

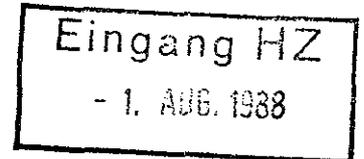
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27 July 1988

G Pott  
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Dear Mr Pott

**REF: HOT CELLS PAPER BY GRAVENOR, HITCHCOCK AND ROBERTS**

There is an omission in the caption of Figure 5. Could you please arrange for the enclosed corrected figure to be distributed with the paper.

Our apologies for the inconvenience caused.

Yours sincerely

J G Gravenor (Dr)

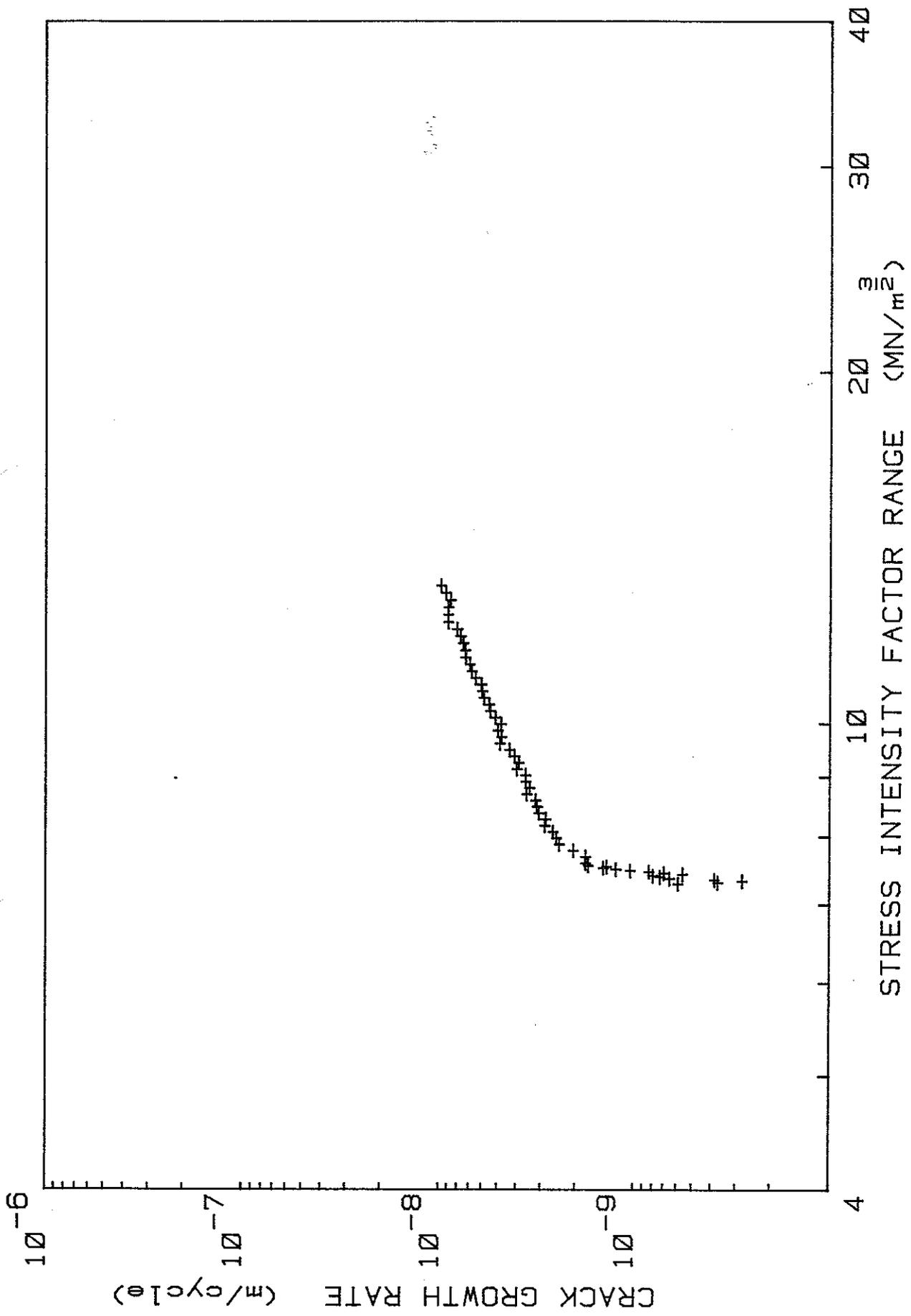


Figure 5 Typical log-log crack growth rate/stress intensity factor range plot

APPENDIX 1. SHIELDED TENSILE AND FATIGUE TESTING FACILITY.

