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AVR prototype pebble bed reactor: a safety re-evaluation of its operation and consequences for future reactors

The AVR pebble bed reactor (46 MWth) was operated 1967–1988 at coolant outlet temperatures up to 990 °C, which are suitable for process heat applications. Also because of a lack of other experience the AVR operation is a basis for future HTRs. This paper deals with insufficiently published but unresolved safety problems of AVR and draws conclusions for future pebble bed HTRs: Although the AVR was operated only for about 4 years at coolant temperatures >900 °C its primary circuit is heavily contaminated with dust bound metallic fission products (^{90}Sr, ^{137}Cs), which create major problems in current dismantling. The end of life contamination reached several percent of a single core inventory. The AVR contamination was mainly caused by inadmissible high core temperatures, and not – as presumed in the past – by inadequate fuel quality only. The high core temperatures, which cannot be easily measured in pebble beds, are probably caused by the insufficiently examined pebble bed mechanics. Safety of future reactors requires a gas tight containment and in addition elaborate R&D or reduction of requirements concerning temperatures and burn-up.

Der Prototyp-Kugelhaufenreaktor AVR: Eine sicherheitstechnische Neubewertung seines Betriebes und Schlussfolgerungen für zukünftige Reaktoren. *Der Kugelhaufenreaktor AVR (46 MWth) wurde von 1967–1988 bei bis zu Prozesswärme-tauglichen Kühlgastemperaturen von 990 °C betrieben. Der AVR ist Vorbild für zukünftige Kugelhaufenreaktoren, da es keine anderen hinreichend belastbaren Erfahrungen gibt. Die vorliegende Arbeit befasst sich mit einigen unzureichend publizierten, aber sicherheitsrelevanten Problemen des AVR-Betriebs und zieht Schlussfolgerungen für zukünftige Reaktoren. Obwohl der AVR nur 4 Jahre bei Kühlgastemperaturen ≥900 °C betrieben wurde ist der AVR Kühlkreislauf massiv mit staubgebundenen metallischen Spaltprodukten (^{90}Sr, ^{137}Cs) kontaminiert, welche den gegenwärtigen Rückbau erheblich erschweren. Diese Kontaminationen betragen einige Prozent eines Coreinventars. Sie wurden in erster Linie durch unzulässig hohe Coretemperaturen verursacht und nicht, wie früher angenommen, ausschließlich durch unzureichende Brennelemente. Die hohen, im Kugelhaufen nur schwierig messbaren Coretemperaturen liegen vermutlich in der nur unzureichend bekannten Kugelhaufenmechanik begründet. Die Sicherheit zukünftiger Reaktoren erfordert neben einem gasdichten Containment entweder umfangreiche F + E-Arbeiten oder Reduktion der Anforderungen bei Nutztemperaturen und Abbrand.*

1 Introduction

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 and the test of many different types of pebble shaped fuel elements. Design data of the AVR [1, 2] are collected in Table 1. A sketch of the reactor and of some relevant components is presented in Fig. 1.

An unintentional coolant flow bypassing the steam generator caused restrictions of AVR power. Diverse fuel types with different coatings were used in AVR (see [2], p. 313). Most fuel pebbles contained 1 g ^{235}U, but the total heavy metal content (U, Th) varied from 6 to 11 g. After a residence time of 4–50 (average 6–8) months in the core the fuel elements reach the defueling tube at the core bottom and are re-fed to the core top (MEDUL-core). This is repeated until the final burn-up is met and then the fuel element is replaced by a

Table 1. Main design data of AVR pebble bed reactor

Power/power density	46 MW$_{th}$ (15 MW$_{el}$)/2.5 MW/m^3
Cycle	Steam cycle with the steam generator (73 bar) inside the reactor vessel
Core height/ diameter	2.8 m/3 m
Coolant/Pressure	He/10.8 bar
He outlet/inlet temperature	1st phase until 02/1974: ≤ 850 °C; 2nd phase until 02/1988: ≤ 950 (990) °C; 3rd phase* until 12/1988: < 750 °C/275 °C
He-flow	13–15.5 kg/s (depending on desired gas outlet temperature) in up flow direction
Fuel	Core: 100 000 matrix graphite pebbles (6 cm diam.) containing coated fuel particles. Diverse fuel types, at end mainly improved TRISO fuel

* after detection of inadmissible high core temperatures the permission for operation at high coolant temperatures became invalid

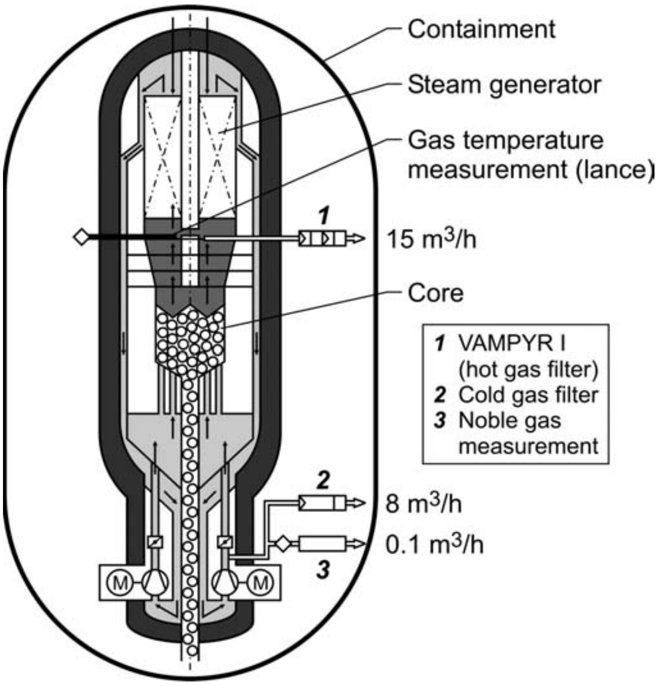

Fig. 1. Scheme of the AVR

fresh one. In order to achieve a radial more even temperature profile low power fuel elements are fed into the core centre and fresh ones into the outer regions (2-zones core). Pebble flow velocities were found to be much higher in the inner than in the outer core region. The continuously moving active core is a unique feature of pebble bed reactors (PBR) which however requires very good knowledge about pebble bed mechanics and stochastics. Helium cools the core in up flow direction. A steam generator is arranged at the top of the pressure vessel.

Although several improvements are foreseen for future pebble bed reactors AVR is in many respect still their paragon, also because the larger demonstration reactor THTR300 (750 MW_{th}, 300 MW_{el}) suffered from diverse partly pebble bed specific problems and was operated thus only for 1.2 fpy (full power years). R&D on pebble bed reactors was limited since 1990 and thus its experience is still of major relevance. Because hot gas temperatures of up to almost 1000 °C were achieved in AVR, which allow for process heat applications, the pebble bed technology finds major interest worldwide. This paper focuses on some unresolved and insufficiently published safety relevant problems which occurred during AVR operation, and outlines consequences for future pebble reactors. The widely published advantageous features [1, 8, 12, 40] of pebble bed HTRs are out of the scope of this paper. More detailed studies to these AVR/HTR problems are found in [4].

At present AVR undergoes dismantling, which became complicated due to a pronounced contamination of the primary circuit with metallic fission products ^{90}Sr and ^{137}Cs, and with the activation product ^{60}Co. The contamination by metallic fission products, which is in the range of some percent of a core inventory, is power weighted by more than 5 orders of magnitude larger than in current German PWRs during operation ($4 \cdot 10^{-3}$ GBq ^{137}Cs/MWth, see chapter 3.6). In PWRs most of this activity is dissolved in coolant water. This water is treated separately before of disposal and thus there remain almost no metallic fission products in the reactor vessel. In AVR particularly highly radiotoxic ^{90}Sr creates safety concern: This β-contamination was found to be the highest of all nuclear installations worldwide, except of those with severe accidents, and to be in the most problematic status for dismantling and safety, i.e. as mobile dust. The reactor vessel will be grouted with light concrete which immobilizes dust and stabilizes the vessel, and will be stored for some decades outside of the AVR site, until a procedure for final dismantling is developed. Grouting is a stabilization method successfully applied to Russian atomic submarines, taken out of service. Storing outside of the AVR site is required, because the soil around AVR became contaminated with ^{90}Sr too (see chapter 3.1) and has to be decontaminated. This requires the removal of the reactor.

A re-evaluation of the heavy primary circuit contamination is performed here. Besides the standard explanation that an insufficient fuel quality was primarily responsible for the pronounced fission product release [1a, 31], also other reasons like overheating of fuel elements, as detected at the end of AVR operation, will be examined. Further, other safety relevant experience from AVR operation will be discussed and conclusions for future reactors will be drawn.

2 AVR core temperatures

2.1 Measurement technique and results

In this paper, the term core temperature means fuel element surface temperature. There is no way of a contemporary measurement of active core temperatures in pebble bed HTRs, in contrast to other reactors, because the pebble movement destroys standard detection equipments. Hot gas temperature profiles outside of the active core were measured occasionally in AVR [3], but core temperatures were based on calculations. For average AVR hot gas temperatures of 950 °C maximum core temperatures were originally calculated to values of 1070 °C to 1123 °C [4]. In the inner core maximum temperatures were about 75 K lower. Depending on the fission power of the fuel element, maximum coated particle temperatures are up to 150 K higher than core temperatures. Modern fuel elements were designed for a maximum coated particle temperature in normal operation of < 1250 °C. However sufficient irradiation tests were until recently available only for lower temperatures or low burn-up [5] and a design temperature of 1250 °C seems to be too optimistic (see chapter 3.3). Fig. 2 shows a scheme with relations between the different temperatures.

