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## Research Article

# Fission Product Transport and Source Terms in HTRs: Experience from AVR Pebble Bed Reactor

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Fission products deposited in the coolant circuit outside of the active core play a dominant role in source term estimations for advanced small pebble bed HTRs, particularly in design basis accidents (DBA). The deposited fission products may be released in depressurization accidents because present pebble bed HTR concepts abstain from a gas tight containment. Contamination of the circuit also hinders maintenance work. Experiments, performed from 1972 to 88 on the AVR, an experimental pebble bed HTR, allow for a deeper insight into fission product transport behavior. The activity deposition per coolant pass was lower than expected and was influenced by fission product chemistry and by presence of carbonaceous dust. The latter lead also to inconsistencies between Cs plate out experiments in laboratory and in AVR. The deposition behavior of Ag was in line with present models. Dust as activity carrier is of safety relevance because of its mobility and of its sorption capability for fission products. All metal surfaces in pebble bed reactors were covered by a carbonaceous dust layer. Dust in AVR was produced by abrasion in amounts of about 5 kg/y. Additional dust sources in AVR were oil ingress and peeling of fuel element surfaces due to an air ingress. Dust has a size of about 1  $\mu\text{m}$ , consists mainly of graphite, is partly remobilized by flow perturbations, and deposits with time constants of 1 to 2 hours. In future reactors, an efficient filtering via a gas tight containment is required because accidents with fast depressurizations induce dust mobilization. Enhanced core temperatures in normal operation as in AVR and broken fuel pebbles have to be considered, as inflammable dust concentrations in the gas phase.

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## 1. INTRODUCTION

Fission product transport during HTR normal operation plays a major role in safety examinations for normal operation and for accidents as also in maintenance and in dismantling. Particularly the fission products released during reactor normal operation and accumulated within the coolant circuit are most relevant contributions to the source terms of design basis accidents (DBAs) for advanced small HTRs. These activities are released into the coolant circuit during long-term normal operation at high temperatures from intact- and defect coated fuel particles and from uranium contamination in the graphite by diffusion through kernel, coating layers, and graphite (metallic fission products) as well as by diffusion out of the kernels of defective coated fuel particles without further retention (nonmetals). The intact coated particle is a very efficient barrier for nonmetals, but low melting metallic fission products diffuse through intact coatings, particularly at temperatures  $>1000^\circ\text{C}$ . Condensable

fission products are mainly deposited in the coolant circuit: plate out on metals competes with sorption on graphitic dust. Except for noble gases, the gas-borne activities are low in steady-state operation due to this deposition.

Advanced modular HTR abstain from a gas-tight containment. Accordingly, there remains only one barrier between the accumulated activity and the environment, the coolant circuit enclosure. DBA-like depressurizations, stand alone or followed by a core heat-up phase, and steam/water ingress accidents may lead to a partial release of these activities accumulated in the coolant circuit [1, 2]. Another still relevant source term contribution is the activity inventory of coated particles with defective coatings (defect fraction: about  $10^{-4}$ ). Nonmetals as iodine are partly released from defective coated particles in course of a water ingress accident due to an interaction between kernel and steam. This holds also for oxide fuel [2, 3].

In contrast, the activity content of particles with intact coatings, which is calculated for modern HTRs to about

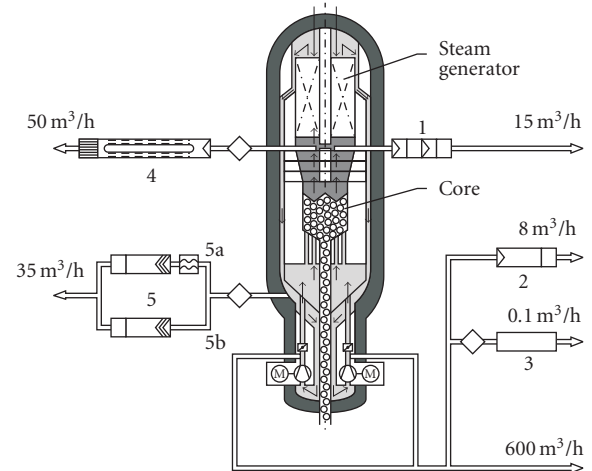
99.9% of the total inventory, does not virtually dominate DBA source terms of advanced small HTRs. Intact coated particles are not affected by stand-alone depressurizations and by water ingress events of the DBA range. However, there are some limited releases from fuel elements by accelerated diffusion and by additional temperature-induced particle failure in case of core heat-up events. Except for Ag-110 m, these releases remain low if core temperatures, as claimed by design, do not exceed 1600°C, and if enhanced temperatures do not last longer than about 200 hours. The total inventory of Ag-110 m is small for uranium fuel but larger for plutonium fuel. Another release-limiting factor in core heat-up events is caused by the fact that high temperatures and corresponding releases occur subsequent to the depressurization of the coolant circuit: for that there remains a very limited gas transport out of the coolant circuit, when high temperatures are reached and releases start. Further, releases out of the reactor building into the environment can be filtered because of the small overpressure in the high-temperature phase. The only credible accidents, which may induce a substantial fission product release from intact coated particles, are severe air ingress with graphite burning [4] and rare reactivity transients. However, these accident are far outside of the DBA range of well-designed HTRs. Source terms related to the inventories outside of intact coated particles dominate, therefore, doses in DBA and probably even the risk of small advanced HTRs [1, 2].

Fission products accumulated in the coolant circuit have also to be taken into account for maintenance purposes. This is particularly relevant for the gas turbine in a direct cycle, which requires periodically hands on maintenance. Contamination limits for gas turbines, allowing hand-on maintenance, are not easy to achieve in HTRs [5] as will be discussed in Section 4.3.

Dose limits for DBA are comparatively low and have to be calculated in a pronounced conservative manner. They allow only a very low activity release into the environment: following German requirements, the maximum tolerable release for key nuclides is—for conditions as assumed in the safety report of the HTR-Module200—in the range of about  $10^{-7}$  (Cs-137) to about  $10^{-8}$  (I-131 and Sr-90) of the total nuclide inventory in that 200 MWth pebble-bed reactor design. Comparing these release limits with inventories present outside of intact coated particles of up to  $10^{-3}$ , which are subject of partial release in DBA due to nonexistence of a gas tight containment, the relevance of fission product transport becomes obvious. Respective experience from the AVR pebble-bed reactor is discussed in this paper.

## 2. OVERVIEW ON AVR AND ITS FISSION PRODUCT RELATED EXPERIMENTS

The AVR was the first pebble-bed HTR worldwide and was operated 1967–1988 in Juelich. Main aim of this experimental reactor was the principal test of the pebble-bed core, test of the fission product transport in pebble-bed reactors, and test of many different types of pebble-shaped fuel elements. Design data of the AVR are collected in Table 1. Presently, AVR undergoes dismantling and its vessel will be



- 1 VAMPYR I (hot gas filter)
- 2 Cold gas filter
- 3 Noble gas measurement
- 4 VAMPYR II (plate-out experiment)
- 5 Dust experiment
- 5a With plate-out section upflow of the dust filters
- 5b Without plate-out section

FIGURE 1: Schematics of AVR and its fission product related experiments.

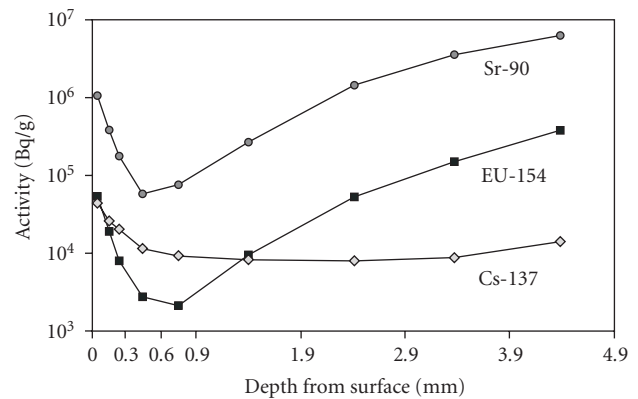


FIGURE 2: Concentration profiles of Cs, Eu and Sr in the outer fuel free shell of an AVR pebble fuel element (17.6% fima, initial U-235/total heavy metal ratio = 0.17).

grouted with light concrete in 2009. A rough scheme of the coolant circuit of this reactor is presented in Figure 1.

