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Investigation of aerosols released at high temperature from nuclear reactor core models

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Abstract

Two experiments were performed to simulate severe reactor accident with air ingress into the hot reactor core. The model bundles contained nine PWR type fuel rods. Their cladding was pre-oxidised by argon–oxygen (test 1) and steam (test 2). The released aerosol was measured continuously by laser particle counters. Morphology and elemental composition of the aerosol particles were studied on samples collected by impactors and quartz filters. The highest aerosol release was detected at the steepest rise of the bundle temperature. A second increase of the aerosol release appeared at the cooling down period. Because of the higher maximum temperature at test 2 about two orders of magnitude more uranium was released than in test 1. The highest emission was found for tin at test 1 and for zirconium and iron at test 2.

1. Introduction

Nuclear power plants contain safety systems assuring that after a postulated accident, the plant can be kept in a controlled state. The prevention of radioactive release – especially of highly volatile species – (see [1]) during a severe accident is an important safety goal. Such accidental situations arise when after vessel failure, air enters into the reactor core. The exothermic oxidation results in strong temperature escalation up to above 2270 K. As a result of high temperature interactions, different kinds of aerosols are formed and released. It is necessary to simulate such type of reactor accident to collect more information about the level of bundle degradation and of the aerosol emitted. In the framework of the oxidation phenomena in severe accidents (OPSA) project (presented in [2]), two integral air ingress tests (AIT) were performed with 9-rod PWR type bundles at the KFKI Atomic Energy Research Institute. Details of the experiments and the post-test examination of the bundle will be published elsewhere. As the fission products were not simulated in the experiments, the main objective was to get measurement data for the release of structural materials and UO_2. This paper summarises the results regarding the concentration, the size distribution and elemental composition of the aerosols released in the two tests.

2. Experimental

2.1. Experimental facility

The core degradation experiment (CODEX) facility was constructed for the investigation of the behaviour of small fuel bundles in severe accident conditions.

The main component of the facility is the test section which incorporated the fuel bundle to be investigated. In the AIT, 9-rod PWR type bundles were used. The rods were arranged on a square lattice, 8 peripheral rods were electrically heated with tungsten bars. The central rod was not heated, it was used for instrumentation. Two spacer grids were applied to fix the bundle. The heated

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length of the bundle was 600 mm. The cladding material was Zircaloy, the shroud part was made of a 2 mm thick Zr2%Nb alloy. Inside the fuel rods, annular UO_2 pellets were placed between the heater bars and the cladding. The depleted fuel had 0.2% U^{235} enrichment. Around the shroud, ZrO_2 thermal insulation was added. The test section was connected to the pre-heater and to the cooler sections as coolant inlet and outlet, respectively. Additional junction was connected to the bottom of the bundle to inject cold – room temperature – air.

The pre-heater unit was able to supply either hot gases or steam to the test section. The outlet temperature of the pre-heater could reach 1073 K. The off-gas streaming out of the test section was 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 filter system.

For the investigation of the aerosol release, a cascade impactor system was connected to the upper plenum of the cooler and two pipelines allowed the continuous measurement of aerosols by means of laser particle counters.

The upper part of the experimental facility is schematically shown in Fig. 1, where the place of aerosol sampling can also be seen at the upper part of the cooler. The most important parameters of the CODEX-AIT facility are summarised in Table 1.

2.2. CODEX-AIT-1 experiment

In the first AIT experiment, Ar + 25% O_2 mixture was used in the pre-oxidation phase to form about 50 μm protective oxide layer on the Zircaloy cladding. The experiment consisted of the following main steps:

- Slow heat up of the facility by 4 g/s hot Ar up to 1023 K in 12 000 s.
- Stepwise increase of power up to 1600 W while the temperature reached 1223 K.
- Pre-oxidation by Ar + 25% O_2. The oxidation heat resulted in a temperature excursion up to 2373 K, therefore the Ar + 25% O_2 was changed for cold Ar and the test section was cooled to 1173 K.
- After a temperature stabilisation at 1173 K, cold air was injected and a second temperature escalation up to 2273 K occurred. Then the test was terminated by cooling down by cold Ar.

The impactors were in use during the temperature escalation of the air ingress phase of the test. They were operated in series and each was used for 60 s, while 1 l/min outlet gas was sucked through the impactor unit. The $10 \times 60 = 600$ s time sampling period meant that the most important phase of the test was covered by impactor sampling. The first unit was switched on when the test section temperature reached 1473 K. In AIT-1, the rod-like structure of the bundle remained almost intact. Failure of some rods were observed close to the top of the bundle. The spacer grid was heavily oxidised and broken. The upper spacer oxidised and fragmented in a larger extent than the cladding.

2.3. CODEX-AIT-2 test

The test conditions achieved in CODEX-AIT-1 test were not favourable to U-bearing aerosol formation,