A crude method for measurement of maximum core temperatures in pebble bed HTRs was developed already 1970 to 1972 [4]. An improved technique was applied from September, 1986 [1b, 2]: 190 graphite pebbles equipped with 20 melt wires (melting points from 650 to 1280 °C) were fed onto the core top. The monitor pebbles detected the maximum gas temperature a pebble passes through during its flow through the core, plus some contribution by γ and neutron heating of 8 ± 2 K [4]. Maximum core temperatures are about 22 ± 12 K higher than maximum temperatures measured by monitor pebbles [4]. This method gives almost no results on spatial or time dependent temperature distributions. Further this temperature measurement is based on the assumption that maximum temperatures are present only at the core top: A continuous operation at high temperatures was possible in AVR only for a limited time but not for the whole passing period of fuel elements through the core. Potential hot spots below the core top, if any, could not be detected because already about 2 months after insertion of monitor pebbles the overall reactor temperature was lowered. Temperature measurement by monitor pebbles is also a time consuming process: First alarming results were obtained not earlier than 15 months after measurement start. Further, the whole measurement campaign was never finished because of the final AVR shut down 1 year later when about 25 % of the monitor pebbles persisted still in the core and thus were not examined. The temperature measurement is even within the discussed restrictions valid only for the last quarter of 1986 but not necessarily completely representative for other operational periods of AVR.

Table 2. Maximum gas temperatures in the AVR active core during normal operation (IV/1986)

Range of maximum temperature [°C]	Percentage of monitor pebbles inner core [%]	outer core
920–1072	35 (25 > 1050 °C)	0
1073–1280	58	69
> 1280	7	31

The temperature results are summarized in Table 2. References to more detailed data on temperatures and insertion of monitor pebbles are found in [4]. The not evaluated, delayed 46 pebbles belong mainly to the outer core region, which may be explained by the much smaller pebble flow speed adjacent to the wall. Except of about 1/3 of the pebbles fed into the radial inner core zone, all pebbles revealed higher temperatures than the maximum of 1070 °C, originally calculated for licensing of hot gas temperatures of 950 °C [4], 1/3 of the pebbles fed into the radial outer zone even by more than 200 K. Gaseous [1a] and metallic (see Fig. 4) fission product release was normal or even comparatively small during the temperature measurement campaign. This indicates that the temperature elevations did not occur during the campaign only, but in whole AVR operation. The absolute maximum temperature was not measured, because it was higher than the highest melting point of the melt wires used. True maximum core temperatures in the inner respectively outer core are coarsely assessed to 1320–1340 °C respectively 1380–1420 °C. Certain fuel elements with higher power (GLE-1) used earlier will have reached even higher temperatures. Maximum fuel temperatures (see Fig. 2) transgressed the permissible limit of 1250 °C defined in the operation license by far more than 200 K.

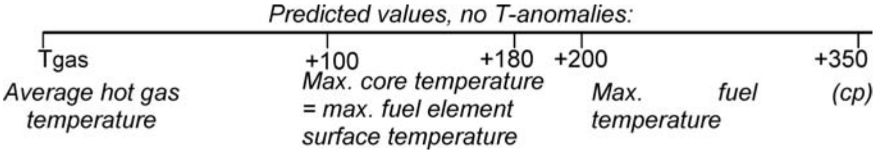

Fig. 2. Relation between hot gas temperature, max. core temperature and max. fuel temperature

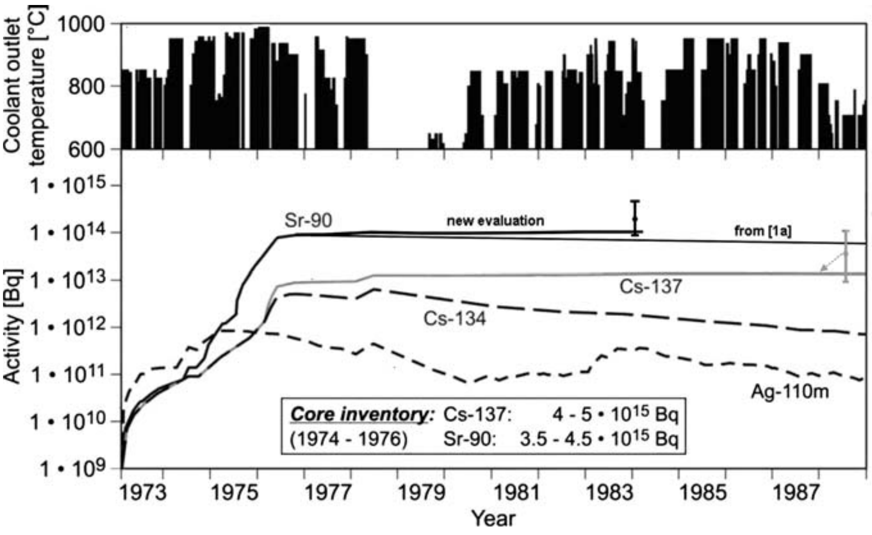

Fig. 3. Time dependent average hot gas temperatures (above) and accumulated activity release into the AVR primary circuit estimated from VAMPYR-I hot gas filter results

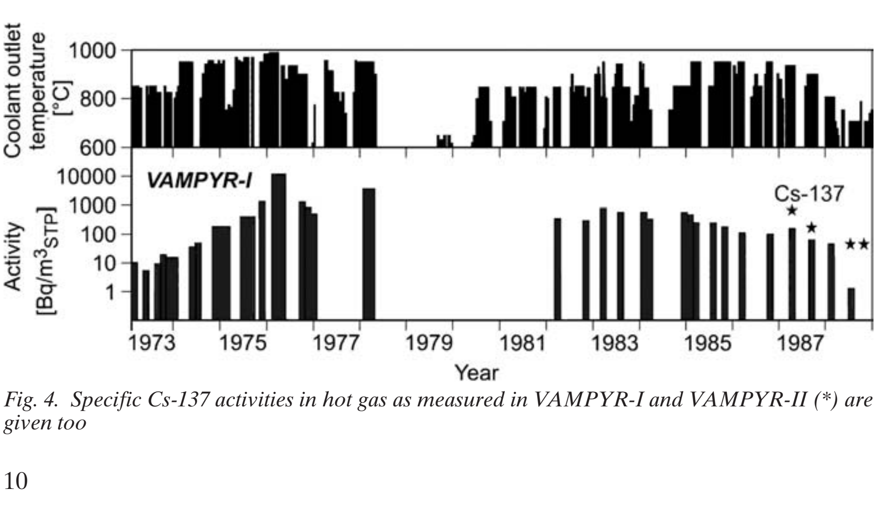

Fig. 4. Specific Cs-137 activities in hot gas as measured in VAMPYR-I and VAMPYR-II (*) are given too

When first temperature results became available the permit for AVR operation at 950 °C hot gas temperature became invalid for safety reasons and the hot gas temperature had to be strongly reduced from beginning of 1988 ([2], p. 190). This temperature drop is seen in Fig. 3. Accordingly, the AVR should no longer be taken as reference for a safe and reliable reactor operation at gas temperatures allowing process heat applications, as done e. g. in [12]. Ironically, the pebble bed HTR concept has probably survived until now mainly as consequence of one of its weak points, its insufficient in-core instrumentation abilities: In case of known AVR core temperatures from beginning of its operation, the AVR hot gas temperatures would have been limited to values far below 950 °C. This means that its main advantage, its apparent capability for process heat generation, would not have been demonstrated.

2.2 Interpretations of inadmissible high temperatures

The reasons for the high AVR core temperatures could not be clarified up to now [4, 9, 13, 43]. It is not even completely clear, whether there were hot spots in the core or even large regions of high temperatures. However, the pronounced fission product release discussed in chapter 3 suggests a substantial fraction of fuel elements with inadmissible high temperatures. Several AVR specific but also pebble bed intrinsic and general reasons and their combinations are in discussion (coolant bypass flows inside and/or outside of the active core, power peaks near to AVR reflector noses, uncertainties in pebble bed stochastics, locally densified pebble bed with high flow resistance, human errors in fuelling procedure, uncertainties in pebble flow behaviour, uncertain burn-up measurements, power asymmetry in the core [4, 13, 43], cones of pebbles on the core top, flow anomalies by the broken AVR bottom reflector [18] etc.). There are indications from the distribution of core and hot gas temperatures that not the whole core is affected by the inadmissible high temperatures. Measured radial hot gas temperature profiles in the hot gas chamber (see Fig. 1, lance) show time dependent differences of more than 100 K for the same average hot gas temperature and the same position. Maximum hot gas temperatures of even more than 1100 °C were observed (see chapter 4.2). Further on a continuous slow change of these hot gas temperature profiles was detected for unchanged overall reactor conditions. This proceeded with time constants typical for changes in pebble bed arrangements. That points to pebble be inherent reasons. Several examinations on AVR temperatures were performed up to now, but only for AVR specific and general, however not for pebble bed inherent reasons. Such pebble bed inherent reasons however, if acting, might create virtually insurmountable problems. In [4] uncertainties in pebble flow mechanics are discussed in correlation to the inadmissible high temperatures more detailed.