AVR was operated until 1974 at coolant outlet temperatures of up to 850°C. From February 1974, the maximum coolant temperature was increased to 950°C, temporarily even up to 990°C (see top part of Figure 4). Whereas the release of iodine and noble gases remained small after temperature increase, there was a pronounced acceleration of release of metallic fission products (Sr-90, Cs-137, Ag-110 m) by diffusion, which may have reached at end of life some percent of one core inventory. There are two contributions for this release: on the one-hand side until

TABLE 1: Main data of the AVR reactor.

Power/Average power density	46 MW <sub>th</sub> (15 MW <sub>el</sub> )/2.5 MW/m <sup>3</sup>
Cycle	Steam cycle with the steam generator inside the reactor vessel
Core height/diameter	2.8 m/3 m
Coolant/Pressure	He/10.8 bar
Gas outlet/inlet temperature	700 to 950 (990)°C/275°C
He-flow	13 to 16 kg/s (depending on desired gas outlet temperature)
Fuel	95000 pebbles (6 cm diam.) diverse fuel types, at end mainly modern TRISO

about 1975, mainly BISO fuel (two pyrocarbon coatings) with UC<sub>2</sub>-kernels and large uranium contamination of the graphite was used. From 1985, modern UO<sub>2</sub> TRISO fuel (one SiC layer between pyrocarbon layers) dominated the core composition. Oxide kernels reveal a better Sr-retention than carbide kernels and modern TRISO fuel elements improve the Cs-retention at least compared to some early fuel elements used in AVR. On the other hand, core temperatures in AVR were substantially higher than precalculated, as being discussed briefly in Section 6.2 and in more detail in [6], and have substantially accelerated the release of metallic fission products [6]. The problem to which extent enhanced releases of AVR have to be considered in future HTR concepts is studied in [6].

Figure 1 also contains the most relevant fission product-related experimental facilities [3, 7–10]. All these experiments were operated in a bypass to the main He circuit. These experiments are as follows.

- (1) VAMPYR-I hot-gas filter: The condensable nuclides activity in the hot gas was measured via a plate out section and of the dust activity in a subsequent dust filter. Materials used in the plate out section are not HTR ones. This is, because VAMPYR-I suffered from a strong neutron activation of the inner plate out section. Mainly for that, plate out sections consisting of Ti, which shows low activation, were used.
- (2) Cold-gas filter: The total activity (except noble gases, but including dust-borne activities also by a separate dust filter) was measured in the cold-gas range.
- (3) Noble gas measurement: These experiments analyzed the total noble gas inventory in the coolant.
- (4) VAMPYR-II plate out experiment in the hot gas region: The plate out section consisted of metallic materials representative for the THTR-300, and was equipped in certain experiments with dust filters upflow and/or downflow. VAMPYR-II was operated only for short periods at the end of AVR life.
- (5) Dust experiment: Dust filter equipment in the cold gas with the intention to examine the dust behavior more in detail, for example by several different filters parallel and in line. The dust experiment was operated from 1984 to 1988 only.

Unfortunately, most of these experiments were not completely evaluated during and in the first years after AVR

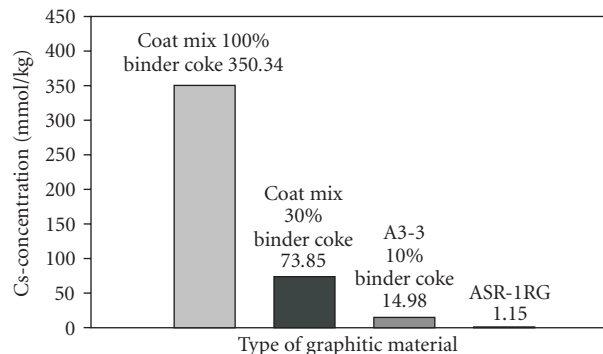


FIGURE 3: Equilibrium concentrations of Cs at 1100°C in different carbonaceous materials (Cs-partial pressure:  $3 \cdot 10^{-2}$  Pa).

operation. This paper gives an overview on the evaluation status of these experiments, including those recently performed. At first, experiments (1, 2, and 4) which cover fission product deposition and plate out, are discussed. Sections 5 and 6 concentrate on details of dust behavior (Experiment 5 and some dust-related results of other experiments) and some general AVR experience on fission product behavior, including Experiment 3. Besides these experiments, several other fission product-related experiments were performed (Tritium reduction by gettering and permeation filters, fission product absorption by metals [8]), which were however not completely successful and will not be discussed here.

### 3. AVR EXPERIMENTS ON FISSION PRODUCT DEPOSITION IN THE COOLANT CIRCUIT

Condensable nuclides released from fuel elements may be reabsorbed on fuel elements, absorbed on graphite reflectors and plated out on metal surfaces, or—in particular in pebble-bed reactors—absorbed on graphitic dust. These deposition mechanisms will be discussed here in detail on example of the AVR.

#### 3.1. Deposition of nuclides on fuel elements and on the graphite reflector

Fission product profiles in depth of such AVR fuel elements, which show a moderate release from coated particles, reveal typically a minimum in concentration in the outer fuel-free zone. This is an indication for fission product reabsorption

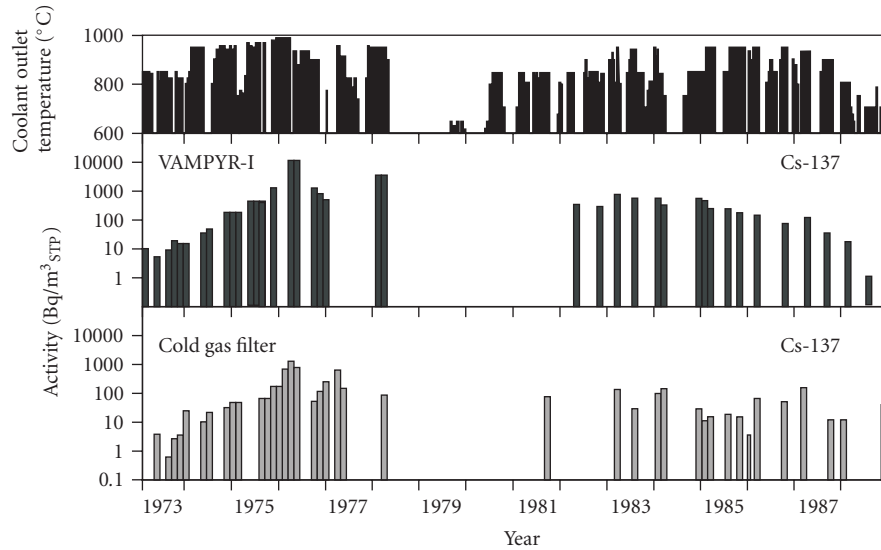


FIGURE 4: Comparison of total Cs-137 activities in AVR hot gas and in cold gas.

on fuel elements. Experimental profiles showing this concentration reincrease towards the surface are presented in Figure 2, which is for a GK (BISO-UC<sub>2</sub>) fuel element. In the AVR core, fuel elements of different type and burn-up were mixed together and accordingly release rates and sorption capabilities varied between neighboring fuel elements, which led to this reabsorption effect. In future reactors with more homogeneous fuel element composition, this effect is expected to become less pronounced. A second contribution may be caused by gas-borne dust-carrying fission products, which circulates in the primary circuit and will adhere on fuel elements. Altogether, the shape of the profile is in line with laboratory sorption experiments for Sr-90 and Cs-137 [3, 11].

As an example, Figure 3 compares the sorption capabilities of fuel element matrix graphite A3-3 as used in AVR, of a standard nuclear graphite ASR-1RG, and of 2 coat mix materials containing different amounts of binder coke, for Cs partial pressures as in accidents at 1100°C.

It becomes obvious that the sorption capability of matrix graphite is about an order of magnitude larger compared to the standard nuclear graphite ASR-1RG, which is—as shown by the comparison with the coat mix materials—due to the content of 10% of ungraphitized (coked) binder in the matrix graphite A3-3. The binder coke absorbs more than 2 orders of magnitude more Cs than the graphite filler, as results on the coat mix materials indicate. This result is beneficial concerning fission product retention in the core, but problematic due to the fission product sorption in the mobile graphitic dust. For Sr, the fuel element matrix graphite reveals an even larger sorption capability than for Cs [3, 11].