|                          | G-2                                  | G-3                               | G-4                                   | G-7                             | G-8                             |
|--------------------------|--------------------------------------|-----------------------------------|---------------------------------------|---------------------------------|---------------------------------|
| Machine type             | Plain bearing actuator               | 1251                              | 1362                                  | 8031                            | 1342                            |
| Manufacturer             | Instron                              | Instron                           | Instron                               | Instron                         | Instron                         |
| Capacity static dynamic  | 50 kN<br>± 25 kN                     | 200 kN<br>± 100 kN                | 200 kN<br>± 100 kN                    | 100 kN<br>± 50 kN               | 200 kN<br>± 100 kN              |
| Valve                    | 10 l/min                             | 20 l/min                          | -                                     | 20 l/min                        | 20 l/min                        |
| Stroke                   | 50 mm                                | 15 mm                             | 50 mm                                 | 50 mm                           | 50 mm                           |
| Strain rate              | $10^{-6}$ - $1.\text{sec}^{-1}$      | $10^{-7}$ - $6.\text{sec}^{-1}$   | $10^{-6}$ - $10^{-1}.\text{sec}^{-1}$ | $10^{-6}$ - $6.\text{sec}^{-1}$ | $10^{-6}$ - $5.\text{sec}^{-1}$ |
| Electronics              | Digital<br>2180 console              | 3-Bay-<br>Analogue                | Digital<br>2180 console               | Digital<br>2160 console         | Digital<br>2160 console         |
| Data-acquis. and control | HP 9816<br>Dual Floppy               | HP 9816<br>Dual Floppy            | HP 310<br>10 Mb harddisc              | HP 9816<br>Dual Floppy          | HP 310<br>20 Mb harddisc        |
| Furnace                  | IR-lamp and vacuum<br>$10^{-7}$ torr | Resistance wire                   | Resistance wire or IR-lamp            | Resistance wire                 | Resistance wire                 |
| Test temp.               | up to 700°C                          | up to 600°C                       | up to 750°C                           | up to 600°C                     | up to 600°C                     |
| Test methods primary     | tensile                              | tensile                           | LCF-axial<br>-diam                    | crack growth                    | fracture mechanics              |
| additional               | crackgrowth                          | crackgrowth<br>fracture mechanics | tensile                               |                                 |                                 |
| Miscellaneous            | AC-Pot.Drop                          | Load-Line<br>Displ.Extens.        |                                       | DC-Pot.Drop                     | Dc-Pot.Drop                     |

# Accident Simulation Testing with Spherical HTR Fuel Elements

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Questions of reactor safety are occupying an increasingly dominant position in the R&D work for nuclear energy. In the KFA Jülich which, since the centre's foundation, has been engaged in the development of the pebble-bed High Temperature Reactor (HTR), investigations into the accident behaviour of spherical fuel elements are now well to the fore, following their successful development and testing. The central problem in the experimental studies is to find out whether the radioactive materials, the fission products, produced in chain reactions, are retained in the fuel element or not.

By means of heating tests simulating accident conditions, it has been possible in the Hot Cell Laboratories to measure the release of the safety-related fission products caesium, strontium and iodine from spherical fuel elements. These data enable us to understand fission product transport processes in the reactor core and are thus of great importance in demonstrating the safety of HTRs.

Because fission product release increases with temperature [1], accidents in which the highest temperatures can occur in the reactor core are being studied in detail. Since 1977, such accidents have been simulated with whole spherical elements, not least to draw attention to the HTR's safety potential. Advances in equipment have led to a high standard of experimental technology, enabling the fission product releases from fuel elements to be measured with precision over the whole range of accident temperatures of interest, i.e. from 1 600 to 2 500 °C.

The geometry of the fuel element, its simple construction and ceramic material are advantageous to safe operation [2] as shown in accident simulation tests. The 60 mm diameter fuel elements with their graphite matrix contain a large number of fuel particles, each fuel particle of 0.5 mm diameter being enclosed by several layers of gas-tight, pressure-resistant ceramic coatings which safely contain fission products.

