accidents in nuclear reactors. It includes 4 corium facilities: VULCANO, where about 50 kg of corium can be melted and poured; COLIMA, dedicated to materials and aerosols studies with a few kilograms of molten corium in a controlled atmosphere enclosure; KROTOS, for corium-water interactions and VITI, dedicated to physical properties measurements of corium (Piluso et al., 2002).

COLIMA (COrium LIquid and MAterials) consists in a 1.5 m^3 tank, where the maximum internal pressure can reach 0.3 MPa. The corium can be melted in a crucible by a thermite reaction or an induction coil that can maintain it hot in order to provide a steady state situation up to 3000 °C. The crucible, designed to contain few kilograms of corium, is surrounded by a thermal shield ring and can be placed at the bottom or at the middle of the tank.

The walls of the vessel tank are thermally controlled at 150 °C. Portholes, dedicated to the instrumentation, are located at its top, half height and bottom.

According to the scientific objectives of each experiment, different configurations of the facility can be used: corium/materials interaction (concrete, ceramics), release of aerosols from the corium (simulating physical-chemistry of oxidic and metallic fission products, without radioactive isotopes except uranium).

As this facility is capable to simulate accidental conditions expected for a PWR containment, it was utilized in this experiment, where prototypical aerosol particles were generated and transported through the crack sample, properly reproducing temperature, pressure drop and gas leakage (Table 1).

3. Crack sample preparation

Concrete blocks were manufactured by RSE according to the composition of a typical Spanish plant (limestone aggregates) (Morandi and Parozzi, 2010).

After 28 days of maturation at constant temperature and hygrometry, cylindrical samples (diameter of 0.13 m and length of 0.3 m) were drilled in each block. Then, a number of samples were prepared by splitting those cylinders by a standard procedure (Fig. 2) to obtain well-characterized cracks, with known geometry and internal surface representing a part of a typical internal crack pathway open to the aerosol leakage. On the basis of the characteristics of the concrete crack obtained (crack surface quite flat and

Table 1
Summary of the parameters assumed to reproduce the aerosol retention in 1000 MWe PWR containment cracks with the COLIMA experiment.

Reference parameter	Reference accident conditions	COLIMA conditions	Adaptation or scaling
Sample material	LWR containment concrete	Limestone aggregates reproducing LWR basemat	Representative
Crack length	>140 cm	30 cm	Representative of a part of the aerosol pathway
Crack shape/section	Flat, few mm^2	130 mm × 0.5 mm	Representative
Crack curvature	Zigzag crack curves	10–30 mm radius curves	Representative
Pressure drop through containment wall	0.3–0.7 MPa/m	0.33 MPa/m	Representative
Gas temperature	150–200 °C	110 °C	Low, but representative
Crack wall temperature	Close to room temperature as the crack opens, then approaching gas temperature	Initially close to 60 °C, then approaching gas temperature	Influencing re-evaporation of volatiles and particle thermophoresis
Gas composition	Air, hydrogen and steam close to saturation	Pure nitrogen	Weakly influent on dry particles deposition
Aerosol concentration	About 1–5 g/m^3	~0.05–0.15 g/m^3	Low, but representative
Aerosol composition	~0.1% I ~1% Cs ~1% Te 100%-complement: fp oxides and inert aerosols	~3% I ~18% Cs ~5% Te 100%-complement: fp oxides and inert aerosols	Enough representative; generated by a representative ex-vessel corium pool
Aerosol size	AMMD ~ 1–5 μm	AMMD = 1.1 μm $\sigma_g = 2$	Representative

with a limited number of porosities) the most suitable concrete sample for the test was chosen.

The chosen cracked concrete sample was opened and cleaned with an air blow gun. To guarantee a sufficient width of the crack, small 0.5-mm-thick Teflon pieces were placed at the 4 corners of one half sample and then the second half of the sample was installed on the top. The two parts were sealed using a silicone paste.

The sample was inserted in a flanged steel holder. The boundaries of the cracks, as well as the connection with the in/out flanges were also sealed, in order to have the aerosol exiting the COLIMA pressure tank transported only through the crack sample (Fig. 2). The sample holder was finally coupled to COLIMA and thermally insulated (Fig. 3).

4. The experimental test

The experiment consisted of the generation of fission product aerosols by a prototypical corium pool within the vessel tank of COLIMA. The aerosol flowed through the concrete crack and partially escaped to the chimney through very high efficiency filters, as already shown in Fig. 1.