Pebble bed densifications afford major attention in this context [4, 43]: A statistically generated pebble bed as assumed in HTRs has a void fraction of about 0.4. Minimum void fractions in pebble beds are however as low as 0.26. Regions with low void fractions show substantially higher temperatures because of their strongly enlarged pressure drop, i.e. reduced cooling, and because of an enlarged power density. A temperature increase of several hundred K is estimated for densified regions of some thousand pebbles. During AVR construction it became obvious that pebble bed movement leads to pebble bed densifications, because the densest pebble bed represents the equilibrium state. In order to avoid densification indentations were worked into the side reflector surface. Experiments on beds of small metal pebbles (room temperature, air) showed that this measure prevents from major pebble bed densifications [1c, 42]. It remains however unclear, whether the latter result is representative for real HTR pebble beds with their large friction and high temperatures: It has to be shown that HTR pebble bed densifications can be excluded also in some distance from the reflector surface. A remarkable increase of pebble bed densities was observed also in THTR300, whose reasons are controversially discussed. Local densifications of packed beds are a common problem in chemical engineering [39]. Clustering of low burn-up, high power fuel elements in the core have to be considered as reason of high temperature regions, too. A general disadvantage of PBRs in comparison with other reactors lies also in the fact that position and local density of fissiles in the core, which are important input parameters for neutronics calculations, are not a priori known, but have to be estimated via insufficiently validated pebble bed mechanics codes. The complex problem of pebble bed mechanics became recently subject of analytical studies using high performance computers under simplifying assumptions [39], leading to the conclusion that this item contains fundamental unsolved problems.

The pebble bed mechanics problems are partly related to an error made at the beginning of pebble bed reactor design and unhealed up to now: Based on Bragg, who investigated the graphite crystal in 1928 and explained the lubrication properties of graphite by its layer like structure, it was assumed that the benign lubrication features of graphite under ambient conditions are an intrinsic property of this material. A smooth pebble flow in PBRs requires low friction. However, as was already discovered in 1948 but not sufficiently noticed by AVR designers, lubrication behaviour and low friction of graphite occur only in presence of sufficient moisture, or – less efficient – of oxygen [38]. There are several reasons under discussion for this effect [38]. Under inert conditions as in HTRs the friction coefficient is by a factor of up to ten larger and the wear rate increases by up to more than 4 orders of magnitude. This leads to the "dusting wear" phenomenon i.e. the formation of large dust amounts by friction and wear [38, 41]. Further, under HTR conditions the friction coefficient of graphite shows strong temperature dependence [1c]. Unfortunately all experimental studies on graphite pebble beds up to now were performed under non representative low friction conditions. The strong underestimation of graphite friction became obvious after some years of AVR operation because of large amounts of graphite dust found, because of pebble movement problems in the fuel reshuffling facilities and because of a pronounced discrepancy between precalculated and observed residence time spectra of fuel elements in the core. The AVR-experience to problems in pebble bed mechanics gained 1967–72 [42] did – in contrast to expectations [42] – not result in a satisfying operation of the THTR300: Its operation 1985–89 strongly suffered from pebble bed mechanics problems, as described below in this chapter. The large friction between graphite and in-core shut down rods of the THTR300 led even to the destruction of a large number of fuel elements. This indicates that pebble bed mechanics is still an unresolved problem.

Core external bypass flows were recently suggested to be the main reason of the inadmissible high temperatures [33]. Some minor contribution by external bypass flows is indeed probable: A core bypass was foreseen by design in order to cool the metallic shut down rods positioned in side reflector buttresses (see Fig. 1 in [4]), whose temperatures must not exceed 700 °C. This bypass flow still indirectly cools the core, but less than in direct core pass. This essential bypass was erroneously omitted in several AVR operational and safety calculations, which thus underestimated maximum core temperatures by about 40–70 K. Accordingly the total operational coolant flow, which was set on basis of these calculations due to the lack of in-core instrumentation, was too low by 5 to 10 %. A major contribution to high temperatures by core external bypasses however can be ruled out: Besides the above discussed fluctuations in hot gas temperature profiles for the same average hot gas temperature, which point to pebble bed inherent reasons, the following facts have to be considered: In case of an external bypass the core cooling is reduced which means that the radial core temperature profile is homogeneously increased. However, the comparison of precalculated and observed radial distributions of temperatures indicates much larger temperature differences, probably due to spacious hot spots. This is also in line with the observed very large scatter of fission product release from the same type of fuel elements, which can be explained by pebble bed inherent hot spots. Altogether the data situation in the AVR core is very poor due to the lack of in-core instrumentation. For that and because of complicated pebble bed mechanics and fuelling there are major doubts, whether pebble bed intrinsic reasons for high temperatures can be excluded. Probably, only a representative large scale experiment or an experimental pebble bed reactor can convincingly answer the remaining questions.

Experience of the THTR300 pebble bed reactor also gives hints to problems with enhanced temperatures and with pebble bed mechanics: Failure of insulation attachment bolts in the hot gas duct occurring in an early operation stage was probably caused by thermal fatigue due to excessive temperature gradients across the core outlet. Temperatures in the THTR core axis were much higher than predicted due to a faster pebble flow in this region, which changed the radial power distribution. One explanation is that debris of broken pebbles has strongly diminished the pebble flow velocity in the outer core zone, in addition to the effect of friction at the walls. Another explanation correlates the lower graphite friction at high temperatures with these high flow rates in the centre. By reduction of the fraction of high power fuel elements in the core axis and by restriction of the pebble flow to low power generation periods (on Sundays, when the electricity demand was smaller), these problems were mitigated. Dust production in THTR300 hindered its operation, e. g. by disturbing instruments and fittings. These problems together with others forced the final shut down of THTR300 after only 1.2 fpy. An evaluation of the THTR300 operation experience is still outstanding and is urgently required.

Reduced flow velocities of fuel elements in pebble beds, which cannot be avoided near to reflectors, may lead to inadmissible high burn-up and thus accelerated fission product release. This is particularly relevant in modern systems with downward coolant flow, having the hottest part at the bottom: By that the combination of high burn-up and high temperature, which leads to pronounced fission product release rates, becomes more probable. In an annular pebble bed core with a centre graphite column the pebble bed surface areas contacting reflectors are particularly large and accordingly, the regions with diminished pebble flow rates, too. Other disadvantages of a downward coolant flow as in modern concepts are due to the higher pressure on the pebbles by the large pressure drop of the coolant: This pressure is enhanced by the three times greater height of the pebble bed in modern reactors compared to AVR. Dust formation and pebble rupture will increase [17] and pebble flow is hindered as consequence of this enlarged pressure. Downward coolant flow avoids potential hot spots in the pebble cone on the core top but there are major flow and temperature irregularities expected in the conic bottom part of the pebble bed.

3 Re-evaluation of fission product release from AVR core into the coolant circuit

3.1 Release data

Measurements of Cs, Sr and Ag release from AVR core by a deposition tube in the hot gas region (VAMPYR-I, see Fig. 1 [16, 17, 29]) revealed that the fission product release into the primary circuit strongly accelerated 1974–1976, correlating with the hot gas temperature increase to 950 °C in February, 1974. Collapse of Sr-retention in graphite, occurring at high Sr-concentrations [29], may have accelerated the Sr-release. Time dependent accumulated release values based on measurements in VAMPYR-I are shown in Figs. 3 and 6. Specific ^{137}Cs coolant activities from VAMPYR-I are presented in Fig. 4, data of the improved plate out facility VAMPYR-II (at same height but in other radial position as VAMPYR-I, see Fig. 1) are given, too. Average hot gas temperatures are given in Figs. 3 and 4. A re-estimation of ^{90}Sr release [4] ("new evaluation" in Fig. 3) revealed that the Sr-release did not stop in 1976 as erroneously assumed in [1a] but continued on a reduced level. The time dependence of the releases in Figs. 3, 4 and 6 was probably approximately correct, although VAMPYR-I is known to underestimate releases, but in the final AVR operation more than in the initial one [4]. The absolute release values are now known to be too low by a factor of up to 10 [4], as also VAMPYR-II results indicate. This is included in the error bars of Fig. 3 and is taken into account in the recent safety report for AVR dismantling [36]. The latter assumes accumulated releases of $7.5 \cdot 10^4$ GBq for ^{137}Cs and $1.1 \cdot 10^5$ GBq for ^{90}Sr at end of operation. Considering this underestimation the core release rate R of ^{137}Cs is proportional to the coolant activity A of Fig. 4 by:

$$R\ [\mathrm{Bq/s}] = 500 \cdot A\ [\mathrm{Bq/m^3_{STP}}]$$

The release rates of Fig. 4 contain 2 rogue results with extraordinary high values. These are the releases in the first quarter 1976 and in the first quarter 1978, whereas the other values fit well into a homogeneous picture without showing a pronounced dependence on the fuel type in core. The latter holds particularly, if the higher VAMPYR-II rates are considered, too. The rate in the first quarter 1976 is influenced by an additional temperature enhancement due to an operation error, which was calculated later to about 40 K, but may have been slightly higher. During the first quarter 1978 the moisture level in the coolant circuit was high, probably because of a steam generator damage, which led a few months later to a massive water ingress (see the end of this chapter and chapter 4.2) and this may have influenced the Cs-release rates.