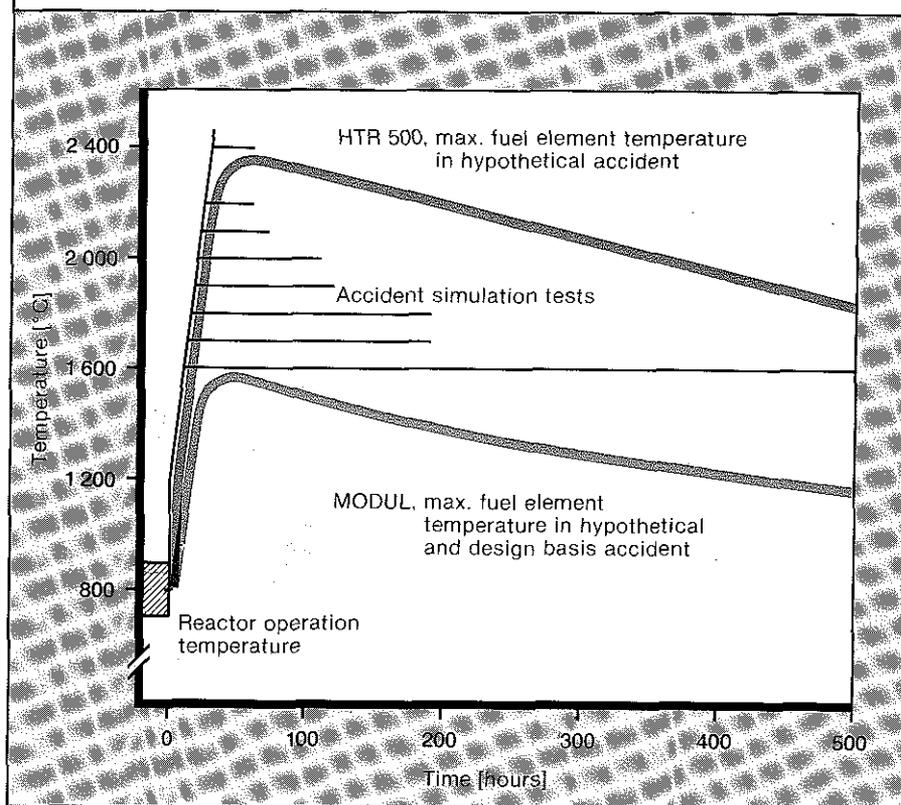
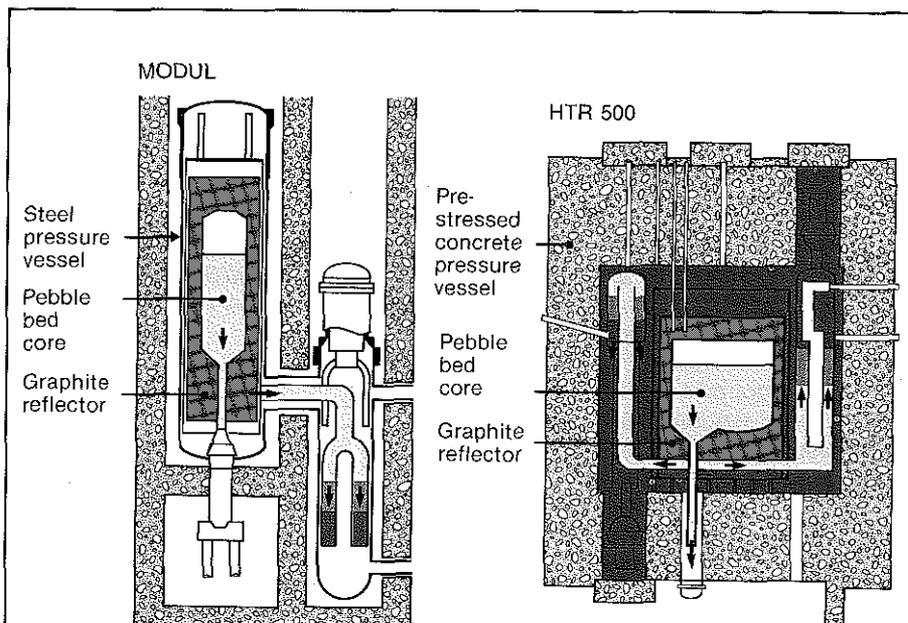


Figure 1: Two of the High Temperature Reactors presently planned are MODUL and HTR-500. Depending on power, power density and geometry of the reactor core, temperatures in HTR-500 would be much higher than in the MODUL in a depressurisation accident. However, the safety philosophy is different between the two reactor concepts. The temperatures in accident simulation tests are determined by the corresponding accident curves.