In particular, the experiment was carried out with a thermite mixture (1800 g) of U_3O_8, CrO_3, Fe_2O_3 and Zr, that gives the prototypical reaction products: UO_2, ZrO_2, Cr, Fe.

Concrete degradation products and elements simulating the fission products were also added. In particular, volatile species, such as caesium and iodine compounds, were included to take into account the aerosols generated by the in-vessel phase of the severe accident that are not trapped in the reactor coolant circuit. The resulting mixture of 2 kg was heated above 2000 °C and melted.

The tank of COLIMA was pressurized with nitrogen. The pressure drop along the cracked concrete sample, maintained by the nitrogen flow, was of the order of 1 bar (it means a pressurization of the COLIMA tank up to 2 bars absolute), and reproduced a 0.3 m-piece of a crack 1.5-m-length (assuming then a total pressure drop of 5 bars along a whole real crack). The gas and aerosol leakage through the tube containing the sample was controlled by a ball valve. The section containing the concrete crack, as well as the downstream tank for the aerosol discharge, was let at room conditions in order to have a transient effect when the ball valve opened. The impactor collectors were heated above 100 °C to prevent condensation of humidity.

The test was performed on Friday November 28, 2008, following COLIMA procedures, after a pre-conditioning of the components (Journeau, 2011):

load preparation
pre-heating of aerosols transports tubes at 110 °C

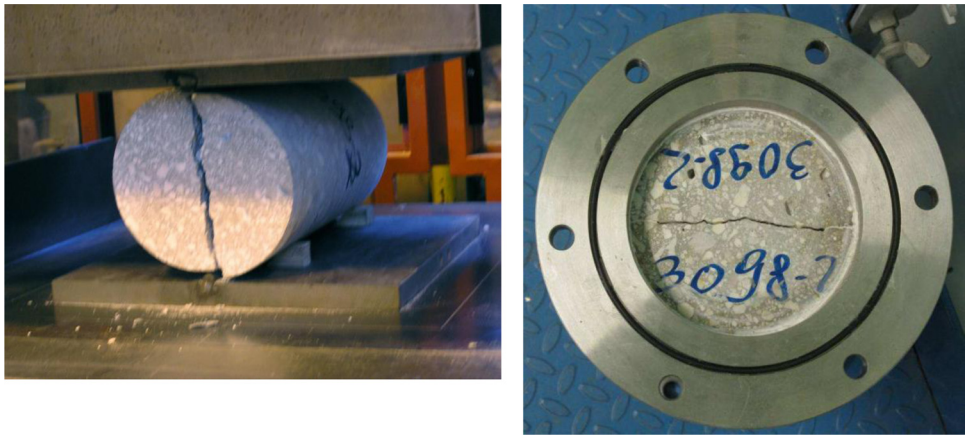

Fig. 2. Instant of the splitting of one of the cylindrical samples (left), and view of the inlet of a crack sample positioned and sealed in the flanged steel holder (right).

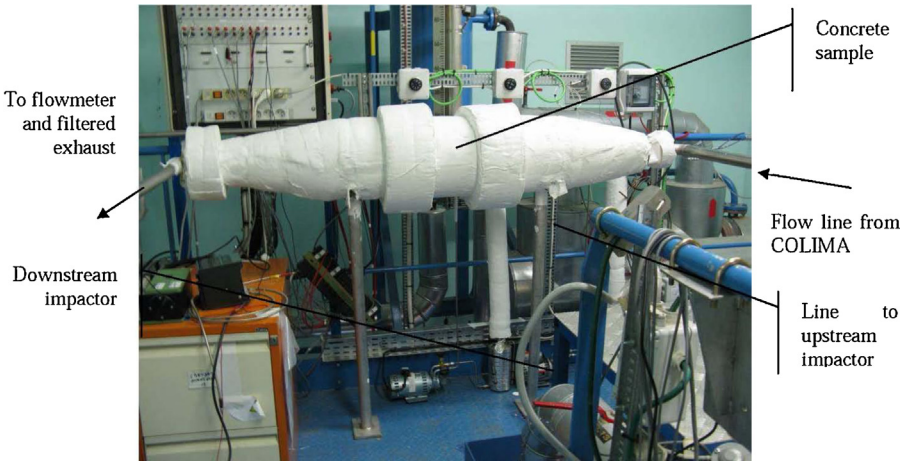

Fig. 3. The concrete sample, placed in the insulated holder, coupled with COLIMA.