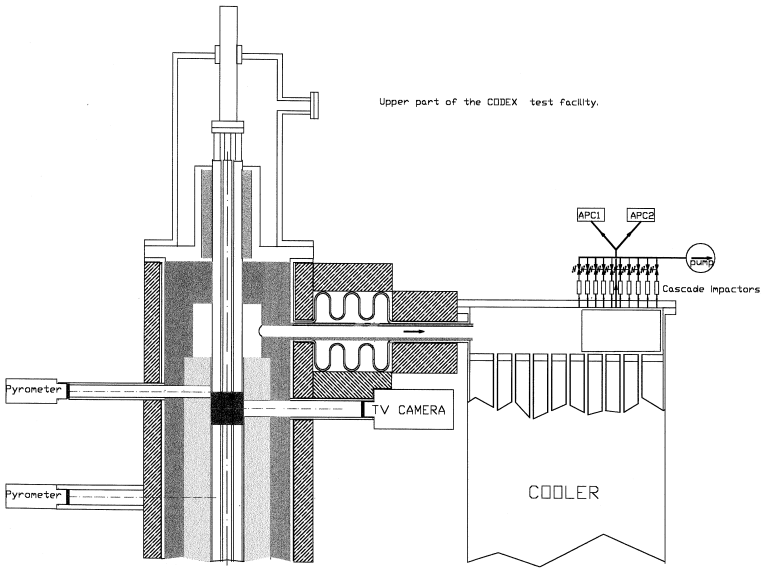

Fig. 1. Schematic view of the CODEX-AIT facility with aerosol samplers.

Table 1
The most important parameters of the bundle used in AIT experiments

Bundle characteristics	
Bundle type	PWR
Bundle size	9 rods
Number of heated rods	8
Number of unheated rods	1
Pitch	14.3 mm
Rod outside diameter	10.75 mm
Cladding thickness	0.725 mm
Cladding material	Zircaloy-4
Heater material	Tungsten (W)
Heater length	600 mm
Heater diameter	6 mm
Fuel pellets – heated rods	UO_2 annular pellets
Fuel pellets – unheated rod	UO_2 annular pellets
Pellet outer diameter	9.1 mm
U-235 enrichment	0.2%
Pellet stack	600 mm
Grid spacer material	Zircaloy-4
Grid spacer location	lower: −15 mm; upper: 535 mm
Shroud material	Zr2%Nb
Shroud wall thickness	2 mm
Shroud inside dimensions	48×48 mm^2
Shroud insulation material	ZrO_2 fibre
Shroud insulation thickness	19 mm

probably because of the lower temperature and the small amount of UO_2 exposed to oxygen. The AIT-2 test was to be allowed to proceed to more severe bundle degradation and higher temperature. Use of steam rather than $Ar + O_2$ mixture for pre-oxidation could help to avoid temperature excursion and it is more prototypic for a reactor accident.

The test sequence consisted of the following steps:

- Slow heat up of the facility with hot argon to 873 K and thermal stabilisation.
- Pre-oxidation in Ar/steam mixture for 60 min in two stages at 1093 K (30 min) and 1173 K (30 min).
- Stopping the steam injection, cooling to 1093 K and flushing with hot Ar.
- Starting the ingress of cold air (2.5 g/s) and increasing the electrical power to reach an initial heating rate of 0.5 K/s. Temperature excursion was observed to a maximum of 2173 K.
- Stopping of the air ingress when the temperature in the central elevation reached 1973 K indicating the melting process.
- Termination of the experiment when the melt reached the lower parts of the bundle, turning off the power and changing the air supply for cold Ar.

The impactors were activated after the pre-oxidation phase. The aerosol sampling strategy by impactors included background measurements in argon before and after pre-oxidation with 60 s sampling, during the excursion and relocation parts of the transient phase with 30 s sampling, and in the cooling down with 60 s sampling.

In the AIT-2 test, the bundle has suffered much more severe damage than in the first test, because the high temperature conditions were kept for longer time in AIT-2 than in AIT-1. The visual examination of the bundle showed strong oxidation and damage both of the shroud and the fuel rods.

2.4. Sample collection and preparation

Filters and impactors were used for sampling. For post-test analysis of aerosols, particles were collected by 10 impactors. At the AIT-1 test, they had two stage cascade impactors, while at AIT-2 they had only one stage. Particles were settled partly on smoothly polished Si and conductive carbon (fixed to the impactor plates), partly on quartz fibre filter placed into each impactor assembly. A nickel impaction plate was placed at the top of the cooler to collect aerosol particles, too (see Fig. 1). At AIT-2 test, this plate was fitted by two pockets and two collector samplers (all of them were made of nickel plate) to study the aerosol distribution versus place of the sampling. The quartz fibre filters were cut out from fibre delivered by Binzer and Munktell Filter Gmbh. Samples for spark source mass spectrometry were prepared by mixing the pulverised fibre filter with high purity graphite powder. The graphite contained 20 at ppm Pt as inner standard.

2.5. Applied techniques and instruments

The on-line aerosol measurements were made by two laser airborne particle counters (signed by APC-03-2 and PAPC-03-2, respectively). The measuring principle of these particle counters is based on laser light scattering with opto-electronic detecting. The data evaluation system comprises filtering, multichannel signal analysing interface and lap-top PC. The system was presented by Czitrovsky and Jani [3,4]. The *APC method* resulted in particle concentrations directly, classifying the scattered light signals into six ranges corresponding to the 0.3–0.5, 0.5–1, 1–3, 3–5, 5–10 and >10 μm intervals. Particle concentrations could be detected up to 5×10^6 particles/l values.

In the CODEX-AIT experiments, the sampling head of the PAPC-03-2 counter was mounted in the top of the cooler using a ~20 cm long copper tube. The APC-03-2 counter was placed out of the container and coupled with a 1.5 m long tube to the cooler head. The velocity of the gas stream was about 3 l/s. The counters were set to 5 s integration cycle time.