Specific activities of long-lived metallic fission products in the graphite/carbon brick core top construction indicate that about 10% of the total inventories of Sr-90 and Cs-137 released from active core are found in this component.

The large He flow velocity and the small geometrical surface of the reflector boreholes limit the mass transfer-controlled sorption, despite of the large sorption capability of graphite.

### 3.2. Deposition on the steam generator

Most condensable fission products are deposited along the steam generator, as comparison of data of the hot-gas filter VAMPYR-I and of corresponding data of the cold-gas filter indicates. This is due to the large geometrical surface of the steam generator. Figure 4 contains data (Cs-137) of both experiments for the AVR operation time 1973–1988: The corresponding average hot gas temperatures are also shown. Similar measurements are available for Cs-134 too. The Cs-134/Cs-137-ratio in VAMPYR-I decreased from 1973–1978 almost continuously from about 1 to 0.4, a value which remained constant in further operation. This observation cannot yet be explained, the respective average ratio in the fuel was about 0.8. The Cs-activity in the dust filter downflow of the plate out section was substantially smaller than the activity in the plate out section. This indicates that Cs occurs mainly in molecular form in the hot gas. This assumption is supported by examination of the Co-60 behavior: Co-60 occurs only dust borne (formed mainly by corrosion/abrasion from activated metallic components). Its deposition behavior in VAMPYR-II is very different from that of Cs (see Section 4.2 and Figure 8). Further the deposition rate of AVR dust is much smaller than that of Cs (see Figure 11 and Table 3). Also, the deposition profile of Cs along the steam generator of Peach Bottom HTR with block-type-fuel elements, which also contained some dust by oil cracking, indicates predominantly molecular Cs-137 and even Sr-90 in the hot gas [12].

VAMPYR-I has unfortunately not sampled hot gas of an average activity composition. Its data were probably influenced by sucking of cold-gas streams, as measurements

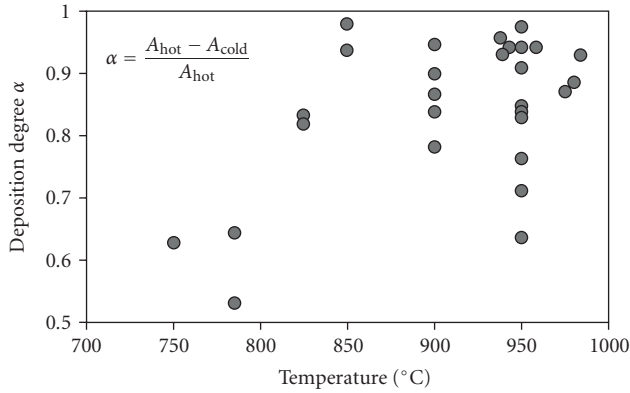


FIGURE 5: Deposition degree  $\alpha$  of gas borne Cs-137 activity ( $A$ ) per pass of coolant.

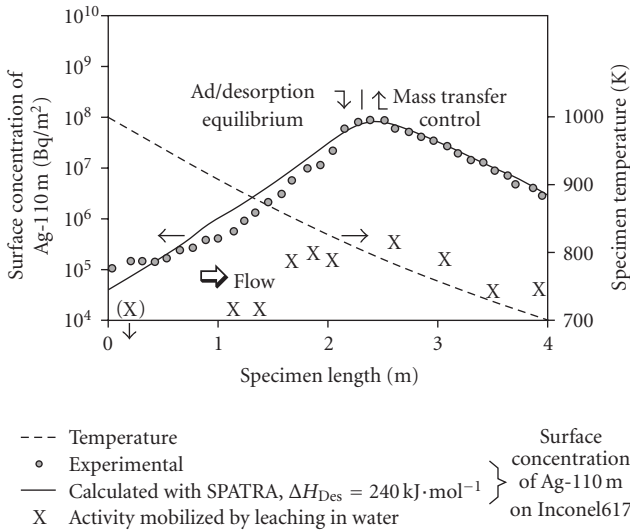
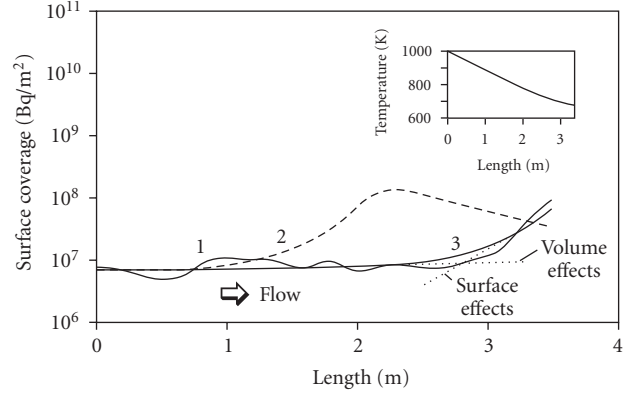


FIGURE 6: Measured and calculated plate-out profile for Ag-110 m on Inconel617 (VAMPYR-II).

of gas temperature profiles in the surrounding of the entrance of VAMPYR-I revealed [13]. The absolute activity values measured in VAMPYR-I were therefore too low.

This underestimation becomes obvious when the total deposition in the VAMPYR-II plate out experiment (see Section 4.2) is compared with respective VAMPYR-I results for about the same time period. Table 2 contains specific activities measured in VAMPYR-II 1987-88 [3] at hot gas temperatures of 700 to 930°C. The activity in the final charcoal filter of VAMPYR-II was unfortunately not measured, so the data presented in Table 2 are lower limits. This holds particularly for Cs-137: examinations of the 5 subsequent segments of the fission product filters of VAMPYR-II-02 revealed no downflow declination in Cs-137 concentration, which means that some unknown amount of Cs-137 passed this facility [14].

VAMPYR-II sucks gas under well-defined conditions, that is, gas of about average hot gas temperature. Having in mind that maximum coolant temperatures at the azimuthal



- 1 Experimental data (Cs-134, -137)
- 2 SPATRA with LAMINAR-loop data ( $\pi_p = 10^{-5}$ ,  $\Delta H_D = 235 \text{ kJ}\cdot\text{mol}^{-1}$ )
- 3 SPATRA with  $\pi_p = 10^{-5}$  and  $\Delta H_D = 180 \text{ kJ}\cdot\text{mol}^{-1}$  (best fit)

FIGURE 7: Cs plate out in VAMPYR-II compared to calculations.

position of VAMPYR-II were measured to more than 100 K higher than average ones [13], an additional reason for underestimations of coolant activities by VAMPYR-II may exist. This is because fission product release follows an exponential temperature dependence. In spite of this underestimation the specific hot gas activities of Cs in VAMPYR-II are still higher than corresponding ones in VAMPYR-I by a factor of 30 at 700°C and 3 at 930°C.

VII-01 data correspond concerning Cs-137 to a release rate of greater than  $10^{-3}$  of the birth rate. The respective value for Ag-110 m amounts to  $2\cdot 10^{-2}$ . Obviously, the main contribution stems from intact-coated particles by diffusion through coatings, because in 1987 the uranium contamination of fuel element graphite and the fraction of defective particles were altogether less than  $2 \times 10^{-4}$  of the total inventory (see AVR Quarterly Progress Reports 1987 and 1988 in [15]). Even the Cs-releases of VII-03 and VII-04 at 700°C can be explained by defect particles and uranium contamination only, if a complete Cs release out of these fractions is assumed, which is unlikely. Accordingly, diffusion through intact coatings has contributed here, too. Mainly unintentional high core temperatures (see Section 6.2) have caused these high rates [6].

Absolute activity values of the cold-gas filter are also assumed to be too low due to deposition along a 30 m pipe guiding to the filter. Comparison of cold-gas filter data with those of the dust filter (10 m pipe length) indicates an average underestimation by a factor of 2 with a large uncertainty scatter.