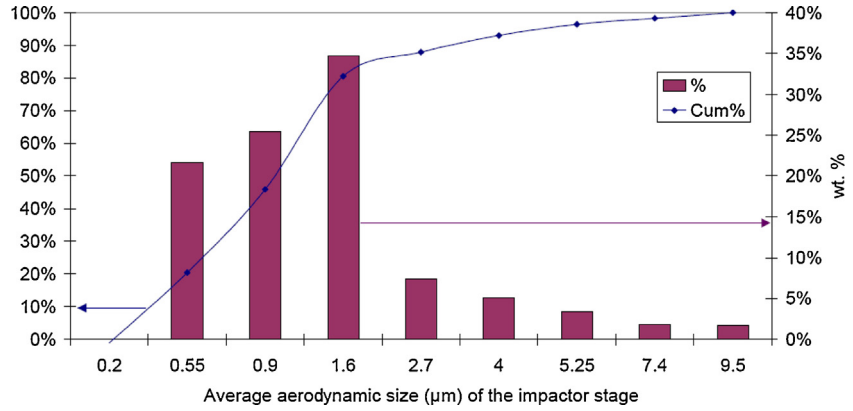

Fig. 4. Aerodynamic size distribution of the aerosol entering the crack sample, as determined from the upstream impactor.

COLIMA set to 1 bar rel. of nitrogen
heating of COLIMA walls at 60 °C
stabilization of flow rate at 400 Nl/min (8.3×10^{-3} kg/s).
ignition of the urano-thermite load
aerosols formation, release and transport to impactors and concrete test section.

5. Results of the analyses

There was no significant change in gas temperature and pressure around the ignition time of thermite. Both impactor sampling lines use an isokinetic pump (28 L/min). They provided the size distribution reported in Fig. 4. A best fit with a log-normal law gives an AMMD of 0.97 μm with a logarithmic standard deviation of 0.72 (i.e. $\sigma_g = 2.05$) for the upstream aerosols.

Very few aerosols were collected at the downstream impactors, indicating an important retention. The error bars plotted in the figure show that the measured inlet and outlet masses are lower than the uncertainties.

Downstream, the only visible aerosols were found in the 0.4–0.7 μm bin of the impactor and were much less than upstream, with errors bars corresponding to the 0.4 μg weighing uncertainties (Fig. 5). It appeared that the other downstream deposits are below the uncertainty threshold, and that retention was larger than a factor of 10 at stage 7 (0.4–0.7 μm) for which some particles escaped the crack, and much larger for the larger sizes. In impactor stage 1 (5.8–9 μm) the outcoming mass, more than incoming ones, can be due to some concrete particles. Silica and calcium oxides were observed on the impactor and on the crack, but, since the impactor collectors are made of silica, no quantitative data are available for SiO_2.

The composition of smallest particles (0.4–0.7 μm stage 7), was typically 18 wt% Cs, 5% Te, 3% I, 3% Cr, 2% Fe. Iodine was not observed in the other stages corresponding to largest particles. In stages 6 (0.7–1.1 μm) to 4 (2.1–3.3 μm), Cs, Te, Cr and Fe were observed with typical ratios of 3–4:1.2:1:1:1. Some uranium (2–3 wt%) was observed in some of the deposits at stage 7. Caesium was the major component at this stage.

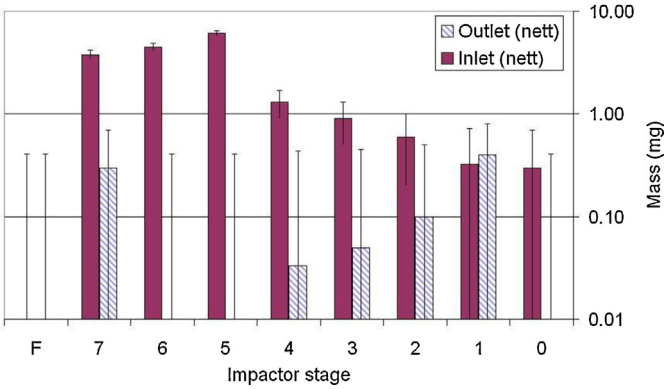

Fig. 5. Distributions of the aerosol mass collected in the impactors at inlet and outlet of the crack sample.