The size and shape of aerosol particles (collected by the impactors) were studied by a Philips SEM 505 electron microscope equipped with a LINK AN 10/55S type of energy dispersive microanalyser (EDS). For quantifying the size of the aerosol particles, the mean Feret diameter, while for the shape, the ratio of the minimum and maximum Feret diameters were chosen. These parameters were measured on SEM images of aerosol samples by a software of the EDS. Elemental analysis of single aerosol particles and their aggregates was made by EDS, while of particles collected by fibre filters was done by a MS 702 R spark source mass spectrometer (SSMS). The sensitivity of the SSMS was about 0.01 at ppm.

3. Results and discussion

3.1. Aerosol released in AIT-1 experiment

3.1.1. Size and shape of aerosol particles

Peak aerosol concentrations of $1–2 \times 10^6$ particle/l were detected simultaneously by APC-03-2 and PAPC-03-2 counters during the air ingress phase of the test. The dynamic range of the total concentration measured by APC-03-2 was $\sim 10^5$. The results of the measurements were obtained in particle concentration at various size ranges. These data were transformed in mass concentration using an average density value for the investigated aerosols. During the AIT-1 test, large scale aerosol release was also detected in the cooling period. The concentration peaks in the pre-oxidation, oxidation and cooling period were registered by two particle counters at the same time. After cooling, the measured concentration returned to the initial value, no saturation or contamination of the devices took place during the measurements. An example of the peaks of the aerosol mass concentration measured by APC-03-2 counter at the oxidation and cooling period is shown in Fig. 2.

There were aerosol particles on each collector plate, but the coverage of the Si surfaces by aerosol particles was different for the aerosol samples taken at different time periods. The greatest number of particles and also the largest ones were settled on the plates of the first impactor. This unit was working in the high temperature escalation period of the air ingress. The degree of coverage was decreasing by increasing time duration, but there was a smaller 'peak' between the 7th and 8th minute after starting the aerosol sampling with the impactors. At the end of the collection phase, only small amount of particles settled. These findings correspond to the results of the laser particle counting.

On the surface of each collector plate, both individual aerosol particles and aggregates could be distinguished. The shape of the particles was mostly slightly elongated. Long plates and rectangular grains were also found. All these features can be seen in the SEI image of Fig. 3. The size of the single aerosol particles ranged from about 0.1 μm up to a few μm and usually did not extend over 5–6 μm. Larger sizes (several tens of μm) were found for aggregates and for long plates.

All these were quantified by the results of image analysis performed on backscattered electron images (BEI) of some aerosol samples. Fig. 4 shows histograms of the mean Feret diameter of aerosol particles measured at magnification of 1000 times for impactor samples 1, 5 and 10. Percentage probabilities of the particles in various diameter intervals were plotted by particle number and particle area, respectively (Figs. 4(a) and (b)). There was a maximum for the large size particles (aggregates) for impactor sample 5 compared with the ones found for samples 1 and 10. This peak will be discussed later in the microanalysis part. It can also be recognised that for particle sizes of 5–10 and 10–15 μm, sample 10 had more particles than samples 1 and 5. This corresponds to the findings of the laser particle counters, where the abundance of the larger sized particles was increased during the cooling down period. The SEM images have shown that large platelets were mostly found at the beginning of the sample collection, however some amounts of plate-like structures were revealed in almost every sample. This was proven by the results of particle shape (see Fig. 4(c)). Histograms of samples No. 1, 5, 6, 7 and 10 proceed almost together, i.e., there was only small difference in the shape of the aerosol particles. At the beginning of the sample collection (i.e., for sample 1), the amount of elongated

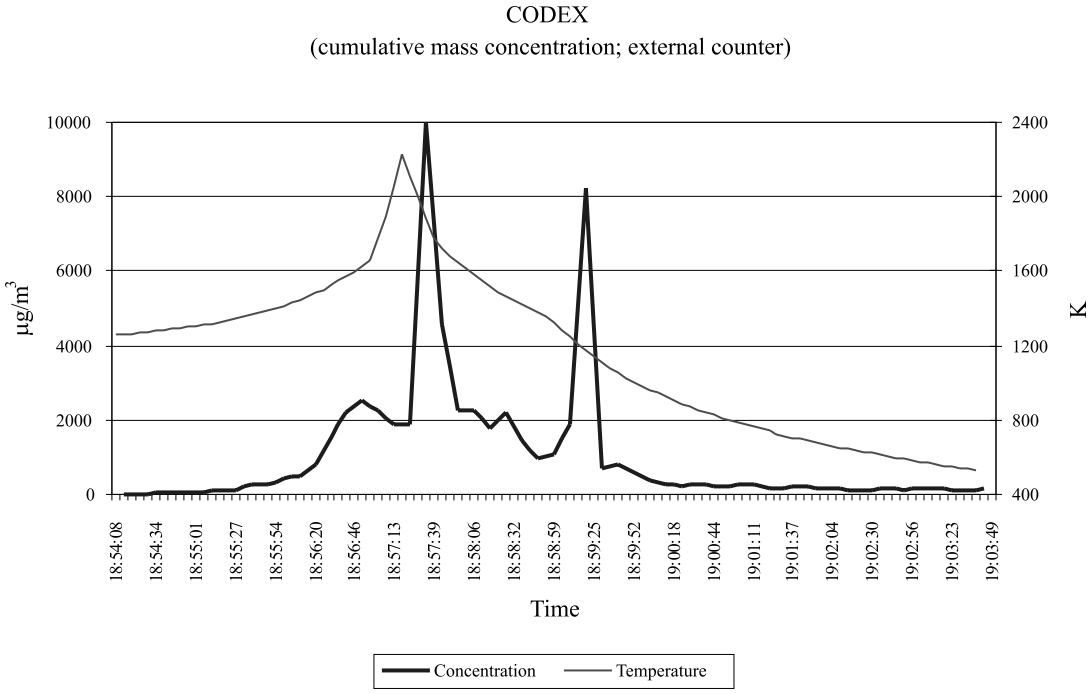

Fig. 2. On-line aerosol measurement during the air ingress and cooling down phase of AIT-1 test.