Activities deposited in the cold-gas filter were mainly found in dust except of iodine. The dust filters in AVR cannot distinguish between molecular activities sorbed on dust present in the filters, or already reach the filter bound on dust. However, the dust concentration in these filters is so small (some mg) that a large sorption effect of molecular

TABLE 2: Lower limits of specific hot gas activities [ $\text{Bq}/\text{m}^3_{\text{STP}}$ ] as measured in VAMPYR-II (1987–88) for hot gas temperatures as given in brackets.

Nuclide	VII-01 (930°C)	VII-02 (900°C)	VII-03 (700°C)	VII-04 (700°C)
Cs-137	402	112	57	55
Ag-110 m	126	48	5	4

TABLE 3: Deposition degree  $\alpha$  in AVR for selected nuclides measured in 1982–1984.

Nuclide	$\alpha$
Ag-110 m	0.87
I-131	0.80
Cs-134 and Cs-137	0.80–0.98

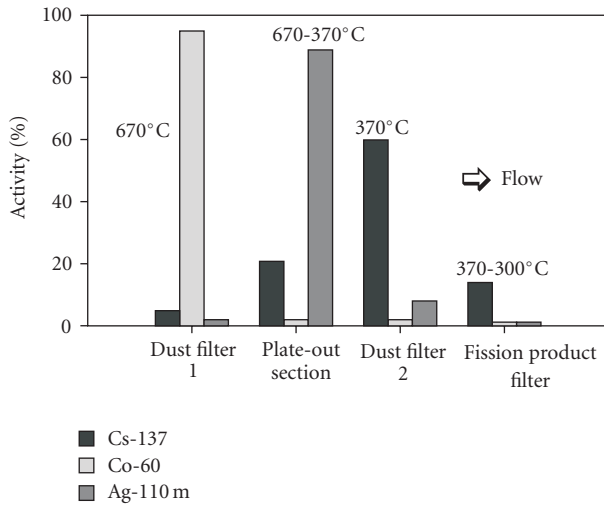


FIGURE 8: Activity distribution in VAMPYR-II-04.

activities on dust has not to be expected. Most probably Cs occurs in molecular form in AVR hot gas but dust borne in the cold gas.

### 3.3. Deposition of nuclides per pass in normal operation

On basis of the data shown in Figure 4, the deposition degree  $\alpha$  of Cs in the steam generator region was calculated.  $\alpha$  is shown in Figure 5 depending on hot gas temperature. There is some tendency of  $\alpha$ -increase with increasing hot gas temperatures; one reason for that might be that at higher hot gas temperatures, the flow velocities are smaller and therefore the residence times of the coolant in the deposition range are larger. An average deposition degree of about 0.85 is found. However, consideration of experimental errors as estimated in Section 3.2 for VAMPYR-I and for the cold-gas filter will enlarge average  $\alpha$ -values to about 0.95. Table 3 contains some estimates on  $\alpha$  for relevant nuclides as found in AVR 1982 to 1984 [7].

Computer simulations based on laboratory scale plate out measurements on metals with codes like SPATRA [14]

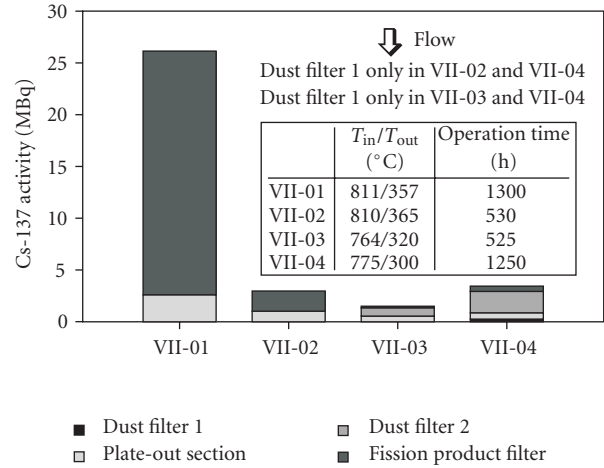


FIGURE 9: Cs distribution in VAMPYR-II experiments.

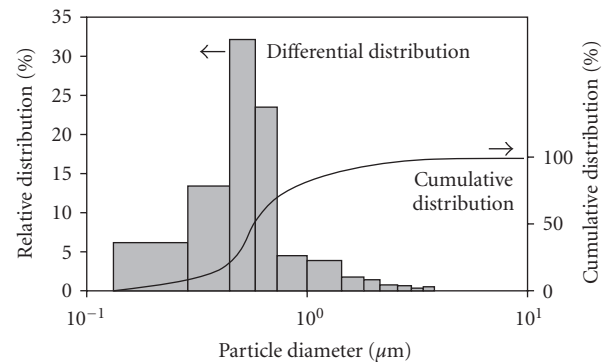


FIGURE 10: Number weighted size distribution of AVR dust.

lead to much higher Cs-deposition degrees ( $\alpha$  greater than 0.99), if no bypasses are considered. The deposition rate is controlled by mass transfer in the boundary layer, except for the hottest part of the steam generator, where an ad/desorption equilibrium is reached. Similar results are obtained, even if very low deposition parameters—as derived in Section 4.2 from VAMPYR-II-data—are taken into account. This discrepancy can only partly be explained by steam generator bypass flows. A similar overestimation of  $\alpha$  in deposition calculations was found for several other nuclides in AVR and also in other HTR-reactors like DRAGON. Here, experimental  $\alpha$ -values for I-131 as low as 0.25 are reported [16], whereas the Cs-deposition was similar to AVR. A very low  $\alpha$  for I-131 was observed also in the COMEDIE loop [17]. This indicates that fission product chemistry (i.e., the chemical form of the nuclides is not

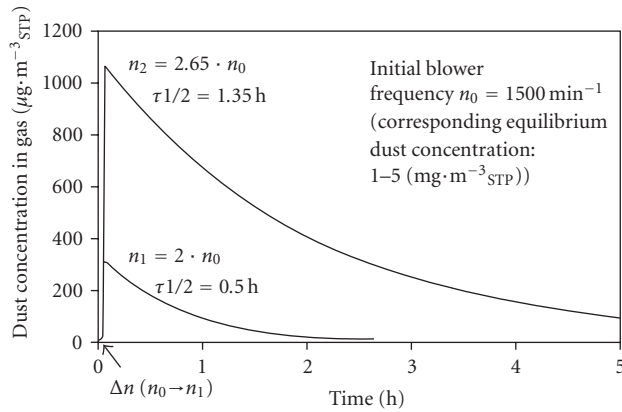


FIGURE 11: Dust concentrations in AVR in blower transient T3 (measured by dust filter experiment).

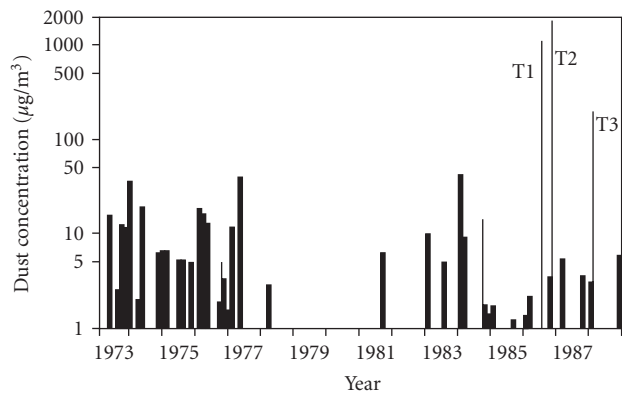


FIGURE 12: Gas borne dust concentrations in AVR operation (dust filter of the cold gas experiment).

the elemental one) and/or particularly in case of pebble-bed reactors the nuclide interaction with dust may play a relevant role in the overall deposition behavior, as is discussed in Sections 4 and 5. Overestimation of  $\alpha$  underestimates the steady-state gas-borne activities.

The  $\alpha$ -value of Sr-90 in AVR (1973-1974) was estimated to less than 0.5. This may be due to the very strong affinity of Sr to dust. However, Sr-90 measurements in VAMPYR-I are not available from mid of 1974 when Sr-90 measurements were replaced by those of Eu-154, which creates some errors. In addition, as will be discussed in Section 5, the dust sampled by the cold-gas filter and by the dust filter contains not only the activities released in the sampling phase but to some extent also activities from prior release periods. This holds particularly for Sr-90.