A large fraction of ^{90}Sr and ^{137}Cs is bound on dust: Up to 7000 ppm of ^{90}Sr were found in graphite dust. The total amount of dust in AVR is about 150–200 kg [17]. The core release rate of ^{90}Sr for the time period 1974–78 at a hot gas temperature of 950 °C was calculated 1979 to 20 GBq/y by standard diffusion models/data without consideration of enhanced temperatures [4], which is orders of magnitude smaller than observed. Unfortunately, the enhanced release of metallic fission products was detected with several months of delay only: This was, because a fast release measurement of metallic fission products does not exist. In addition – in contrast to expectations – no conspicuous release of easily detectable noble gases by fuel failure was observed. Further, first calculations of the total primary contamination on basis of VAMPYR-I data were erroneous [4]. Accordingly, during AVR operation the contamination was not taken as serious as it was. In rating the releases of metallic fission products it has to be noted that AVR-operation at hot gas temperatures > 900 °C, when most of the releases occurred, sums up to only about 4 years.

Graphite dust of the AVR primary circuit contained also small but radiotoxic relevant quantities of actinides (^{241}Pu, ^{241}Am), mainly caused by pebble rupture and destruction of coated particles. Because of larger compressive loads in advanced HTRs [16, 17] this problem has to be examined more detailed. Here, unexploited THTR experience should be considered, too [10]: The large fuel element rupture in THTR can be partly attributed to rod movement in the core, but other compressive loads may have contributed, too [17].

Besides amplification of AVR dismantling costs another consequence of this contamination was discovered 1999: In course of a slow accidental steam/water ingress of about $3 \cdot 10^4$ kg in 1978 [2], about 1500 GBq of the accumulated ^{90}Sr was washed out, together with 10^5 GBq of ^{3}H. Some of the water leaked incidentally into the reactor grounding and reached the surrounding soil [4, 18]. Because the steam generator leak remained small, this accident was not a design basis accident: Core temperatures were already low when large water amounts were present and thus the extent of the graphite/steam reaction remained limited.

3.2 Influence of unintentional temperatures on fission product release

Assuming that maximum temperatures measured in 1986 are roughly representative for AVR operation at 950 °C hot gas temperature it becomes clear that the enhanced fission product release is correlated to the observed overheating of fuel to temperatures far higher than the maximum permissible value: Hot gas temperatures of 800 to 850 °C (1973) led to release rates by 2–4 orders of magnitude smaller than for 950 °C (1975–76), see Fig. 4. Results from US-Peach Bottom HTR with well known core temperatures (operated 1967–74 with block type fuel, representing a similar fuel development stage in its core 2 as AVR 1970–78). The carbide BISO fuel particles of GK fuel elements in AVR, which were found to be the main source of Sr, were about the same fuel type as that used in Peach Bottom core 2: Particularly the Sr release in Peach Bottom core 2 was by several orders of magnitude smaller than in AVR [7, 22], but also Cs-release was by greater than one order of magnitude lower. Increase in particle defects in AVR operation remained small and cannot explain the majority of the release [4].

3.3 Release mechanisms of metallic fission products

We have to distinguish 3 sources of fission products: Intact coated particles, particles with defective coatings and the uranium contamination of the graphite resulting from manufacturing. Progress of fuel element technology (e. g. SiC coating) diminished the fraction of uranium in defective particles and uranium contamination during AVR operation from 10^{-2} to $< 10^{-4}$. Noble gases are virtually completely retained by intact coatings, i. e. the fuel element acts as an efficient barrier and a partial release occurs from defective particles and uranium contamination only. Metallic fission products however diffuse through intact coatings which become penetrable at high temperatures [23, 37]. For that the fuel element is a sufficient barrier for metallic fission products only up to a certain temperature limit, which depends on the mobility of the metal and on the irradiation time. For temperatures below these limits the release of Cs and Ag is caused by the small level of uranium contamination and defective coated particles.

Diffusion through barriers is characterized by 2 parameters: The breakthrough time tB reflects the initial diffusion phase before steady state diffusion is reached and characterizes the retention time of a barrier:

$$t_B = l^2/(6\,D)$$

with l = coating thickness, D = diffusion coefficient of fission product in coating. Steady state diffusion rates in coatings are proportional to D/l. Because of the temperature dependence of diffusion in solids the breakthrough time decreases and the stationary diffusion rate increases with increasing temperature. Besides temperatures there are other parameters accelerating diffusion rates in HTR fuel, as burn-up and neutron fluence. However, the physical mechanisms are not yet fully understood.

Table 3 contains the breakthrough time at 1250 °C (maximum permissible coated particle temperature) for different barrier components of HTR fuel pebbles. Buffer layer and LTI-PyC are not considered, because their retention capability is small compared to other barriers. The data scatter of about one order of magnitude reflects uncertainties of diffusion coefficients. Altogether, not only the temperature is a relevant parameter for release estimates but also the temperature dose (temperature · time). Diffusion calculations indicate that in oxide fuel the Sr-retention is additionally reduced by Sr-interaction with the kernel, which has to be considered in addition to the data of Table 3. As shown in [35] this retention is however diminished in normal operation by recoil. For carbide fuel kernels the graphite is in any case the dominant Sr retention barrier. Table 3 indicates that for TRISO-fuel breakthrough is possible at 1250 °C for all nuclides during fuel irradiation time (2 to 4 y, about 1/3 at high temperatures), but the release rate remains small for Sr due to the mentioned retention by the oxide kernel. For HTI-BISO fuel no Cs-breakthrough has to be expected, but Ag-release and in case of carbide kernels also Sr-release is larger compared to TRISO oxide fuel. Cs-retention in intact TRISO coated particles is worse compared to former HTI-BISO fuel. A similar conclusion for Cs is drawn in [37]. On the other hand, modern TRISO fuel strongly reduces the uranium contamination of graphite and thus the release rates of iodine and noble gases in normal operation. TRISO fuel represented 30 y ago, when it was developed, a still acceptable solution, although already at that time the limited Cs-retention ability of intact SiC layers created some concern [23, 37]. However it does not fulfill todays requirements. Altogether, modern TRISO fuel represents a compromise and may cause greater problems concerning Cs-release at high temperatures [4, 5, 37].

A recent irradiation (EU1bis) of 5 modern German GLE-4 fuel elements at maximum pebble central temperatures of 1250 °C [24] discovered a pronounced release of gaseous and metallic fission products: GLE-4 LEU fuel elements representing the highest quality of the German HTR program were present in AVR final operation. Goal of the experiment was to demonstrate the applicability of present HTR fuel for Very High Temperature Reactors (VHTR). Full power irradiation time was about 8.5 months, the burn-up reached by error only 11.1 % fima (^{235}U/Utotal = 0.16) instead of the foreseen 16 % fima. Nevertheless the fraction of defective particles increased by a factor of > 10 more than expected and also a “significant” release of Cs and Ag out of the fuel elements was observed. ^{133}Xe release was far larger compared to other noble gases [24] which may be due to an untypical high release of its precursor ^{133}I. A detailed postexamination is ongoing. A later heat-up of one of these pebbles to 1600 °C revealed a high instantaneous ^{137}Cs release, which is due to the inventory released in the irradiation phase out of the coated particles into the graphite. A release of Cs out of intact coated particles, which starts at intermediate burn-up and ex-

Table 3. Breakthrough time t_B [d] for metallic fission products at 1250 °C in components of HTR fuel pebbles

Nuclide	Breakthrough time t_B [d] in		
	HTI-PyC (85 µm, BISO)	SiC (35 µm, TRISO)	A3 matrix graphite (5 mm)
^{90}Sr	1–10	10–100	≤ 200
^{110m}Ag	1–10	10–100	< 0.5
^{137}Cs	1000–10 000	50–500	1

ceeds the level of defective coated particles by an order of magnitude at final burn-up, is also in-line with release calculations using diffusion codes as discussed in chapter 3.5. An ongoing similar experiment with GLE-4 fuel elements at about 150 K lower temperatures does not show up to now higher noble gas releases than expected. These results underline that even modern HTR fuel is not yet suitable for VHTR applications, and that the maximum design temperature of fuel particles of 1250 °C for current TRISO fuel is too optimistic and should be reduced by about 100–150 K.

3.4 Interpretation of AVR release rates after hot gas temperature increase to 950 °C

Looking on Figs. 3, 4 and 6 it becomes obvious, that the temperature increase starting February, 1974 leads with a delay of about 0.5 to 1.5 y to release rates of Sr and Cs by about 3 orders of magnitude larger than before. The ^{110m}Ag release became even almost complete soon after temperature increase and thus its increase is smaller. The delayed start of the release is mainly due to diffusion effects (see chapter 3.3), but a slowly increasing particle failure fraction (GLE-1, see below) has contributed for Cs, too. An AVR fuel element undergoes temperature cycling down to temperatures of about 300 °C during its flow through the core which enlarges the diffusion break through times.