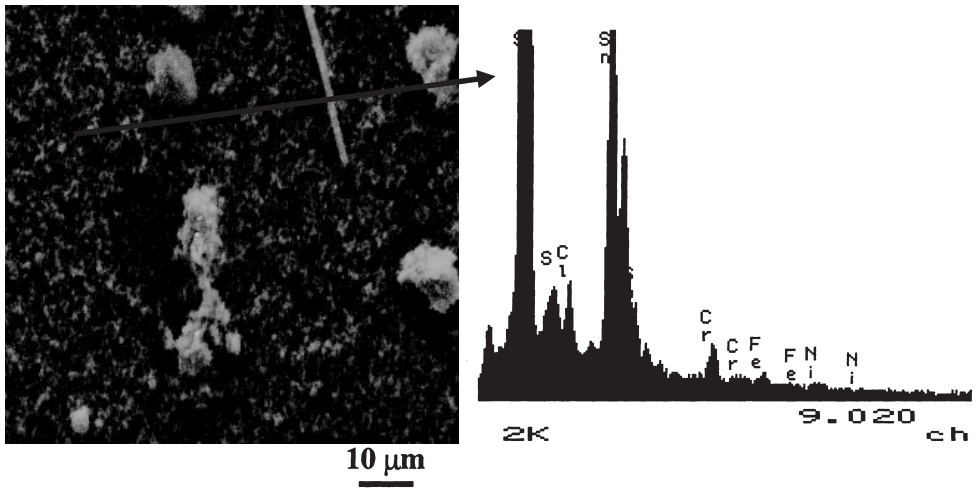

Fig. 3. SEM image and ED spectrum of impactor sample No. 1 in AIT-1 test.

particles was larger. Sample 5 had particles with shape more closely to the sphere than in the other samples. However the shape of the aerosol particles deposited in different time periods did not show very large differences than the size of them. All these findings are corresponding to the results of the laser particle analyser where a second peak was detected for the larger particles between samples 6 and 8.

3.1.2. Elemental composition of aerosol particles

U-rich particles with a few times 10^{-11} g were detected on impactor sample 1 (during the high temperature escalation) by EDS. Uranium was present mostly in single particles (1–3 µm) with rectangular shape. The uranium content of the aerosols settled on the filters was below the detection limit of SSMS (1 ng). On the Ni impaction plate, 7 ng/cm^2 uranium

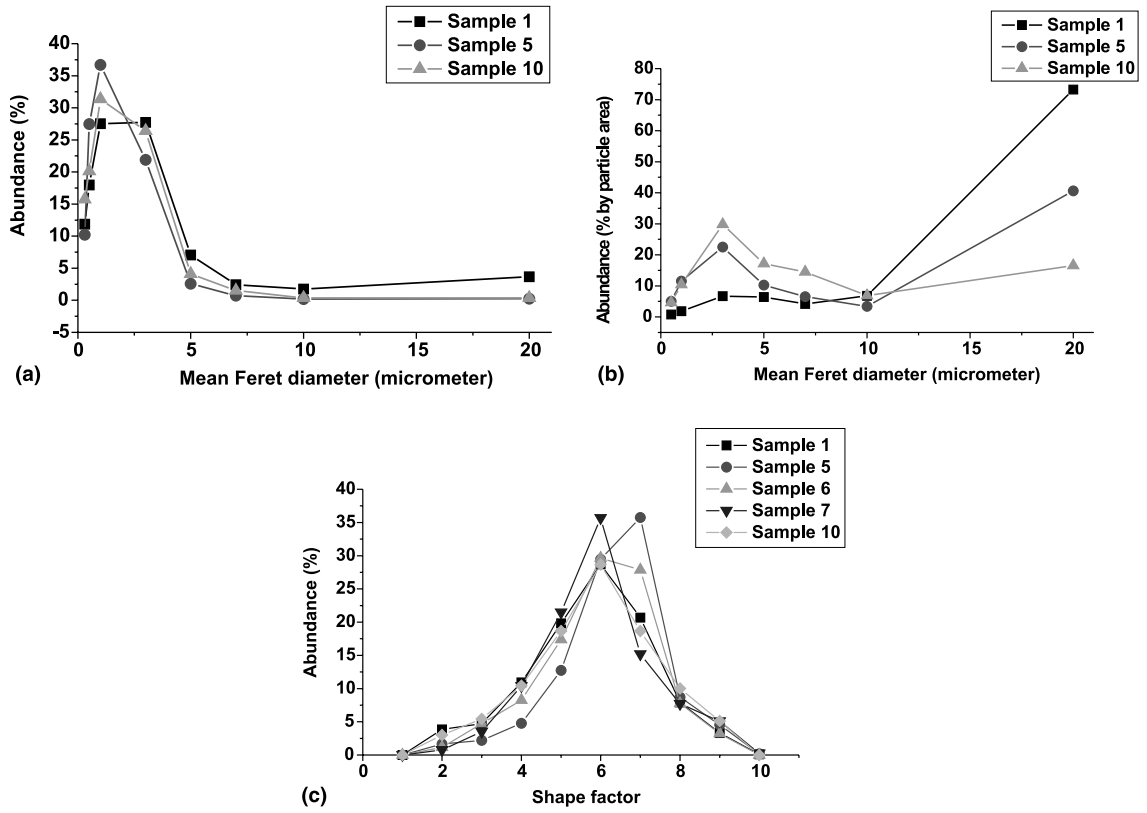

Fig. 4. Results of image analysis for some aerosol samples collected by impactors in AIT-1 test: (a) mean Feret diameter plotted by particle number; (b) mean Feret diameter plotted by particle area; (c) shape factor.

was detected by SSMS. On the basis of these findings, the total uranium release could be assessed as a few tenth of μg.