#### 4. AVR EXPERIMENTS ON PLATE OUT OF CONDENSABLE NUCLIDES ON METALS

##### 4.1. Experimental results

The main facility for generation of fission product plate out data in AVR was VAMPYR-II (Figure 1), which contained an annular plate out section. The inner slab consisted of

Inconel617, whereas the tube was made from Incolloy800H. The total length of the plate out section was about 3.5 m, the diameter of the slab was 9 mm and the inner diameter of the tube was 16 mm. Gas heating allowed operation at similar temperature conditions. Nevertheless, the gas inlet temperature of individual experiments (see Figure 9) varied slightly from 765 to 810°C depending on the average hot gas temperature which is given in Table 2. Maximum wall temperatures of the plate out section were between 660 and 725°C. A temperature gradient along the length of the plate out section of about 100 K/m was maintained by heaters. In contrast to VAMPYR-I, VAMPYR-II was installed in a range without strong activation by neutrons. There were 4 plate out experiments performed in VAMPYR-II; their duration was between 1 and 2 months. Main nuclides examined in VAMPYR-II were Cs-134, Cs-137, Ag-110 m, and Co-60. Some limited data on I-131 are available for the first VAMPYR-II experiment. Two of the VAMPYR-II experiments (2 and 4) were equipped with a dust filter upflow of the plate out section. Experiments 3 and 4 contained a dust filter downflow. Also, all experiments contained a so-called fission product filter behind the plate out section or the second dust filter, which however did not work properly [14].

A general result of all VAMPYR-II experiments was that Ag-110 m behaves as expected from classic plate out theory, which means ad/desorption equilibrium at high temperatures and mass transfer-controlled plate out at low temperatures which means that the plate out section acts at low temperatures as a perfect sink [14, 18]. Figure 6 contains as an example the deposition profile of Ag-110 m in Experiment 4 on Inconel617 together with postcalculations. A physical explanation of the plate out model, used, for example, in the SPATRA plate out code, is discussed in ANNEX I of [2].

Model calculations with a desorption energy of 240 kJ/mol easily fit the measured profile. An upper limit of a “penetration coefficient” of  $10^{-6}$  (see [14] and annex I of [2]) was extracted from these experiments too. The penetration coefficient takes volume effects as absorption in the metal bulk into account. Profiles of similar shape were obtained in the other three VAMPYR-II experiments. Additional post examinations on the removal of plated out Ag-110 m by water leaching were performed and are presented in Figure 6 too. Only a few percent of the plated out activity can be removed by water leaching, that is, decontaminations are not easy to perform. This is a relevant finding particularly for maintenance and repair work in direct cycle facilities, where plated out Ag-110 m belongs to the dose-determining nuclides (see Section 4.3).

The more surprising output of VAMPYR-II is connected to cesium: in all VAMPYR-II experiments, virtually flat (temperature independent) deposition profiles were observed, connected to a very low deposition degree in the plate out section as shown in Figure 7. This deposition behavior can be interpreted by a low desorption energy in conjunction with a penetration coefficient of  $10^{-5}$ , but one has to bear in mind that laboratory scale experiments on Cs-plate out like LAMINAR-Loop [19] led to completely different results.

Figure 7 also contains a postcalculation of Cs-plate out using parameters measured in laboratory scale experiments, which obviously do not fit to the VAMPYR-II results. This also may be taken as an indication that Cs does not occur in elemental gaseous form in the AVR. Taking that into account, there are two explanations on this deviating Cs-behavior:

- (i) Cs is already dust borne in the hot gas;
- (ii) Cs occurs as a chemical compound, which has a smaller desorption energy than elemental Cs.

Comparing VAMPYR-II experiments with and without dust filter upflow of the plate out section, however, does not lead to different results concerning Cs plate out, because Cs passes this dust filter. This is taken as additional indication that Cs is not mainly dust borne in the AVR hot gas. As also seen in Figure 8, Co-60 is retained with high efficiency already in the first dust filter in front of the plate out section. Co-60 is known to be dust borne. However, as seen from Figure 8, most of the Cs in Experiment 4 is retained in the second dust filter, which is situated downflow of the plate out section. The main difference between the 2 dust filters is the temperature, so condensation of a Cs-compound (CsOH) may be the dominating deposition mechanism. Formation of CsOH was found to be possible in AVR normal operation from kinetic and chemical thermodynamic point of view. Similar holds for future reactors because a conservation of a protecting oxide layer on metals requires a sufficient oxidation potential, which is supplied by sufficiently high  $H_2O/H_2$  ratio. The deposition behavior of those Cs-compounds under HTR conditions is not exactly known but desorption energies are expected to be much smaller than for atomic Cs. Figure 9 contains the distribution of Cs-137 onto the different components of VAMPYR experiments.

The fission product deposition in VAMPYR-II occurred mainly on metallic surfaces because a complete dust layer could not be formed within the short period of operation, and because some experiments contained a dust filter upflow of the plate out section. As will be discussed more detailed in Section 5, VAMPYR-II conditions are therefore not completely representative for pebble-bed reactors.

A similar plate out behavior of Ag as above is observed in the COMEDIE experiments [17, 20, 21]. Concerning Cs plate out, it is interesting to note that in the first COMEDIE series, a flat profile with altogether low deposition was observed too [20, 21], whereas in the second [17], a more classic (decreasing) deposition profile combined with a higher deposition degree was found. A major source for graphitic dust in COMEDIE is not seen, also because no movement of graphitic components occurred. Thus, fission product chemistry seems to be the reason for the "unusual" plate out behavior of Cs in [20, 21]. It is worth to look into differences in experimental conditions between both COMEDIE experimental series, because here, one key for understanding the Cs-behavior might be discovered.

Iodine plate out was measured on a stainless steel tube in VAMPYR-I [3, 14]. The plate out profile can be crudely approximated by a desorption energy of 170 kJ/mol.

#### 4.2. Influence of dust on fission product plate out

Plate out experiments for HTR metallic components are performed so far on metal specimen. However, there is some doubt from HTR experience, whether that represents most components of pebble-bed HTRs because of the presence of carbon-based dust. On virtually all surfaces in the coolant circuit of the THTR-300 a graphitic dust layer was present, which had a thickness of about 5-to-10  $\mu\text{m}$  (total full power operation time of THTR-300: 16 months) [22]. A visible black dust layer is present also on most AVR surfaces. Further, Cs and Sr in the coolant circuit of the Peach Bottom HTR with block-type fuel were found to be greater than 80% in carbonaceous layers, which covered almost all surfaces [12]. In the Peach Bottom HTR, the carbonaceous layers were formed almost completely by continuous small lubrication oil ingress from blowers. Examinations on the AVR blower nozzle revealed that deposited fission products can be removed by wiping to the same extent as by an electrolytic surface treatment of 1.8  $\mu\text{m}$  in metal depth [23]. This however does not hold for fast-moving parts of the blower, where shear forces are too high for dust deposition. Accordingly, the activity may be present dust-bound on the nozzle but not in other fast moving parts of the blower. Conclusions drawn from blower examination suffer however from the fact that the operating blower came in touch with liquid water for several days during a water ingress in 1978 and that the blower examinations occurred shortly after the water ingress. There may have happened some decontamination of the blower already during the water ingress.

The arguments presented before support the assumption that these fission products are not plated out on the metal (and are thus less mobile) but mainly stuck in the mobile graphitic dust layers. An additional argument lies in the very large sorption capability of matrix graphite compared with metal surfaces for Cs and Sr, as demonstrated in Figure 3. The sorption capability of graphite is already for a layer thickness of 1  $\mu\text{m}$  substantial larger than that of the underlying metal surface as parameter calculations using SPATRA revealed. These calculations were performed on plate out of elemental Cs in an HTR-Module200-like reactor, with and without a 1- $\mu\text{m}$  matrix graphite layer on all metallic surfaces. As expected, the results differ substantially in the high temperature range, where deposition on graphitic dust dominates. Whereas for metal surfaces an ad/desorption equilibrium is rapidly reached, the sorption capability of dust is much larger and thus it acts as a perfect sink. In the low temperature range, the deposition is mass transfer controlled for both, metals and graphite, that is, perfect sink behavior. Accordingly, the deposition will be proportional to the accessible geometrical surface of graphite and metal. The results are qualitatively similar for desorption energies from LAMINAR-loop and from Experiment 4 of VAMPYR-II. Because of the pronounced chemisorption enthalpy, associated with sorption of Cs to graphite, and in line with chemical equilibrium calculations [11], it has to be assumed that fission product chemistry does not change this result in general although the deposition kinetics may be different.