^{90}Sr -release was dominated by diffusion out of high enriched UC_2-kernels of GK BISO fuel elements: Only one GK-fuel element (of about 25 examined) without significant release of Eu (as indicator for Sr) was found after 1975. The contribution of the uranium contamination of the matrix graphite and of defective particles to enhanced Sr-releases remained small, because in BISO-UC_2-fuel the graphite is the dominating barrier (Table 3), which is acting also for matrix contamination and defective particles. Further, fission products generated from uranium contamination are predominantly located in the interior of crystallites and not on pore surfaces: This leads to an additional retention, which was also observed in AVR. From 1980 dominance of oxide fuel and the lower temperatures led to smaller release rates from core, which are however still on a safety relevant level and will be discussed later.

For Cs-release the situation is more complex, also because of the pronounced retention capability of HTI-BISO coatings. The influence of a small charge of 2400 wrong designed fuel elements (GLE-1) on Cs-release in AVR is discussed in [4]: A re-estimation discovered that defect particles of GLE-1 fuel elements were not predominantly responsible for the ^{137}Cs release [4, 31] in contrast to earlier assumptions [1a]. However the average Cs release rate of GLE-1 was significantly larger than for other fuel elements. Considering all uncertainties the contribution of GLE-1 to the overall Cs-release 1974–78 is assumed to be in the range of 10 to 30 %. There are major contributions from fuel elements with smaller particle defect fractions and from uranium contamination of the matrix graphite, but particularly from diffusion out of intact coated particles for all types of fuel elements. In line with this interpretation even HTI-BISO GK fuel elements with high Cs-release out of intact particles of up to 25 % were found. The latter may be taken as an indication of very high temperatures and/or excessive burn-up.

For future reactors it is relevant that obviously no credit can be taken from the originally assumed strong correlation between the coating failure rate, which is easily detectable by noble gas release, and the release rate of metallic fission products (see also chapter 3.1).

3.5 Dependence of activity release from AVR core on the core composition

For future reactors it is also important to know the behaviour of modern TRISO fuel. For that, we compare fission product releases from core for the period 1974–1978, when predominantly carbide BISO fuel was in the core, with release rates from 1983–1988, when oxide fuel with an increasing content of TRISO particles was used. Irradiation tests of modern fuel elements, usually applied for the proof of a satisfying behaviour in normal operation, are not available for the range of maximum AVR temperatures.

^{90}Sr release rates dropped between 1978 and 1984: ^{154}Eu release rates (standing for ^{90}Sr) decreased by about a factor of 20 in AVR after change to oxide fuel. This still significant rate is in line with accelerated irradiation tests of oxide fuel elements at temperatures > 1250 °C: The FRJ2-K3 irradiation experiment at surface temperatures of 1400 °C resulted in a release of even almost 10 % of ^{90}Sr already within 37 days [11]. At 1300 °C there was only minor release out of the fuel element in 37 days, but substantial release from particles into graphite occurred. Irradiation experiments at 1250 °C surface temperature but 104 d of irradiation (FRJ2-K9/B3) came to similar results as FRJ2-K3 (1300 °C). The fuel particle temperature was high in FRJ2-K3, which partly explains the large release rate at 1400 °C. In addition, recoil increases the release from kernel [35]. Although these fuel elements belong to an early development stage they are still representative for Sr: The main Sr-retaining components, oxide kernel and matrix graphite are already present. Results on cold gas and dust measurements of Sr/Eu in AVR, which fit into these interpretations, are discussed in [4].

^{90}Sr release shows a pronounced temperature dependence in oxide fuel, but a much smaller one in carbide. Measured diffusion coefficients of Sr in UO_2 and UC_2 are presented in Fig. 5: Comparing the higher UO_2 diffusion coefficient with that in UC_2 we find, that both are of the same order at 1500 °C, whereas at 1200 °C the diffusion in UO_2 is by a factor of about 500 slower. Taking recoil into account, the difference at low temperatures becomes however smaller. Taking these values a reduction factor of 20 ± 10 for modern oxide fuel is in line with maximum AVR core temperatures of 1300–1400 °C, which correspond to somewhat higher coated particle temperatures. Considering that in carbide fuel the matrix graphite determines the Sr-release rate over a wide range of temperature the behaviour of oxide and carbide fuel converges even more.

Whereas ^{90}Sr is mainly retained in the oxide kernel and in graphite but less in coatings, the principal diffusion barrier for ^{137}Cs is the particle coating (SiC in TRISO or even more BISO-HTI pyrocarbon). For ^{137}Cs a reduction factor of about 8 was coarsely estimated by AVR assuming an operation only with improved and modern fuel [1a]. This was calculated on basis of the release behaviour of ^{137}Cs and of ^{134}Cs depending on the AVR core composition and temperature as presented in Fig. 6.

Fig. 6 contains the VAMPYR-I-data of both Cs-isotopes for the operation time 1973–1988 in a linear scale. Particularly the declination of ^{134}Cs ($t_{1/2} = 2$ y) reveals that during use of improved fuel the Cs-release rate in the AVR dropped between 1976 and 1987 by almost 1 order of magnitude, but remained in a still significant order. We have to bear in mind that a negligible release rate corresponds to a declination of the accumulated activity, as observed in the shut down period of 1978/79. In the period of predominant use of improved fuel in AVR (from 1983) the average temperature was altogether lower than in the period 1973–1978 (see Figs. 3 and 4). As outlined in chapter 3.3 the release rate does not depend only on actual temperatures, but on the whole temperature history the fuel element has seen, respectively on the temperature dose (i. e. temperature · time). Estimating the reduction factor of 8 the latter was not considered, which means that the true reduction is expected to be smaller. The accumulated release from 1981 to 1988 amounts to about 30 to 35 % of that in the phase 1973 to 1978. For periods when GLE-1 fuel elements discussed in chapter 3.4 do not contribute to release rates, an even larger Cs-release of improved TRISO fuel may occur, as comparison of Cs-release rates of 1973 (BISO-fuel) with those of 1987 for similar temperatures indicates (see Fig. 4).

The following observations underline that Cs-diffusion out of intact coated particles in AVR was still high for improved and modern TRISO-fuel: At first the evaluation of VAMPYR-II, whose Cs-release/birth ratio at hot gas temperatures of 930 °C (1987) was about 10^{-3}, i. e. significantly larger than the fissile fraction outside of intact coated particles (see Fig. 4). Further, postexamination of 12 modern GLE-3 fuel elements (TRISO, 1 g ^{235}U, 10 g total heavy metal) irradiated in AVR from mid 1982 revealed for 2 low burn-up fuel elements (< 3 % fima) remarkable Cs diffusion out of coated particles. Finally the recent experimental results on GLE-4 [24] described in chapter 3.3 indicate that the retention capability of modern fuel was overestimated for high temperatures in the past. As discussed in [4] the evaluation of ^{137}Cs/^{134}Cs profiles in fuel elements are difficult to interpret but may indicate a significant Cs-source below the core top by a hot spot.

For ^{110m}Ag, which is one crucial isotope for gas turbine contamination/maintenance, a by 1 order of magnitude better retention is expected in TRISO fuel, which is however compensated by the larger ^{110m}Ag inventory in modern low enriched fuel. ^{110m}Ag release rates are generally higher than those of other metals. Large ^{110m}Ag inventories in dust on the steam generator were detected 1986 [1a]. Their comparison with those for ^{90}Sr, which are only about a factor of 15 larger, indicate that ^{110m}Ag values of Fig. 3 underestimate the release by about 1 order of magnitude.

As discussed in [4] these interpretations are in line with short term heat-up tests of irradiated modern fuel elements to temperatures of 1600–1800 °C: The observed retention of metallic fission products at these temperatures holds – in contrast to more optimistic claims – only for a limited period of time. A complete release of ^{137}Cs is expected at 1600 °C after 2 to 5 months, depending on conditions of the irradiation before [4]. Extrapolation of heat-up data to lower temperatures is consistent with the t_B values of Table 3.