Components of the cladding, the spacer and some impurities from the insulating materials were found by EDS and SSMS as well. The following elements were detected frequently: tin, zirconium, iron, chromium, nickel, niobium (see Fig. 5) and some other elements such as aluminium, silicon, magnesium, sulphur, chlorine etc. The amounts of these elements were the highest for impactor sample 1 – except Zr and Nb which had maximum value for samples 7 and 5, respectively (Fig. 5). The quantities of these important elements were decreased during the cooling down phase, however for most of the components, there was a peak at filter 7.

There is a good correlation between the impactor and laser particle counter results showing a second maximum between impactor 6 and 8 for all particle sizes and especially for those below 1 μm (Fig. 6). It can be recognised that the number of the small sized particles (0.3–0.5 μm and 0.5–1 μm) decreased with the time in the same way as the tin concentration (see Fig. 5). Presumably, tin was enriched in aerosol

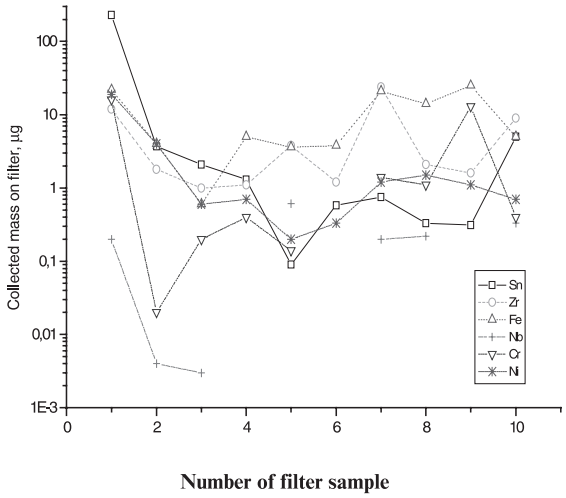

Fig. 5. Distribution of some elements collected by quartz filters in AIT-1 test.

particles of small sizes. They were collected mostly by the filters of the impactor assemblies. The increase of the particle number and element concentration

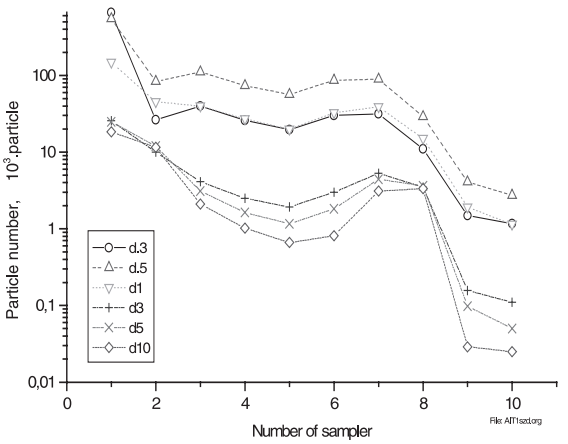

Fig. 6. Number of particles measured by internal laser particle counter during impactor sampling in AIT-1 test.

between impactor 6 and 8 reflects a strong post-excursion emission, probably from thermo-mechanical stresses in the oxide layer. Similar effect was observed in our separate effect test for other high temperature aerosol experiments.

By both EDS and SSMS, the largest amount of element was detected for tin – 230 μg in filter 1 and about 200 μg on collector plate 1, in impactor No. 1. Tin was enriched in small sized particles (mostly below 1 μm) as mentioned above. All the results correspond to each other showing significant (about three orders of magnitude) decrease in the tin content and in the number of the small sized aerosol particles collected at the beginning of the cooling down. Later, there was a second peak both in tin (and in almost every element) concentration and in the number of the small sized particles at filter 7. Making a rough estimation, it might be supposed that 2% of the total tin content escaped from Zircaloy 4.

Zr was found by EDS both in platelets and in particles with a few μm size. The highest amount of platelets were found on collector plate No. 1, while Zr-rich particles were more frequently detected at later phases of the aerosol sample collection (i.e., during cooling down). This can be correlated with the SEM – image analysis result showing increasing amount of relatively large particles, for e.g., impactor samples 5 and 10, respectively (see Fig. 4(b)).

By SSMS, it was found that the amount of zirconium decreased monotonously only at the first few filters, then it changed irregularly. This element was present in larger particles – proved by SEM and EDS – those numbers had larger statistical fluctuations. The relatively large amount of Zr found on the last filter (No. 10) could be correlated with the EDS results described above. These Zr containing particles came probably from the molten β-zirconium which was found to be missing from some parts of the cladding. This was proven by studying cross-sections of the bundle at some elevation.

The maximum Nb content (Fig. 5) – which corresponds to the second peak in the Zr content at filter 5 – might have originated from the shroud.