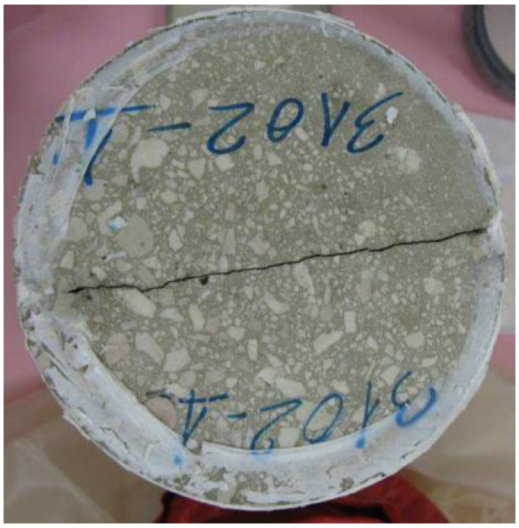

Fig. 6. Upstream face of the concrete sample after the test. The sample sealing is visible on its periphery.

Fig. 7. Post-test view of the two sides of the concrete crack sample. The arrow indicates the direction of flow.

After the test, the concrete sample was dismounted from the sample holder. No aerosol deposit was observed on the concrete faces, even near the crack upstream inlet (Fig. 6).

The sealing was cut and the two halves were separated. Fig. 7 shows that there was an intense aerosol deposition (in black) in the first 5 cm of the crack sides, and that there was almost no deposit after 20 cm. In the 5–20 cm range, some preferred flow path traces are visible through of the cracked surface (Fig. 8).

The sample was divided into 5-cm zones and the aerosols were wiped from one of the halves and collected. Table 2 presents the

Table 2
Collected aerosol masses in the concrete sample.

Zone	Collected mass	Mass fraction
0–5 cm	210 mg	70%
5–10 cm	68 mg	23%
10–20 cm	21 mg	7%
0–20 cm (total collected mass)	299 mg[a] (retention 95%[b])	

[a] Calculated on the basis of collected mass from the sample after the experiments.
[b] Evaluated on the basis of the comparison between the inlet and outlet impactors results.

Table 3
Average mass composition of the samples collected along the concrete crack.

	Mg	Si	Ca	Cr	Fe	Te	Cs	Ti	U
B1 (0–5 cm)	10%	17%	9%	19%	15%	12%	18%	0%	0%
B2 (5–10 cm)	5%	11%	2%	12%	9%	6%	56%	0%	0%
B3 (10–15 cm)	3%	27%	5%	0%	45%	0%	0%	2%	19%

collected masses for the three sampling zones (zones 10–15 and 15–20 cm have been grouped due to the small amount of aerosol). The total collected mass is about 300 mg. The aerosol injected to the crack was evaluated after the experiments on the basis of the results of the impactor sampling and of the total collected mass from the concrete sample: a total of about 200–300 mg of aerosols was estimated to be directed to the sample. Taking into account the gas flow (400 Nl/min), and the aerosol cloud duration (about 5–10 minutes), the aerosol concentration entering the crack sample turned out to be in the range 0.05–0.15 g/m^3. These numbers are not fully consistent with the masses estimated from impactors, maybe because of the mixing of aerosol with some concrete particles during collection, and due to difficulties to get an exact mass balance, it is recommended to consider mainly relative fractions.

Representative samples of aerosols were taken to perform post-test analyses. The samples were first observed under optical microscopy, following this step, analysis by means of Scanning Electron Microscopy (SEM) coupled with an Energy Dispersion Spectrometry analysis system (EDS), OXFORD ISIS 300 to obtain local composition of the solid phases obtained and deposited.

The laboratory analysis of those deposits reported that numerous elements were deposited in the first 5 cm of the crack, including caesium, tellurium and uranium. A large particle of (U, Zr)O_2 was also found, together with particles of concrete decomposition products (Mg, Ca, Si)O_x with some chromium (4 wt%) and caesium (2 wt%), as well as particles of Te, and iron and chromium oxides.

Between 5 and 10 cm from the crack inlet, the analyses indicated a significant presence of Cs, while in the last part of the crack (more than 10 cm away from the inlet), the deposits were mainly made of iron and uranium (sometimes with some zirconium).

Table 3 compares the average compositions from the collected samples. It appears that Cs and Te are mainly deposited in the first 10 cm (caesium representing more than half of the deposits in the 5–10 cm zone). In proportion, the fraction of uranium oxide aerosol is negligible in the first 10 cm deposits but becomes significant (19%) in the remainder (since most off the other aerosols have been deposited, except iron). It is difficult to assess in which proportion the measured Si, Mg and Ca are due to concrete decomposition product aerosols and to debris from the concrete surface.

In conclusion, it must be noted that some radioelements such as uranium (found in the B3 sample) and caesium (found in the downstream stage 7 collector) are within the least deposited particles.

6. Interpretation of the results

Even if the duration of aerosol generation was shorter than planned, the test was successful and confirmed the intense aerosol deposition in a representative crack path. Retention of about 95% was achieved, mainly in the first 10 cm of the crack.