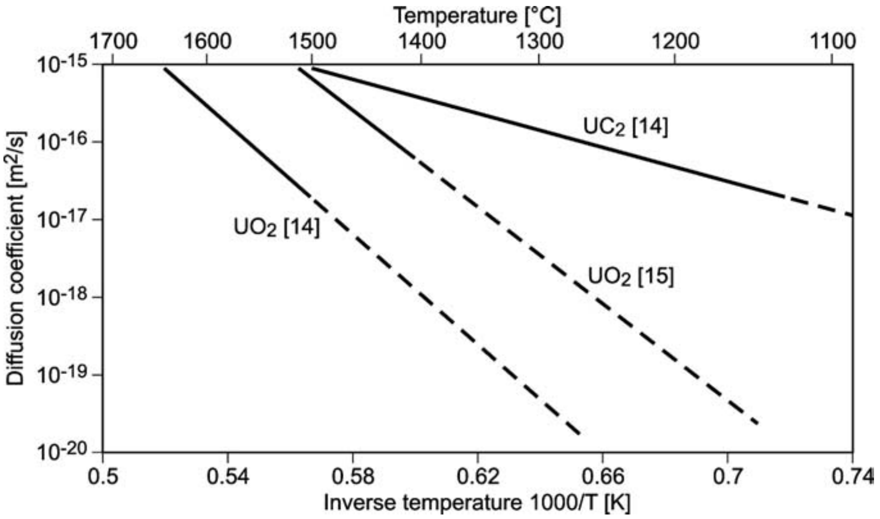

Fig. 5. Measured Sr-diffusion coefficients in UC_2 and in UO_2

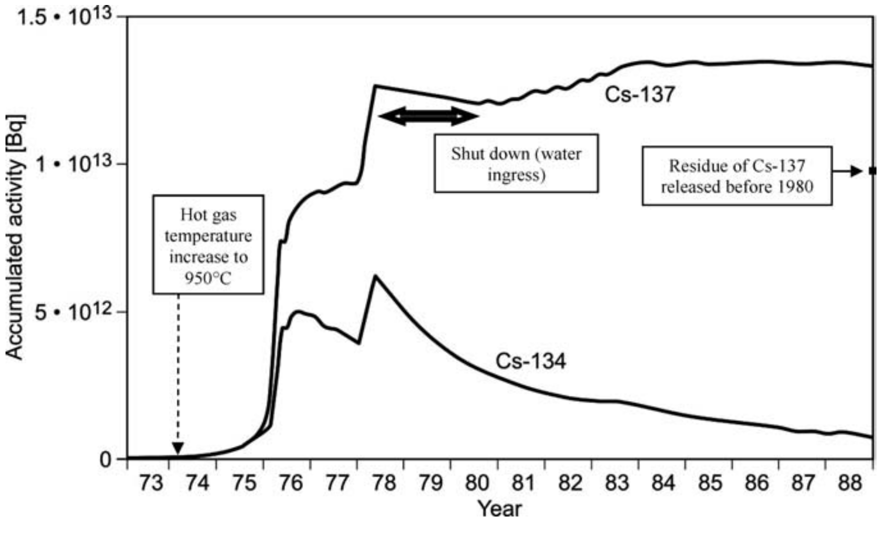

Fig. 6. Accumulated Cs in AVR primary circuit as calculated from VAMPYR-I results [1a]

A recent publication on AVR fuel [31] comes to similar results concerning the relative contribution of modern fuel elements to the release of ^{90}Sr in AVR. However, for ^{137}Cs publication [31] concludes – in contrast to the results of this paper – that modern fuel elements did virtually not contribute to the observed release. This discrepancy respectively matching is due to the fact that diffusion out of intact coated particles was completely neglected in [31] for Cs, but not for Sr. The omission of Cs-diffusion in intact coated particles is not in line with the state of science and technology [21, 23, 35, 37]. For that and because of an overestination of partile failures in old fuel the Cs release for modern fuel is heavily underestimated in [31] and conclusions drawn for future reactors on basis of these results lead to a far too optimistic figure for ^{137}Cs.

Fig. 7 contains calculations with a complete diffusion model (FRESCO-II [32]) for the Cs-release behaviour of a modern fuel element pebble during 11 cycles in reactor normal operation. The maximum fuel temperature is assumed only to 1200 °C, i. e. 50 °C below the maximum design value. The total irradiation time of 2.5 y is representative for future HTR concepts. Diffusion data are taken from [21].

It becomes obvious from Fig. 7 that after 1.3 y of irradiation the release from intact coated particles becomes the dominant Cs-release mechanism, i. e. the release from intact particles exceeds the inventory in defective particles plus the U-contamination. There is some Cs-retention in the graphite, but at end of life about $1.5 \cdot 10^{-4}$ of the inventory is released from a fuel element. Extrapolation to 32 fpy leads for a 400 MWth facility to an accumulated release of 200000 GBq of ^{137}Cs, i.e. more than found in AVR. Similar calculations for maximum fuel temperatures of 1100 °C and 1250 °C lead to accumulated releases of 25000 GBq respectively 10^6 GBq. This is also in-line with results for HTTR normal operation, obtained with two different diffusion codes and Cs-diffusion parameters [35]. For maximum fuel temperatures greater than 1100 to 1150 °C the Cs release from intact coated particles becomes in modern fuel elements dominant in course of operation. Because maximum fuel temperatures in AVR were > 1400 °C, probably up to 1600 °C, neglecting of Cs-diffusion in intact particles as in [31] leads to major underestimations of release rates particularly for modern fuel. Ongoing parameter calculations for modern fuel elements in AVR with FRESCO-II support this statement.

3.6 *Comparison of HTR and LWR fission product behaviour*

Compared to primary circuit contaminations in PWRs (about $4 \cdot 10^{-3}$ GBq/MWth, see also below) the accumulated ^{137}Cs activities in pebble bed HTRs are many orders of magnitude larger. This holds even for low temperature conditions as in the HTR-Module200 concept (hot gas temperature 700 °C, max. fuel temperature < 1000 °C), where still 1200 GBq of ^{137}Cs were expected as accumulated activity by release mainly from defective particles. Also the coolant circuit contamination with most other nuclides as ^{90}Sr and ^{131}I is in HTRs with modern fuel significantly larger than in current PWRs. This does however not hold for radiological less relevant noble gases. Considering these numbers the often claimed superior fission product retention capability of HTR TRISO fuel element pebbles becomes questionable. A superior fuel behaviour holds only for certain accidents, particularly short term core heat-up, but not for long term normal operation or air ingress accidents. The reasons for the unfavourable contamination situation in HTRs are as follows:

Fuel centre temperatures may reach in LWRs values of up to 2500 °C. However on the pellet surface, temperatures are < 800 °C, which significantly limits diffusive release. The second barrier in LWRs, the Zircalloy cladding is at temperatures of 320–600 °C only, which suppresses any diffusive release of metallic fission products via intact claddings. In contrast a diffusion of metallic fission products from intact coated fuel particles cannot be avoided in HTR normal operation because of the higher temperatures and because of the small thickness of the diffusion barriers. The latter is a coated particle immanent disadvantage. Also for these reasons the occasionally claimed compensation of a gas tight containment by the allegedly excellent retention of coated particles in HTRs [40] is far too optimistic. This is underlined by the fact that protecting coatings of fuel particles do not withstand air at temperatures > 1400 °C [19].

Another important difference involves fission product behaviour in the coolant circuit. Because of their strong affinity to liquid water (chemical interaction and physical dissolution), fission products except of noble gases remain in PWRs in the coolant, until they are removed by the purification facilities. Typical values of ^{137}Cs are only 1–5 GBq in the coolant of a PWR (700 MWth, high burn-up) and 500–1000 GBq in the resins of the purification facility [26]. This is in line with German experience for 3000 MWth PWR, where 3–30 GBq are measured in the coolant water. This allows an easy handling of fission products released in normal operation. In contrast the affinity of reactive fission products to the coolant He in HTRs is small and thus these fission products tend to plate out rapidly on primary circuit components, even in the hot regions, and on graphitic dust, as occurred in AVR [1a, 16, 17, 29]. Accordingly the removal of reactive fission products via a gas clean-up plant is not efficient and the fission products accumulate in a not well defined manner in the whole HTR primary circuit. Fission product filters for the main coolant flow were tested in an AVR bypass for up to 2 months and did not work successfully for Cs, but indicated some limited efficiency for Ag [29].

Decontamination of the HTR primary circuit was found to be difficult, also because in hot parts diffusion of fission products into primary circuit components occurs. The fission product accumulation in the primary circuit is a major unresolved disadvantage of HTRs with respect to their safety, maintenance and dismantling/disposal. As long as this HTR problem is not resolved, the requirements on fission product retention of fuel elements have to be far more stringent for HTRs, compared to LWRs. This is particularly true for pebble bed reactors with their large graphite dust content which acts as fission product carrier.

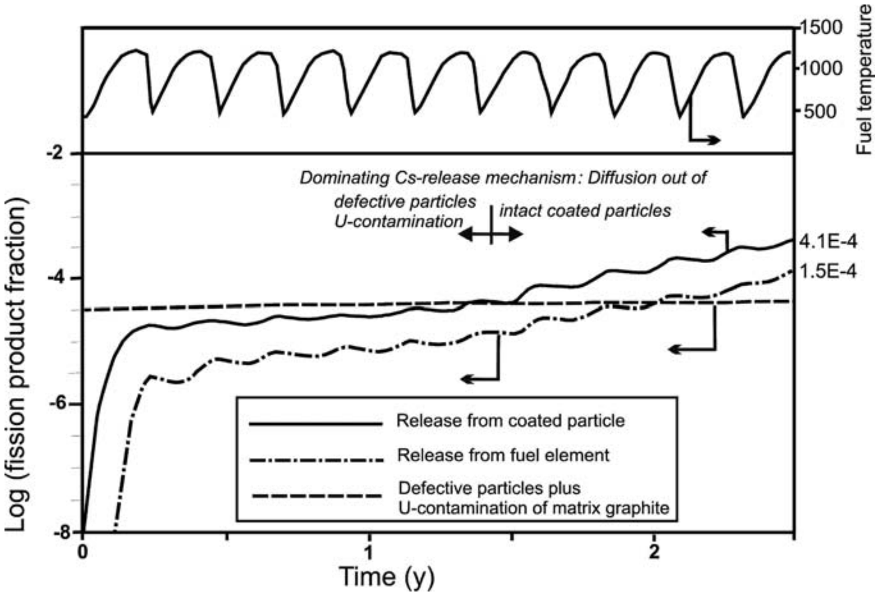

Fig. 7. Fractional release of Cs-137 from a fuel element during normal operation at fuel temperatures up to 1200 °C

4 Relevance of AVR experience for future pebble bed reactors

4.1 Maximum permissible environmental release in design basis accidents

What are the implications of AVR experience including temperature uncertainties for future pebble bed HTRs? Enhanced fission product accumulation in the primary circuit during normal operation is a major safety concern, because these activities were found by safety analyses to be main source term contribution in design basis accidents (see [16] and literature cited there), and may even contribute significantly to the risk.