Some other elements – such as Ta and W – were also found by EDS and SSMS. They were detected mostly in impactor sample 1, i.e., during the high temperature escalation. There were only few particles having these elements, their quantities were between 10^{-11}–10^{-10} g. Tantalum and tungsten are structural materials used in the test facility. Ta was the shielding material of thermometers, while the heating rods were made of W.

3.2. Aerosols released in AIT-2 experiment

3.2.1. Size and shape of aerosol particles

During the test, the highest aerosol release was achieved at the moment of the steepest rise of the temperature slope. The cool down period was accompanied again with a change of particle composition and size distribution.

An example of the measured results of the cumulative mass concentration measured by PAPC-03-2 (internal) particle counter during the AIT-2 test together with the temperature curve is shown in Fig. 7.

The mass increase of the impactor samplers had a peak value of 0.81 mg at samplers No. 4 and 8. (see Fig. 8). This result could be correlated with the one obtained by the internal laser particle counter (Fig. 7). On the basis of the fractions collected on the quartz fibre filters, the total aerosol mass flow rate was estimated to be 0.6 g/min. In the pockets 10.4 mg, while in the cooler samplers 21.3 mg aerosol was collected.

Fine particles (below 1 μm) were settled more frequently on the Ni impaction plate, than on the collectors. It was found that the particle number was much less for the collectors of this test than for the first experiment. Particles below 0.5 μm were settled on the Ni impaction plate, while they were not settled on the Ni plate used in the first test. In the second test, the greatest number of particles was found on impactor No. 4. It was working during the highest temperature region of the test. There was a second maximum in the aerosol particle number and mass, between collector samples No. 6 and 8. This corresponds to the second maximum in the curve of the particle cumulative mass measured by the internal particle counter (see Fig. 7). Impactors 9 and 10 were working in the cooling down phase of the test. The coverage of their collectors by particles corresponds to the last two maximums in the curve of the cumulative particle mass mentioned above. The degree of coverage of the Ni impaction plate was found to be changing in

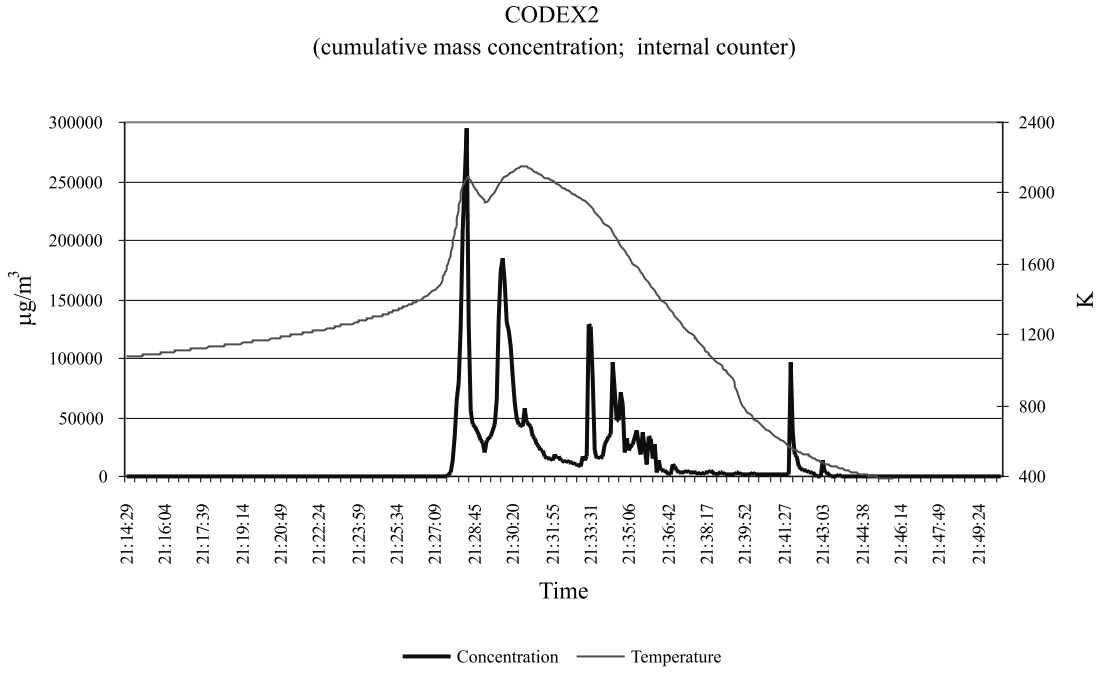

Fig. 7. On-line aerosol measurement during the air ingress and cooling down phase of AIT-2 test.

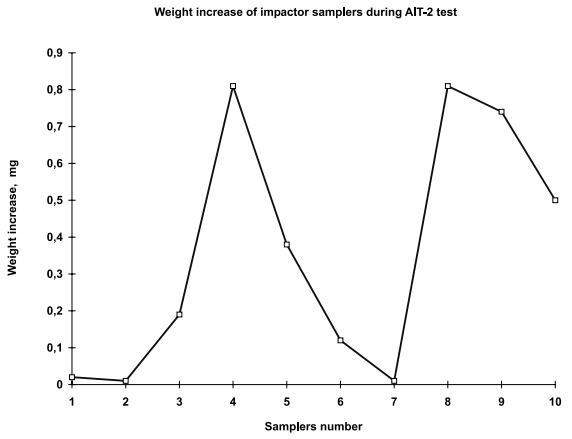

Fig. 8. Results of weight measurements of impactor samples used in AIT-2 test.

function of the place. The middle part was more densely covered than the edge.