The transported chemical species and particle diameters were not retained in the same way: only the smallest particles (below 0.7 μm of aerodynamic diameter) were not totally deposited, even if within experimental uncertainties. The main elements of the undeposited aerosols were steel elements (Fe, Cr). Small quantities of caesium, and to a slightly lesser extend molybdenum and cadmium, were also transported through the crack sample.

It must be stressed that the transport time in the crack sample was of the order of 4 ms, and the observed retention can be

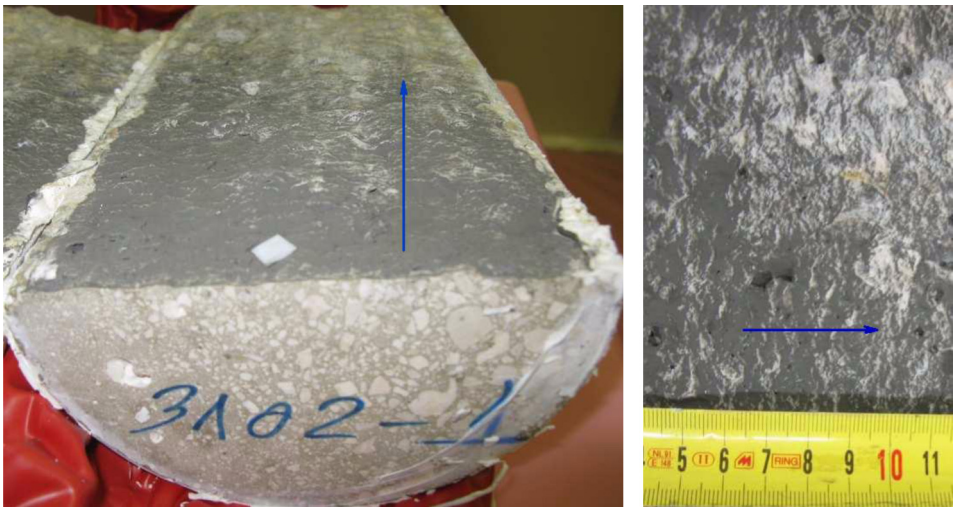

Fig. 8. View from upstream of one of the concrete halves (left) and zoomed photograph showing the effect of crack tortuosity on deposits (right). The Teflon 0.5 mm spacer is visible (small white square); the arrows indicate the direction of flow.

explained only assuming that adhesive forces strongly prevail the lift forces. As a matter of fact, a preliminary analysis with the support of aerosol models of ECART code (Parozzi and Paci, 2006; Allelein et al., 2009) indicates that it is likely that most of the retained particles were removed by inertial impaction due to the crack tortuosity (average path curvature estimated of the order of 1 cm). The retention cut-off is estimated, in that preliminary run, around 0.1 μm, giving the right order of magnitude.

7. Open issues and conclusions

An experiment with the facility COLIMA was addressed to investigate the aerosol retention within concrete containment cracks under severe accident conditions in close connection with the SARNET severe accident research network. The experiment was carried out with a crack sample reproducing a 0.3 m piece of a real containment crack and the aerosol was generated with a thermite reaction that gave representative reaction products.

The experiment results confirmed a significant retention of aerosol in the crack. Furthermore, the results, even if they are preliminary as they are from a single test, show that the most of aerosol deposition occurred in the first 0.05 m of the crack sides, while negligible deposits were found after 0.2 m. In the 0.05–0.2 m range, some preferred flow path traces are visible.

As regards the aerosol behaviour, the analysis of the test results with the support of aerosol models highlighted the role of inertial impaction accounting for the path tortuosity, as the main responsible of the adhesive forces inside the crack pathway (the simulations with ECART are in good agreement with the experimental aerosol mass deposition and distribution along the crack). In fact, in an equivalent straight path, the aerodynamic lifting forces would continuously remove any particle from the wall surfaces.

The different retention of elements could be caused by the size-dependent composition of the particles due to the aerosol nucleation mechanisms (depending on the vapour pressure of the involved chemical species).

As only one test is not sufficient to establish the range of experimental uncertainties and validate or adjust the code models, future studies should be addressed to understand how the particle inertia really acts in zig-zag pathways and what happen if the particles are already deposited and saltation can occur. It would therefore be useful to conduct experiments with different conditions in terms of crack thickness and flowrate. Iodine behaviour and, more in general, the influence of chemistry on the aerosol particles characteristic and transport should require specific investigations.

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