Aggravating, future HTRs are currently not designed with a gas tight containment, as the AVR had. In direct cycle HTRs the activity deposition on the gas turbine, which at present cannot be sufficiently decontaminated, hinders the required hands on maintenance [6]. Further on, maximum temperatures in core heat-up accidents may be enlarged by inadmissible high normal operation temperatures [43]. In order to suppress graphite oxidation with formation of burnable gases in design basis accidents, the normal operation core temperatures in steam and process heat generating HTRs must remain limited.

Table 4 contains release limits of individual key nuclides into the environment in case of design basis accidents for German licensing conditions. ALARA holds here and therefore the releases limits must not be reached, if that is reasonably achievable.

The ^{90}Sr activity accumulated in the primary circuit outside the AVR active core (Fig. 3) exceeds German release limits into the environment for design basis accidents by more than 5 orders of magnitude. From data in Table 4 the maximum tolerable mobile fraction of fission products accumulated in the primary circuit can be estimated from the point of view of protection of the public. In case that unfiltered releases into the environment in design basis accidents have to be assumed (i.e. no gas tight containment), a conservatively estimated mobile fraction of accumulated activities must remain sufficiently below the evaluated exclusion release limits of Table 4 as is subsequently estimated:

Metallic fission products occur mainly dust borne in the coolant circuit of pebble bed reactors [16, 17, 29]. The dust is deposited by gravitation in dead water regions or by adhesive forces on surfaces. The mobilization of deposited activity in pebble bed reactors in course of accidents is not well examined, but some effort is at present spent into that item. There are however data on activity mobilization of specimen from Peach Bottom HTGR. These specimens were similar to AVR/THTR covered with a carbonaceous layer and contained more than 80 % of Cs and Sr released into the coolant circuit of the Peach Bottom HTGR. Blow down tests revealed that at shear forces by a factor of 5 larger than in normal operation between 2 and 25 % of the deposited activity is released within 2 minutes [30]. Tests on specimens without a carbonaceous layer did not show a mobilization of metallic fission products, but a small release of iodine. For conservative estimations as required in design basis accidents a mobilization degree of 25 % is assumed for depressurization accidents of pebble bed reactors. Formation of carbon dust cannot yet be avoided in current pebble bed reactors, but in HTRs with block type fuel.

Table 4. Calculated exclusion release limits of single nuclides into the environment for design basis accidents (distance to site fence: 100 m; release duration: 8 h)

Nuclide	Halve life	Inventory HTR-Module (200 MW_{th}) [GBq]	Exclusion release limit [GBq] for emission height	
			20 m	50 m
^{90}Sr	28.8 y	$1.37 \cdot 10^7$	0.4	0.6
^{110m}Ag	250 d	$1.89 \cdot 10^5$	270	410
^{131}I	8 d	$2.07 \cdot 10^8$	5.5	10
^{133}Xe	5.3 d	$4.44 \cdot 10^8$	$5.7 \cdot 10^7$	$1.1 \cdot 10^8$
^{137}Cs	30.1 y	$1.67 \cdot 10^7$	30	50

In presence of a gas tight containment with filtered release an accumulated mobile activity by at least 2 to 3 orders of magnitude larger than exclusion limits of Table 4 becomes tolerable from the point of view of public protection. However other limiting factors may become effective, as there are the above discussed requirements of maintenance and of dismantling: In order to allow hand on maintenance of a direct cycle gas turbine, the total accumulated release of ^{137}Cs in the primary circuit should not exceed about 500–1000 GBq for ^{137}Cs or 120–150 GBq for ^{110}Ag [6], in order to allow an efficient decontamination of the turbine and of other components. Restrictive limits are expected also for a complete exchange of the side reflector after 16 fpy, which is under discussion for pebble bed reactors.

The following Table 5 contains maximum tolerable accumulated activities and core release rates (32 fpy) for cases with and without presence of a gas tight containment. ALARA is taken into account: Because releases of German Generation II LWRs remain in design basis accidents far below the limits of Table 4, the exclusion release limits of Table IV are reduced for calculations of the values in Table 5 by a factor of 5. Similar data were generated by Flowers already 1973 [20], but for a large size block type HTR (3000 MWth) with about 500 m distance to the fence: Flowers calculated a maximum tolerable release fraction of ^{137}Cs into the coolant circuit of $6 \cdot 10^{-6}$ (related to a core inventory, maintenance criterion) and a maximum tolerable Sr-release fraction of 10^{-7} into the environment (public protection). This agrees reasonably with values of Table 5, having also in mind that current dose factors are larger than those used by Flowers and that the power and the distance to the fence are different in both calculations.

Typical calculated values for accumulated primary circuit contaminations (32 fpy) of advanced modular HTRs (200–400 MWth) below VHTR temperatures, i.e. at 700–850 °C, are in the range of 1000 to 20000 GBq ^{137}Cs, depending on hot gas temperatures, power and burn-up, but without consideration of major core temperature enhancements. For 400 MWth and hot gas temperatures of 900 °C the end of life Cs contaminations in the same order as in AVR at final shut down. Here we have to note again that AVR was operated for

Table 5. Maximum tolerable accumulated activities in the primary circuit of future HTRs and corresponding maximum tolerable core release rates [4]

Nuclide/ criterion	No containment: Max. tolerable		Gas tight containment: Max. tolerable	
	accumulated activity [GBq]	release rate from core [Bq/s]	accumulated activity [GBq]	release rate from core [Bq/s]
^{90}Sr/ public protection	0.4	0.5	≥ 200	≥ 250
^{137}Cs/ public protection	40	50	≥ 20000	≥ 25000
^{137}Cs/ maintenance	1000	1200	1000	1200

only less than 4 y at hot gas temperatures ≥900 °C. Detailed calculation data also for VHTR-temperatures are presented in chapter 3.5 in conjunction with Fig. 7. Corresponding ^{90}Sr-values are calculated to up to about 3 orders of magnitude smaller, which means that ^{90}Sr is a less stringent problem than ^{137}Cs for these conditions. It becomes obvious that systems without gas tight containment exceed limits for protection of the public by far, even if no temperature uncertainties are considered. For presence of a gas tight containment the limits for protection of the public however can be met. Deposition of fission products in the containment, occurring before the filtered release starts, may additionally increase the maximum tolerable activities/releases; accordingly, the values of Table 5 for the containment case represent lower limits. However criteria derived from maintenance of a gas turbine, which do not depend on the presence of a gas tight containment, are not met. Meeting of criteria derived from maintenance requires that the Cs-release fraction from a fuel element remains $<10^{-6}$, which is more than an order of magnitude smaller than the fraction of defective coated particles in modern fuel. This release fraction limit of an individual fuel element corresponds to a release fraction limit of $<10^{-5}$ after 32 fpy, related to a core inventory. Accordingly this requirement is achievable only at very low real temperatures (maximum core temperatures <950 °C, average hot gas temperature ≤700 °C). Summing up, a gas-tight containment is required also in future systems, but has to be complemented by other measures. The gas tight containment is necessary in pebble bed HTRs in order to allow a high efficiency filtration of the activity without losses by leaks. Based on past operational experience such leaks cannot be sufficiently excluded by confinement buildings without gas tight inner liner, which are foreseen in HTRs up to now. Such a confinement was in the past assumed to be sufficient because the activity release during depressurization out of the coolant circuit was underestimated. Compared with LWRs an HTR containment has to be larger, because of the larger volumes of the core and because He in contrast to steam is not condensable. Further He-tightness is more difficult to achieve than air/steam tightness as required in LWR containments. On the other hand LWR containments have to retain short lived noble gas activity, which cannot be filtered, until sufficient decay; this is not required in HTRs because of the comparatively well retention of the low radiotoxic noble gases.

Considering additionally enhanced temperatures as observed in AVR the following results are obtained: Upscaling of AVR core release rates for ^{137}Cs, as measured in the final AVR operation period 1987–1988 in VAMPYR-II [17], to a reactor with 400 MWth leads to values of >35 000 [Bq/s] at 700 °C hot gas temperature and > 300 000 [Bq/s] at 930 °C hot gas temperature. Calculation results with FRESCO-II for high fuel temperatures, outlined in chapter 3.5, underline these unacceptable high Cs release rates. ^{90}Sr with its distinctive radiotoxicity shows a more pronounced temperature dependence of release rates than Cs. Thus ^{90}Sr becomes a major problem when high hot gas temperatures are required and inadmissible high core temperatures as in AVR cannot be excluded: For maximum core temperatures >1300 °C design values of Sr-release rates exceed substantially the lower limits of Table 5 for the containment case. Altogether, the core temperature uncertainties significantly aggravate existing safety problems.

As long as pebble bed immanent reasons for inadmissible high core temperatures cannot be excluded the following strategies are possible in order to guarantee a safe operation of future pebble bed HTRs: On the one hand side a reduction of demands e.g. reduction of average hot gas temperatures and of average fuel burn-up. Recently several proposals for HTR applications at hot gas temperatures as low as 640 °C were published, e.g. in [34]. The latter paper even proposes for future nuclear high temperature applications to leave the insufficiently proven He-based HTR technology and to rely instead on the well proven British AGR technology, using CO_2 as coolant and applying conventional superheating to requested temperatures. On the other hand, major R&D effort may be spent in order to overcome the existing problems. The latter will be discussed in chapter 5. These strategies complement a gas tight containment. Summing up a safe operation of VHTR pebble bed reactors can currently not yet be guaranteed, i.e. for hot gas temperatures suitable for driving of chemical processes (950–1000 °C).