TABLE 4: Dose rates on a gas turbine [5].

Nuclide	Dose rate [mSv/(GBq·h)] in a distance of 1 m from a gas turbine
Cs-137	0.10
Ag-110 m	0.45

The assumption of predominant fission product sorption on graphitic dust in pebble-bed reactors changes, as outlined before, the deposition profile in a gas circuit, but has also major safety implications. Because dust is easier to remobilize than activities plated out on metals, the source terms in a fast depressurization with its substantial shear forces will increase remarkably. A detailed discussion of the dust behavior in AVR and of its safety implications is presented in Section 5 of this paper.

### 4.3. Plate out limits for a gas turbine

Plate out on metals/metal oxides remains relevant for HTRs with block-type fuel and also for fast-rotating components of pebble-bed reactors, particularly for a gas turbine. During the required hand on maintenance of a gas turbine, the annual dose limit of a radiation worker of 20 mSv has to be met, which limits the contamination of the turbine during maintenance, for example, for Cs-137 to about  $10^{-6}$  of a core inventory (400 MW<sub>th</sub> reactor), as the dose rates of Table 4 indicate.

Consideration of AVR core release rates for the final stage of operation with modern fuel at 950°C (see Figure 4), corrected with a factor of 5 for the known underestimation of core release rates by VAMPYR-I and assuming a plate out of 10% of the core release on the gas turbine results leads to contamination values much higher than tolerable. Thus, satisfaction of dose limits becomes a challenging item and requires further examinations. Enhanced AVR core temperatures have to be discussed in this context too (see Section 6.2) [6].

## 5. AVR EXPERIENCE ON DUST AND DUST-BORNE ACTIVITY

Dust played a major role in AVR fission product transport and will be relevant for safety examinations of future pebble-bed reactors too. This is due to the mobility of dust and due to its large specific activities.

### 5.1. Dust: Characterization and formation mechanisms

Maximum specific activities in AVR dust were 100 GBq<sub>Cs-137</sub>/kg, 30 GBq<sub>Ag-110m</sub>/kg and 400 GBq<sub>Sr-90</sub>/kg. These activities, which are not necessarily typical for future reactors, were measured in 1986 on dust sampled from the steam generator [24]. In 1976/1977 specific dust activities of even up to 2200 GBq<sub>Sr-90</sub>/kg (700 ppm) were measured in the cold-gas filter. Part of the activity is present already in dust during formation from fuel elements, but the major

part comes from sorption of molecular activities by the dust. The latter is concluded from the observation that specific activities of dust sampled from fuel element surfaces and from the fuel handling system are smaller than maximum specific activities given above. In the past, it was assumed that only about 10 to 15% of Cs and Sr are dust borne in AVR [24]. However this value is based on the assumption that the deposition on the steam generator occurs as plate out on metals and not on an overlying graphite layer. As already discussed before, most surfaces of pebble-bed HTRs are covered with carbon layers, which predominantly absorb metallic fission products.

Analyses of dust sampled in filters revealed a metal content of 5 to 15%, the main part is graphite. The metal and metal oxide fraction was particularly high until 1975. Co-60 is formed by activation of Co-containing alloys and of Co-impurities in carbon components, and distributed dust borne in the whole circuit. Typical specific activities of Co-60 in AVR dust were 1 to 5 GBq/kg, its stationary gas-borne activity was about 10 Bq/m<sup>3</sup><sub>STP</sub>. Because that hinders maintenance, Co-alloys should be avoided in HTRs as much as possible.

Dust formation in AVR by abrasion occurred mainly in the active core and in the fuel element handling system. Abrasion from reflectors plays some role too. Neutron irradiation may enhance abrasive dust production. Besides by abrasion, a large amount of dust was produced in AVR 1982 by a unintentional oil ingress (0.12 m<sup>3</sup>) and subsequent cracking of the oil, producing up to about 70 to 75 kg of loose carbon dust and of carbonaceous layers on surfaces (see AVR Quarterly Progress Report III/1983 in [15]). Some soot-like dust was observed, but the consistence of the solid cracking product depends on the cracking temperature and on the nature of the solid substrate where the carbonaceous layer is formed on. In 1971 a unintentional feeding of 100 m<sup>3</sup> of air into the primary circuit induced a peeling of outer fuel element layers, which probably increased the dust level by about 8 kg. The detailed mechanism of this peeling effect is however not understood. The total dust amount is not exactly known, but assumed to about 100 to 200 kg. A former published value of 60 kg [24] had to be increased after inspections during dismantling and after consideration of the oil ingress. Dust produced by abrasion is roughly estimated to 50 to 80 kg, corresponding to a dust production rate of 5 kg/y<sub>operation</sub>. Neglecting reflector abrasion, which cannot yet be quantified, this is equivalent to a fuel element weight loss of 0.2%. There were several out-of-pile experiments on dust production rates, which were performed in air. These experiments are not sufficiently representative for reactor conditions because friction coefficients of graphites are strongly reduced by a chemisorbed oxygen layer, which is not present under reactor conditions.

Size analyses of AVR dust performed at the end of AVR life revealed an average number weighted diameter of 0.6 μm only, independent on the origin of dust (see Figure 10). This was originally not expected for dust formed by abrasion, because typical grain sizes of fuel element matrix and reflector graphites are much larger. However, experience on interactions of dust particles with a turbine indicated that

large dust particles are crushed. The crushing probability increases strongly with the particle diameter. Some crushing may have taken place in the AVR blowers too. Dust found in filters consists partly of agglomerates. Agglomeration may have taken place in filters and/or already gas circuit; a separation of agglomerates into original particles was possible by ultrasonic treatment.

It has to be noted that preliminary measurements performed during the early AVR operation led to dust sizes in the range of  $5\ \mu\text{m}$ . These measurements were however performed before of the oil ingress 1982. It cannot be excluded that the dominating presence of dust less than or equal to  $1\ \mu\text{m}$  is connected to the oil ingress. Visual inspections of dust just after the oil ingress support this assumption. Only one large graphite particle was found in an AVR dust filter: its weight was 130 mg. Large metal particles were more frequently observed in these filters. A remarkable amount of coarse-grained dust was found during AVR dismantling on the bottom reflector and in cracks of it. The latter was however not examined in detail. Based on the early diameter measurements, dust filters in AVR were mainly designed for average particle sizes of greater than  $2\ \mu\text{m}$ . For that, an incomplete filtration of dust may have taken place in some dust filters, but not in the cold-gas filter (Experiment 2 in Figure 1).

### 5.2. Results of dust filter experiments

Besides dust sampling from VAMPYR-I and VAMPYR-II and from cold-gas experiment for stationary operation, there are results from 15 stationary and 3 transient campaigns with the dust experiment 1984–1988. Condensable fission products in the cold gas were found mainly dust borne, particularly for transient operation.

The most surprising results came from dust experiments with blower transients with online detection of some relevant nuclides. The blower was operated 2 days after reactor shut down at about  $1500\ \text{min}^{-1}$  until stationary conditions were achieved. Then, the blower frequency was increased within less than 1 minute to  $3000\ \text{min}^{-1}$ . Already for these frequencies, which are smaller than in normal operation ( $3300$  to  $3600\ \text{min}^{-1}$ ), the dust concentration in the coolant increased by a factor of about 50 to 100 [7, 25]. Acceleration from  $1500$  to  $4000\ \text{min}^{-1}$  increased the dust concentration even by a factor of 200 to 400. Obviously already small perturbations in the flow pattern may lead to a significant dust remobilization. Mobilization at low shear forces was observed also on dust layers out of pile [26]. Vibrations of components during transients may have played a role too. The transient dust concentrations in the dust filter experiment are shown in Figure 11 indicating that dust redeposition occurs with time constants of 1 to 2 hours.