Similar to the first test, both individual aerosol particles and aggregates could be distinguished by SEM on the surface of the collector plates. The size of single particles has started at about 0.8–1 μm and generally has not extended over 5–6 μm. On the Ni impaction plate, particles below 0.5 μm could also be found. Both for the cooler sampler and for the Ni impaction plate, the relative frequency of particles between 1 and 3 μm was found to be enhanced, while in the first test it was found for particles between 0.5 and 1 μm. The results of image analysis are shown in Fig. 9. It can be seen that the largest particles, aggregates were settled on the upper part of the cooler sampler and of the Ni plate, while at the edge, the largest particles were found on the lower side of the plate. The reason is that the middle upper part of the impaction plate was almost opposite to the aerosol stream, while the edge was further from the direct stream.

The particle shape was similar for the samples collected in the two experiments, however the abundance of the more elongated particles was slightly larger for the first test than for the second one (see Fig. 9(c)). Small differences can be found for samples collected on the upper and lower part of the cooler sampler, i.e., there were greater number of particle with more elongated shape on the upper part, where the largest particles were settled.

3.2.2. Elemental composition of aerosol particles

About two orders of magnitude more uranium could be detected in the aerosol released in AIT-2, than in AIT-1.

In the aerosol samples taken in AIT-2, uranium could be found both in single particles (1–3 μm) and in aggregates (5–10 μm) (see Fig. 10). The shape of the particles was mostly rectangular. The aggregates had

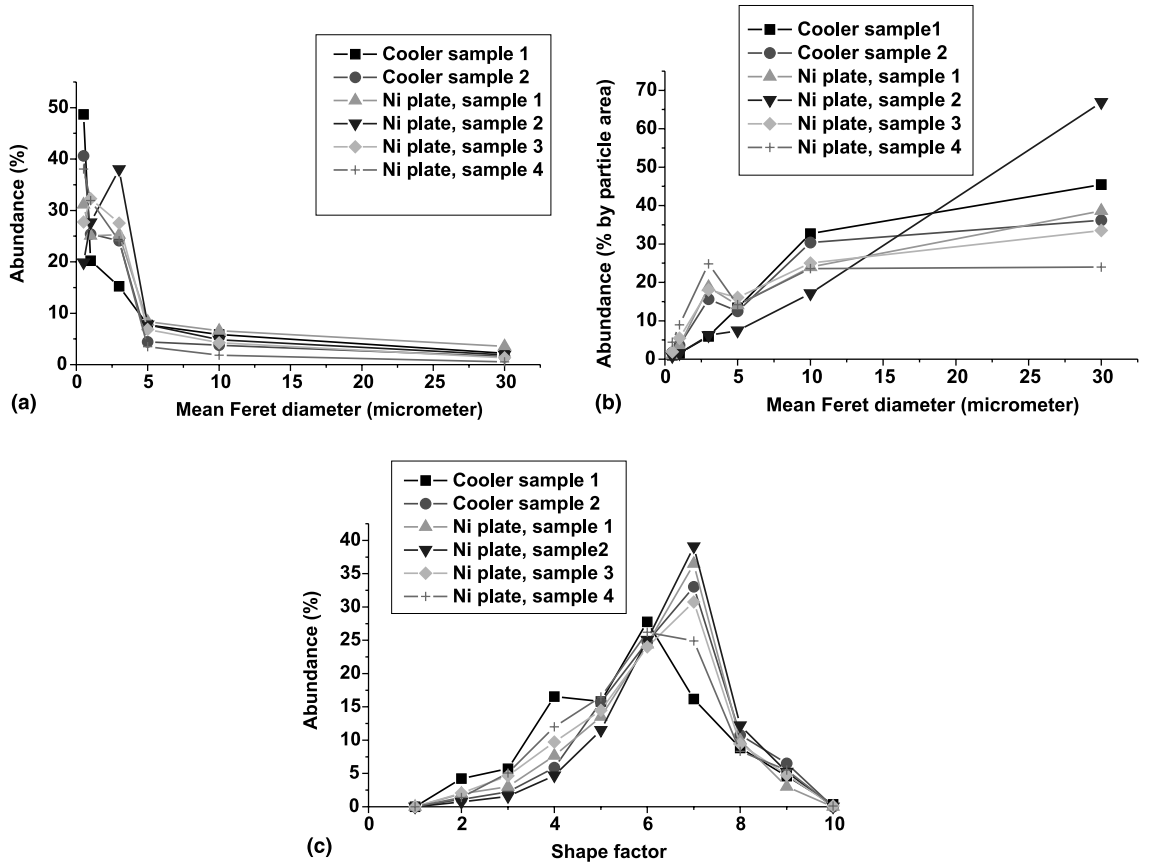

Fig. 9. Results of image analysis for aerosols collected in various parts of the impaction plate in AIT-2 test: (a) mean Feret diameter plotted by particle number; (b) mean Feret diameter plotted by particle area; (c) particle shape.

elongated or irregular forms. They contained other elements such as Si, Al, Fe, etc. besides uranium.

More uranium could be found on the Ni impaction plate than on the collectors. SSMS analysis of the quartz fibre filters indicated no uranium content. On the impaction plate, 0.07 μg/cm^2 uranium was determined by SSMS. In particles collected by pockets of the impaction plate, 1.5 μg, while in the cooler sampler 0.65 μg uranium was measured by SSMS. The total uranium release from the bundle of AIT-2 was estimated to be 50–100 μg.