4.2 Consequences of AVR experience for water ingress accidents, air ingress accidents and core heat-up events

The formation rate of burnable gases (CO and H_2) in design basis water ingress accidents increases exponentially with temperature: This problem occurs in steam cycle and in process heat generating concepts without an intermediate heat exchanger, but not in present direct cycle ones. A primary circuit depressurization is hardly avoidable in this accident sequence. In order to prevent explosive gas mixtures after depressurization, maximum graphite surface temperatures must not exceed 1100–1200 °C [4], depending on the core temperature distribution. This corresponds to maximum tolerable hot gas temperatures of <750 °C, if maximum core temperatures of 250 K higher than calculated with standard methods i.e. as occurring in AVR are assumed. The presence of burnable graphite dust in pebble bed reactors may worsen the situation [16, 17]. An enlarged fission product release into the primary circuit followed by water ingress, remobilization of the accumulated activity, depressurization and destruction of last barriers by a gas explosion cannot be accepted as a potential scenario for a design basis accident. The prevention of these accidents may require major design changes as reduction of temperatures or an explosion proven or inertized containment.

Hot gas currents as observed in AVR may lead to overheating of parts of the steam generator or of other metallic components, which may increase their failure probability. Temperatures in hot gas currents below the steam generator of up to 1100 °C were measured in 1985 [4] but the long term stability of AVR metal components was limited to <900 °C only. The steam generator leak of 1978 may be induced by such overheating.

Another lesson to be learned from AVR water ingress of May, 1978 belongs to reactivity effects: The reactor was operated at low power and temperature (10 MWth, 500 °C) for several days in order to dry the primary circuit [2]. During that, liquid water run out of the leak in direction of the core. As exhibited later, a fraction of about 3 % of liquid water in the void volume of a pebble bed may lead to a positive void coefficient of reactivity [4, 28], which may induce reactivity excursions. These conditions were not reached in this AVR water ingress accident, also because the steam generator leak did not enlarge. In order to exclude such potential reactivity problems future concepts must not foresee steam generators on top of the core. In a later stage of the accident, when the reactor was already shut down, the blowers even touched the level of liquid water accumulated at the vessel bottom (30 000 kg) and merged water droplets to the coolant.

Beyond design air ingress accidents with subsequent core burning are treated in the safety philosophy of pebble bed HTRs as follows: There is some grace time between end of depressurization and massive damage of fuel elements by burning, ranging from a few hours to some days [19]. It was assumed in the past that no relevant activity release occurs by the depressurization preceding the air ingress, i. e. that the reactor surrounding remains accessible and thus intervention measures can easily be performed during the grace time. Considering the large and mobile AVR contamination the latter may however not be true.

Some aspects of the influence of inadmissible high core temperatures in normal operation on core heat-up accidents are discussed in [4, 43]. Diffusion of fission products into the barriers of the fuel element already during normal operation particularly for high burn-up will diminish the time span in core heat-up accidents, required for initiation of major fission product release.

5 Conclusion

The following major problems of pebble bed HTRs were identified during a re-evaluation of the safety behaviour of the AVR operation:

- Inadmissible high core temperatures and other temperature anomalies, which heavily accelerated the release of metallic fission products from fuel elements into the coolant circuit, and whose reasons are not yet understood
- An insufficient retention capability of present TRISO fuel elements for metallic fission products particularly in high temperature, long term normal operation as required for process heat applications
- Safety, maintenance and disintegration problems due to the uncontrolled accumulation of partly mobile metallic fission products all over the primary circuit
- Unexpected and unresolved behaviour in pebble bed mechanics and in pebble bed stochastics.

Major effort is needed in order to resolve the above listed open safety problems which query the licensibility of pebble bed HTRs. In any case, a gas tight containment, as included in almost all other modern reactor systems, has to be foreseen for future HTRs. The containment should be explosion proven or inertized in order to prevent from potential dust or burnable gas explosions in accidents. However, a gas tight containment will not sufficiently eliminate all problems listed: Thus pebble bed HTRs require additional safety related R&D effort and updating of safety analyses before of construction. This includes a careful examination of AVR and THTR experience. In detail the following tasks remain to be examined. This list takes also parts of the results of the NRC and CSNI evaluations of pebble bed reactors into account [4, 9, 13, 43]:

- full evaluation of operational experience and problems of AVR and THTR300
- development of a new fuel element sufficiently retaining metallic fission products in long term operation
- development of a reliable quality control for fuel elements
- experiments on iodine release from fuel elements in core heat-up accidents
- full understanding and reliable modelling of core temperature behaviour and of pebble bed mechanics including pebble rupture
- examinations on unexpected particle failures as observed in experiments with realistic core heat-up transients
- experimental and theoretical examinations on dust formation and on dust/fission product interactions under real PBR conditions
- full understanding of the pronounced contamination of the whole primary circuits of AVR and THTR300 with the activation product ^{60}Co
- proof of applicability of the second emergency shut down system, based on small absorber pebbles falling into the pebble bed (KLAK), under real reactor conditions (high friction, dust and broken pebbles, pebble bed densification)
- fast and reliable local measurement (direct or indirect) of safety relevant parameters in the pebble bed core (e. g. temperatures)
- full understanding of fission product transport in the coolant circuit, including development of measures to avoid activity accumulation in the circuit
- development of a fast detection system for metallic fission product release from core
- material development for process heat components
- HTR specific dismantling and disposal items.

Other open problems of pebble bed reactors are discussed in [4, 9, 13, 16, 17, 43]. Before initiation of the above listed comprehensive R&D, a feasibility study including an estimate of the required R&D effort is advisable in order to quantify the economical risk of this development. A representative experimental reactor, which is sufficiently equipped with instruments also for in-core measurements, is required for a reliable examination of the open questions.

Concerning beyond design basis accidents there are still unresolved questions connected to a massive air ingress with graphite burning, which may lead to massive fission product releases from the reactor [19]. However priority has to be given to solutions of safety problems related to operation and design basis accidents.

It is advisable to perform a comparative study of pebble bed reactors with block type fuel HTRs, which do not show most of the problems discussed before (no graphite dust, in-core instrumentation possible, no uncertainties as by pebble flow) and also have a smaller proliferation problem than pebble bed HTRs due to their discontinuous fuelling. More general a comparative probabilistic safety assessment considering Generation III LWRs and pebble bed reactors is required: Improved knowledge to safety of pebble bed HTRs since 1990 leads to the conclusion that former comprehensive safety assessments were too optimistic. Previously a superior safety behaviour of pebble bed reactors was claimed compared to other nuclear systems including an allegedly catastrophe free design. According to the above presented arguments there are doubts, whether this depicts reality.

The claim of a "catastrophe free" design is mainly based on the fact that PBRs show advantages in areas, where conventional reactors have problems, but does not sufficiently consider disadvantages of PBRs: A first advantage is the ceramic core of low power density, which will not melt-down in case of lack of cooling. This advantage is however achieved by a large core containing a huge amount of graphite. This design leads to a very expensive core and its high costs have to be compensated in reality by omission of some safety measures outside of the core. In addition, graphite is a burnable material: A core fire as possible in HTRs is a severe accident class not existing for conventional reactors. Also graphite disposal is an unresolved problem because of its remarkable content of C^{-14}. In this context the claimed stability of fuel elements in accidents up to 1600 °C has to be discussed, too: This stability means that short term heating (100 h) of modern fuel elements of low and medium burn-up at 1600 °C does not lead to additional failures of coated particles and that the diffusive release of fission products, except of ^{110m}Ag, remains small. This temperature limit of 1600 °C must however not be generalized: It is too optimistic for high burn-up fuel, for prolonged heat-up periods or for fast transients. In addition, fuel elements have to be treated as damaged after a real core heat-up event reaching 1600 °C and have to be exchanged. The second advantage of PBRs is their strong negative temperature coefficient of reactivity and their low excess reactivity required in operation. It was demonstrated in AVR and in the Chinese HTR-10 that in case of a stop of cooling without reactor shut down by operators the reactors were shutdown by the negative temperature coefficient in a self-acting manner for 24 h. Also this benign effect should not be generalized: A fast withdrawal of control rods in presently planned PBRs leads to a heavy reactivity insertion, which at least destroys the fuel elements, before the reactor is self-acting shut down. Further the positive void coefficient of reactivity, which may occur in steam generating PBRs in course of water ingress accidents, is a PBR disadvantage not found in conventional reactors.

Altogether PBRs contain certain inherent safety features compared to conventional reactors, which are however at least compensated by PBR inherent safety problems. Some of these PBR inherent safety problems can be solved by adequate safety measures or by R&D, but it remains uncertain whether this is possible in an economic manner.

6 Remark

The views given in this paper with regard to the relevance for future pebble bed reactors are those of the author and do not represent a common opinion of the Institute for Energy Research of Forschungszentrum Juelich. The paper is approved for publication according to the regulations of the Forschungszentrum Jülich.

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