In line with this slow dust deposition is the postexamination result that no particle in these dust filter experiments with a size greater than  $1.5\ \mu\text{m}$  was found after ultrasonic treatment. Provided that dust agglomeration happened mainly in the dust filter, this result might support the hypothesis that no dust from dead water areas is remobilized (where mainly large particles are expected to be sedimented),

but dust adhesively bound on surfaces. Dust is perhaps deposited on surfaces in multilayers where adhesion forces decrease with increasing layer thickness; the maximum layer thickness is in that case given by the shear forces of stationary operation.

Looking at the specific activities of dust in transient experiments, it becomes obvious that I-131 activities are with up to  $3.5\ \text{GBq/kg}$  relatively high, having in mind that the total I-131 activity outside the active core is in the range of  $50\ \text{GBq}$  [24]. Probably, the dust which is remobilized contains some more short-lived nuclides than the remainder and there is not much iodine plated out on metal surfaces (as already discussed before for Cs), respectively, sorbed on graphite components. The specific activities of long-lived nuclides as Cs-137 in the dust, mobilized during transients, was by a factor of up to 4 larger than in comparable stationary experiments and almost of the same order as in 1976, when specific activities were highest. Here, we have to assume that old dust with larger specific activities is mobilized in larger amounts too. The same conclusion has to be drawn from dust sampling by the cold-gas filter under stationary conditions. Sr-90 shows for all cold-gas experiments the highest sampled activity, even for experiments at low hot gas temperatures, when Sr-90 release is expected to be low. For the time period 1986–1988, the sampled Sr-90 activity in the cold-gas experiment is by about a factor of 4 to 8 higher than the Cs-137 one and by about a factor of 20 to 200 higher than the Ag-110 m one. This may be explained as follows: dust is settled and mobilized several times during operation until a stable position is found. For the same reason, a correlation between sampled Sr-90 activity in dust and hot gas temperatures is not always found. Precise Sr-90 measurements are however not available for all runs of the cold-gas experiment. Summing up, dust filters integrate particularly for Sr-90 to some extent the prior release history of AVR. Not only fresh dust is sampled, but also dust which is already present for a long time in the coolant circuit.

The total stationary dust concentration in the coolant, as measured by the dust filter of the cold-gas experiment, is shown in Figure 12, indicating its low values. The dust concentrations do not show a dependence on coolant temperatures. Their large scatter reflects variations of the flow pattern in the coolant circuit and of dust formation rates during sampling. Dust concentrations measured in the cold-gas experiment are a factor of 2 to 3 smaller than in the dust filter experiment and by a factor of 3 to 4 smaller than in the dust filter of VAMPYR-I. This is due to differences in dust deposition along the pipes to the filters.

A reliable model for dust behavior in HTRs including interaction with fission products does not yet exist. For that, there are no model-based evaluations of AVR dust-related data.

### 5.3. Relevance of dust for source terms in accidents of pebble-bed reactors

In modern HTRs, friction forces in the active core are about an order of magnitude larger than in AVR (inverse He flow, greater core height), so dust production in the active core will

increase. On the other hand, future reactors will be supplied with a dust filter in the fuel handling system. Conservative design values for dust production rates in modern small pebble-bed HTRs (400 MW<sub>th</sub>) are estimated to about 50 to 100 kg/y. Detailed knowledge of the dust amount is not as relevant for analyses on dust-borne activities as on dust explosions. The sorption capability of dust for fission products is even for the lower uncertainty limit of dust production rates sufficient for absorbing the activities in normal operation. On the other hand, for dust explosions exists a concentration limit, which is 70 g/m<sup>3</sup> for 4 μm graphite particles in ambient air. The maximum overpressure of dust explosions is 6.5 bar, as experiments on related dust problems in fusion reactors revealed [27]. Dust explosions are mainly relevant for depressurizations into a building containing air. Here, dust sedimentation and lift-off and their influence on the probability of a local inflammable mixture is a problem, not present in solely gaseous mixtures. The possibility of incineration or even explosion of dust in filters, affecting their retention ability, should be examined. Matrix graphite dust shows a higher reactivity in air at low temperatures [28], and thus will have a lower ignition limit than the referenced 4 μm dusts.

Altogether, despite of the small dust remobilization in the transient AVR dust experiments the state of knowledge indicates that for fast depressurizations with shear forces substantially higher than in normal operation and/or very different flow pattern, a remarkable mobilization of the dust-bound activity has to be considered excluded [17, 26], at least not within the conservative treatment of design basis accidents (DBAs). Unfortunately, there are no additional representative experiments on mobilization of dust as in pebble-bed reactors available. A depressurization experiment of the AVR primary circuit with measurement of the mobilized activity was planned for the last operation year but was not commissioned. An experimental facility for lift-off experiments on AVR specimen is at present in Juelich under construction. However, it is not clear to which extent AVR specimen representative for future pebble-bed reactors are still available. Representative specimen should consist of dust formed by abrasion, but the incidental oil ingress of 1982 has probably changed most AVR surfaces by oil cracking and has also changed the average dust morphology. For that, specimens from THTR might be more suitable. The lift-off behavior of dust formed by abrasion is expected to be different from that of carbonaceous layers formed by oil cracking. There is some experience on lift-off of carbonaceous layers formed by unintentional oil ingress in the Peach Bottom HTR [29]. Results of these lift-off experiments of 2-minute duration indicate that lift-off of Cs and Sr remains less than or equal to 1% for shear forces up to those present in normal operation of the reactor. Depending on temperature history and on substrate composition of the specimen, a lift-off of 2 to 25% was measured for shear forces of a factor 5 higher than in normal operation. Even larger shear forces are expected for fast depressurizations.

Plate out activities on metals are much less mobilized in depressurizations than dust-borne ones as lift-off experiments indicate [17]. Past safety analyses on pebble-bed

reactors assumed most activities in the coolant circuit to be plated out on metals and only a minor fraction dust bound. Accordingly, source terms in depressurizations were underestimated and the former conclusion that filtration is superfluous due to low activity content of the released gas is no longer valid. Thus, a carefully filtered release of fast depressurizations is essential for pebble-bed HTRs. This is particularly true for arrangements of several HTRs on one site, as is foreseen, because the accident frequencies per site increase proportionally to the number of reactors there. Such filtration is state of the art for nuclear systems equipped with a gas-tight containment. However, it is not yet clear whether a sufficient fast and efficient filtration is achievable for the vented, not pressure-retaining confinements of present pebble-bed HTR concepts. Summing up the efficient filtration of dust with its small particle diameter remains a challenging problem.

## 6. OTHER SOURCE TERM RELEVANT AVR EXPERIENCE

### 6.1. *Noble gas measurement and related quality control of fuel*

The noble gas measurement is not directly related to the problems addressed here, because noble gases are not deposited and do not create major safety problems. Their tolerable release in DBA following German regulations is up to 8 orders of magnitude larger than for key nuclides as Cs, Sr, and I. However, the noble gas level was a direct indicator for the number of defective-coated particles in the AVR core and its increase during operation. Accordingly, this experiment allows a direct determination of the quality of the fuel elements present in the reactor concerning coated particle failure; but in contrast to original expectations, easily detectable noble gas release cannot be correlated with the release of metallic fission products by diffusion: a late detected pronounced release of metallic fission products leading to a heavy contamination of the coolant circuit was a major problem of AVR operation.

This item has implications to the quality control of HTR fuel elements too: at present, the fuel manufacturing parameters guaranteeing reasonable activity retention are known, but the related physical fuel parameters are not sufficiently examined. Thus, monitoring of fuel manufacture is the main quality-ensuring measure, but quality control after manufacture is difficult. One reliable, but elaborate proof after manufacture is irradiation of fuel specimen under conditions simulating normal operation, and subsequent heatup to accident temperatures. This deficit in fuel quality control abilities was thought to be compensable in AVR by a fast detection of wrong fuel in operation by an enhanced noble gas release. However, as AVR experience teaches, this does not hold for releases of metallic fission products by diffusion. For that, additional effort in the fuel quality field is required. Because the fuel element acts as the central element of the HTR barrier concept, the presence of fuel giving no fast warning by a high noble gas level in normal operation but failing in core heatup accidents has to be excluded at least for DBA.