Besides uranium, components of the cladding, of the spacer, of the heating rod and of the other structural materials were detected. Both by EDS and SSMS, the most frequently found elements were the zirconium and the iron. By SSMS analysis of the quartz fibre filters, 1–7 μg/ min total escaping rate was found for Zr and Fe, and about one order of magnitude lower values were detected for Sn and Ni (Fig. 11). Quantities of the Sn, Fe and Ni had maximums at filters No. 3 and 8 (or at No. 7 for the Fe), which corresponded to the mass increase found both by weighting of the impactors and by the time dependence of the aerosol mass by the laser particle counter. The same tendency was found by EDS studies of the aerosol particles settled on the collectors.

The larger lamella-like particles collected by the pockets and by the cooler samplers were formed mainly from oxides of Zr, Si, Al, Fe and Cr with decreasing concentration in this order (see Table 2). There is a correspondence between the results of EDS and SSMS got for these lamellas.

The Ni impaction plate and the Ni sheets of the cooler sampler had mainly Mn, less Fe, Zn, Sn and W stacked to the surfaces. The quantities extended from 1 to 200 μg/cm^2.

4. Summary and conclusions

The two experiments with slightly different conditions resulted in rather different results regarding the amount and composition of the aerosol emitted. In the first test, the highest emission was found for tin and the uranium release was supposed to be a few tenth of μg. It

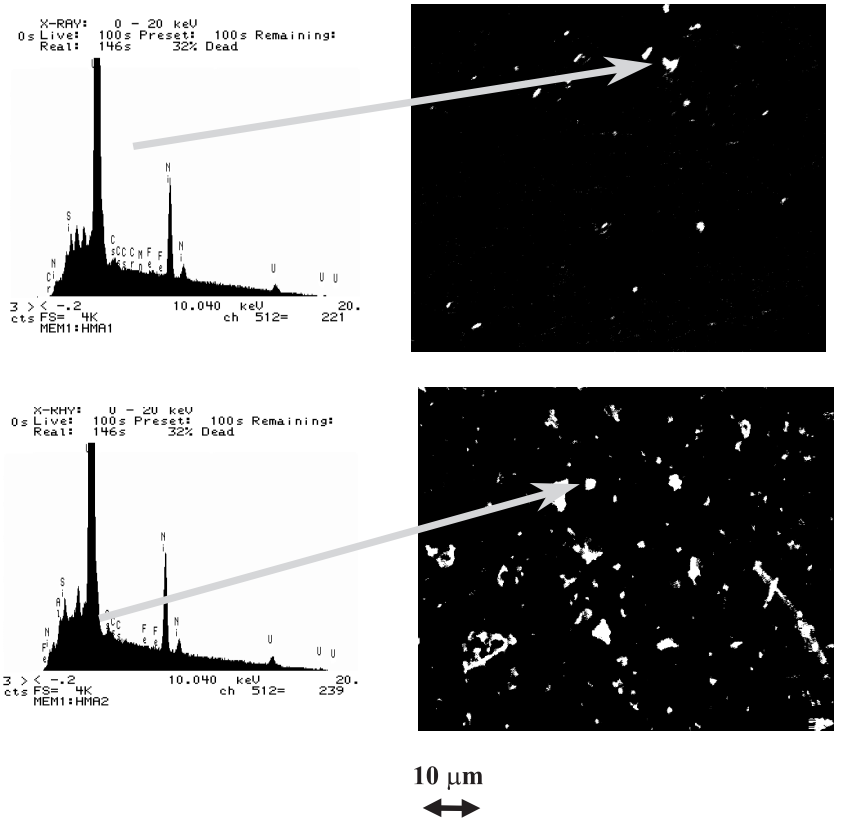

Fig. 10. SEM images and ED spectra of some U-rich particles collected in AIT-2 test.

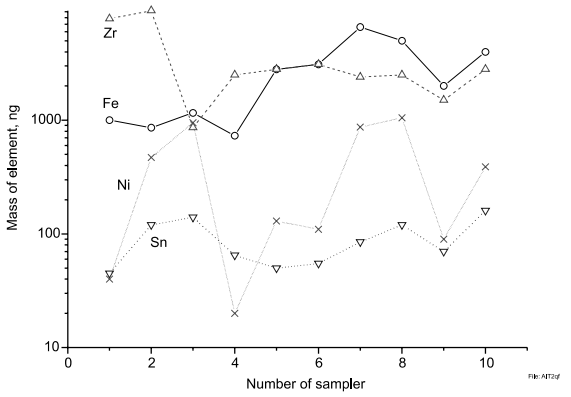

Fig. 11. SSMS results for some elements collected by filters in AIT-2 test.

seems that the shorter duration of air ingress was not enough to maintain the high temperature to degrade seriously the fuel bundle. Also, interaction between the fuel and the cladding occurred only in some parts. About two order of magnitude more uranium was released in the second experiment. All the results gained by laser particle counters, SEM, EDS and SSMS correspond to each other regarding the amount, the size and/or composition of the aerosols released. The results

Table 2
SSMS results measured for aerosol collected by the pocket and cooler samplers in AIT-2 test

Element	Pockets on impaction plate (μg)	Cooler sampler (μg)
Mg	100	150
Al	1200	850
Si	1800	2000
Ca	8	4
Cr	160	500
Mn	30	100
Fe	850	1400
Ni	60	60
Zn	3	5
Zr	1300	6000
Mo	1.4	3
Sn	36	90
Hf	20	30
Pb	2.5	1.5
W	2	15
U	1.5	0.65

of both AIT experiments are unique because the consequences of the air oxidation were not yet investigated in integral facilities.

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