During AVR operation, the total noble gas inventory in the coolant varied from 200–6000 GBq [9, 24], depending on operational temperatures and was particularly high during partial failure of a charge of BISO fuel elements with large fuel content (GLE-1), which reached very high temperatures. A high noble gas level was also observed even at lower temperatures after the water ingress of 1978 with its increased steam impurity level. This was due to the interaction between steam and defective particles. Concerning safety, noble gas precursors require some attention, particularly for Sr-90, but also for Cs-137.

### 6.2. Enhanced AVR core temperatures

There is no way of online or local measurement of pebbled (active core) temperatures in contrast to other reactors. This is, because the moving pebble bed destroys all equipments. Average hot gas temperatures outside of the active core were occasionally measured online in AVR at the same positions as VAMPYR-I and VAMPYR-II as discussed in Section 4.2. Talking about temperatures in the active core, we have to distinguish between coated particle temperatures, fuel element surface temperatures, and gas temperatures. Depending on its power, the coated particle temperature of a fuel element positioned near the hot gas exit was in AVR up to 120 K higher than the surface temperature. Gas temperatures in this position were calculated to about  $30 \pm 10$  K lower than fuel element surface temperatures. In this paper, core temperature means fuel element surface temperature. Maximum (best estimate) core temperatures in AVR were calculated to 1070°C for average hot gas temperatures of 950°C [30], assuming a core composition as in 1974. In later operation with different core compositions, higher maximum core temperatures of up to 1140°C were calculated for small fuel-element fractions (less than 1% at temperatures greater than 1100°C) and limited operation time.

Some effort was spent in measuring maximum AVR core temperatures, which was however not completely successful before 1988: In September 1986, 190 graphite pebbles equipped with melt wires (melting points between 650 and 1280°C) were fed onto the core top. For most of the time, when the monitor pebbles were calculated to pass the hot core top, the AVR average hot gas temperature was at 950°C. These monitor pebbles record the maximum gas temperature occurring around the monitor pebble, plus some minor contribution of about 6 to 10 K by  $\gamma$  and neutron heating. Accordingly, the corresponding core temperature is about 20 K higher than the recorded maximum temperature. The results for 144 monitor pebbles, extracted until end of AVR operation, are as follows [31–33]: except of about 1/3 of the pebbles fed into the radial inner core zone, all pebbles revealed core higher temperatures than the originally calculated maximum temperature of about 1100°C, 1/3 of the pebbles fed into the radial outer zone even by more than 200 K, that is, all melt wires were molten. Whereas almost 90% of the fuel elements fed onto the inner core zone were evaluated, this holds for only about 55% of the monitor pebbles fed onto the outer core zone. The unevaluated

monitor pebbles belong probably to the outmost part of the outer core zone, where pebble flow velocities are expected to be lowest. Accordingly, a complete figure of maximum temperatures in AVR is not yet available. Figure 4 reveals that Cs-release was comparatively low in the last quarter of 1986, when monitor pebbles passed the core top. The same holds for noble gas release [32]. Accordingly, the impermissible high temperatures occurred probably during most of the AVR operation.

The unintentional high core temperatures transgressed safety limits. Accordingly, the permit for an operation at 950°C hot gas temperature was withdrawn and hot gas temperatures were limited to 810°C for the rest of its operation time, that is, from February 1988, when first temperature results became available [6, 32]. Thus, a repetition of temperature measurements at 950°C hot gas temperature in order to determine the real maximum core temperature was not permitted.

The reasons for the high AVR core temperatures are not yet understood, several AVR specific but also other reasons immanent to pebble beds are in discussion (gas bypass flow, power peaks near to reflector noses, pebble bed stochastics and locally densified pebble bed, error in axial power distribution, radial asymmetric power profile, etc.) [34]. At present, some theoretical effort is spent in order to clarify this problem.

The unintentional high temperatures are relevant also for examinations of source terms: as discussed in Section 2, a pronounced increase in release rates of metallic fission products (Sr, Cs, Ag) was observed after the hot gas temperature increase from 850 to 950°C in February, 1974. As outlined in [6], this enhanced release has to be mainly attributed to the unintentional high AVR temperatures and not, as assumed in the past, to insufficient fuel quality only. Other safety-relevant implications of the unintentional high core temperatures are discussed in [6] too.

### 6.3. Implications of water ingress on fission product behavior

A slow water ingress of 27500 kg in 1978 led to a substantial transfer of activity (mainly Sr and H-3 to a minor extent Cs and I) from primary circuit components into the water [35]. Some of the water went unintentionally into the grounding of AVR, where it is still present. Traces of Sr-90 were detected in the soil surrounding the reactor [36]. The exact size of this Sr-90 contamination is not yet known but will be examined after dismantling of the AVR.

There was also some graphite oxidation during this accident and in the subsequent drying phase. However, when large amounts of water had entered the primary circuit, core temperatures were substantially lower than in normal operation. So the maximum oxidation degree observed on fuel elements depending on the type was only 0.2–0.5%. After water ingress, some standard oxidation tests (1 kPa steam in He, 1000°C) were performed on fuel elements: A substantial acceleration of oxidation rates was observed for some fuel elements by catalytic effects. On the one hand side metallic corrosion products formed by the water ingress

were absorbed by the fuel elements and induced pitting corrosion. On the other hand, the large Sr-concentration in the fuel element graphite increased oxidation rates in steam homogeneously over the fuel element surface by up to a factor of 5 by catalysis.

It remains to be examined, whether local high hot gas temperatures as observed [6, 13] may have initiated the AVR steam generator leak.

#### 6.4. Experiments on coolant chemistry

There were continuously experiments with impurity injections into the AVR coolant circuit ( $\text{CO}_2$ ,  $\text{H}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{N}_2$ ,  $\text{CH}_4$ ,  $\text{NH}_3$ ) [32, 37]. Interesting for understanding of the H-3 behavior are hydrogen injection experiments, which led—by exchange reactions with H-3 sorbed on graphite—to a large emission of H-3 into the gas phase. Based also on such experiments, it was concluded that nitrogen and hydrogen are present gas borne in HTRs only to about 0.1%, but mainly sorbed on graphite.

#### 6.5. Pebble rupture in operation

Rupture of fuel element pebbles in operation leads to destruction of coated particles and to the presence of actinides, particularly Pu-241, in dust. Similar was observed in the THTR, where a far more pronounced pebble rupture than estimated (8000 pebbles in 16 months full power operation) [22] was observed. Whereas the majority of the pebble rupture in THTR may be attributed to control rod insertion, some may also be due to the pebble movement. Although the total actinide activities in AVR are small (some ten GBq of Pu-241 in AVR dust), they cannot be neglected because of their pronounced toxicity due to their  $\alpha$ -activity and because of the mobility of dust. Altogether, only 220 of 2.4 Mio cycled pebbles ruptured in AVR [32]. There exists, however, no reliable model for pebble rupture up to now, as also the THTR experience indicates [22]. Extrapolation to modern facilities has to consider the same arguments as discussed before for dust formation, that is, a higher rupture rate than in AVR has to be taken into account.

### 7. CONCLUDING REMARKS

Major discrepancies between in-pile and out-of-pile results of plate out experiments indicate that common laboratory scale experiments on fission product deposition of HTRs should be handled with care. They did not completely simulate the real deposition behavior in a reactor, because of a different coolant composition (impurities like CO,  $\text{CO}_2$ , steam, etc., and other fission or activation nuclides) and because of the presence of fission product absorbing dust also on surfaces preferentially in a pebble-bed reactor. Also, for the same reason, a sufficient understanding of the deposition degree  $\alpha$  in physical or chemical terms is not yet available. Plate out on a gas turbine is a challenging problem due to the required hand-on maintenance. As consequence of their safety relevance, these problems require detailed additional examinations.

The data situation concerning dust formation, interaction with fission products, and dust behavior in the coolant circuit of HTRs and in depressurization is still insufficient. The same holds for respective modeling tools. Accordingly, major research and development effort on dust is required. Oil ingress events as a dust source need additional attention. As long as uncertainties are unresolved, the need for mitigation of dust-related problems advises a pebble-bed reactor design equipped with a gas tight containment and measures against dust explosions. A gas tight containment may diminish many of the problems associated with enhanced normal operation temperatures as observed in AVR and with pebble rupture, as problems due to the insufficient fuel element quality control, but not those associated with maintenance and dismantling.

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