## HIGH TEMPERATURE REACTORS

There are at present two HTRs with spherical fuel elements in operation in West Germany and there exist three concepts for future HTR-reactors.

The AVR experimental power station, which has successfully operated for 20 years at Jülich, is of great importance for large scale testing of fuel. By means of this reactor, it is possible to supply the required number of spheres for accident testing under realistic conditions. From 1977 to 1982, extensive investigations were carried out with spherical fuel elements of the type used in the Thorium High Temperature Reactor (THTR) at Hamm-Uentrop [3].

More recently, accident simulation tests have concentrated on fuel elements of advanced design which are intended for the future reactors: MODUL, HTR 100 and HTR 500. Figure 1 shows schematic sections through MODUL and HTR 500, and the temperature curves predicted for depressurisation accidents in the two reactors [1]. The curves are not directly comparable due to the respective safety philosophies. In the case of the larger HTR 500, the maximum fuel design temperature in the licensing procedure is 1250 °C, because redundant cooling systems prevent any further core heat-up. Temperatures of 2300 °C thirty hours after the start of an accident would however be reached during an unrestricted core heatup accident in the highly unlikely event of all cooling systems in the reactor failing simultaneously with complete depressurisation. Simulation of fuel element behaviour during such accidents is necessary for risk analyses.

For small HTRs, such as the MODUL, the temperature curve up to 1500 °C (figure 1, bottom) is much more significant. This reactor is designed so that even in the event of an unrestricted core heatup accident with depressurisation, the fission products are completely retained within the fuel elements, which makes the safety tech-

| Reactor                           | AVR  | THTR | MODUL           | HTR 100 | HTR 500 |
|-----------------------------------|------|------|-----------------|---------|---------|
| Start of operation                | 1967 | 1986 | Future concepts |         |         |
| Thermal power, MW <sub>th</sub>   | 46   | 750  | 200             | 258     | 1250    |
| Electrical power, MW <sub>e</sub> | 15   | 308  | 80              | 100     | 500     |

nology much more simple. Design and safety concept of this reactor are based on the results of accident simulation tests.

Apart from the temperature range to be covered by an appropriate heating programme, the definition of the key fission products is of prime importance to the relevance of the test results. In the first weeks after a reactor accident, radioactive iodine plays a governing rôle in activity release. Because of their long half-lives, caesium and strontium nuclides are more significant in the long term. As a consequence, the fission products of these three elements dangerous to man must be investigated as accurately as possible. The radioactive noble gas nuclides of krypton and xenon are less important because they are not incorporated into the human body. They are, however, relevant in the study of particle failure mechanisms.

## TEST FACILITIES

In the early Seventies, tests at accident temperatures were carried out in the FRG and the USA only on single or small quantities of fuel particles. The facilities necessary for such small sample size have the advantage that they are simple, cheap and readily accessible since, because of the small amounts of activity, it is not necessary to shield them by thick walls. Some major fission product transport data were determined from these tests and then applied in release calculations. The results, however, varied considerably and lead to a large error band; but

| Important fission products |         |           |
|----------------------------|---------|-----------|
| ELEMENT                    | ISOTOPE | HALF LIFE |
| Solid fission products     |         |           |
| Caesium                    | Cs 137  | 30 years  |
|                            | Cs 134  | 2 years   |
| Strontium                  | Sr 90   | 29 years  |
| Iodine                     | I 131   | 8 days    |
| Fission gases              |         |           |
| Krypton                    | Kr 85   | 11 years  |
| Xenon                      | Xe 133  | 5 days    |

this is not the only reason why tests on complete spherical fuel elements are much more desirable:

Depending on its type, there are 10,000 to 40,000 particles in a spherical fuel element. This means that in a heating test of an element, very low particle defect fractions can be determined and a reliable statistical mean value may be obtained for fission product release from the very large number of particles. Moreover, there are small quantities of heavy metal in the matrix graphite, due to manufacture, from which fission products originate and may be released. The matrix graphite may also house fission products com-

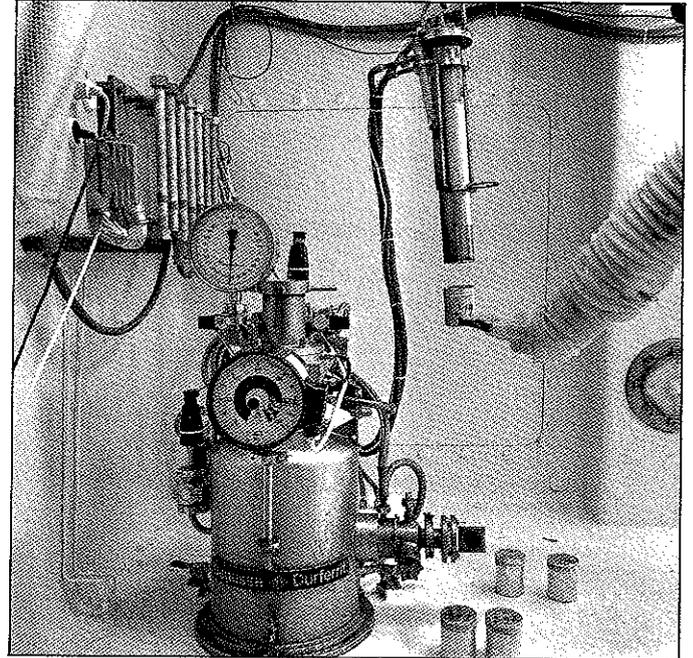
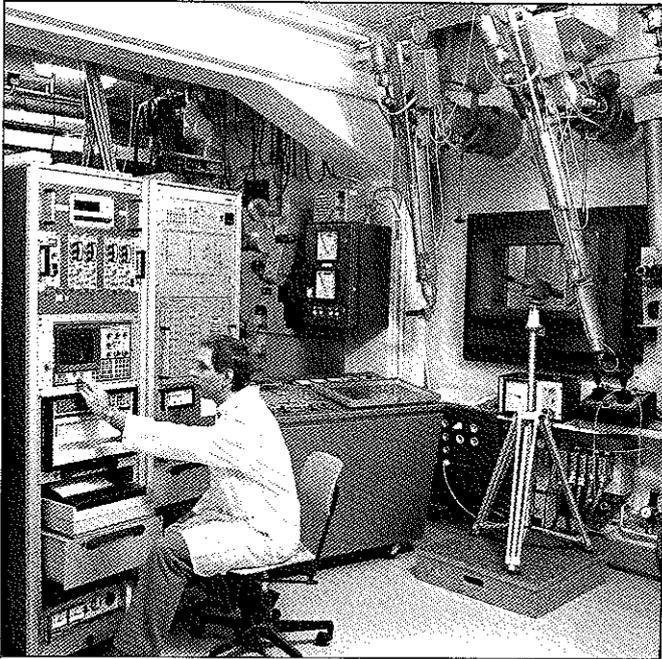


Figure 2: The photograph on the left shows the operating equipment for the cold finger apparatus, viz. electronic controls for fission gas measurements, control panels for the sweep gas circuit and furnace, and hot cell window with manipulators. The right-hand side photograph shows the cold finger furnace inside the cell box. The manipulator arm is gripping an aluminium can for reception of a condensation plate from the cold finger.

ing from defective particles and which will be delayed in the graphitic material.

The test requirements arising from this problem led to the development of highly-specialised test equipment capable to cope with complete fuel elements. With the heatup rig designed for accident simulation tests, temperatures up to 2 500 °C can be reached. Apart from the fission gases such as krypton, the release of caesium was determined via the overall gamma-spectrometric measurement of the spheres before and after a heating test. This meant that small caesium releases remained undetected. Therefore, a new design became necessary, especially suited to demonstrate that the fission product release due to an accident of small HTRs is sufficiently low. The equipment for this rig is shown in figure 2. The differences between the two rigs are compared in table 3.

Table 3: The accident simulation facilities enable heating tests to be carried out on complete spherical fuel elements in flowing helium. With the cold finger test rig, solid fission products released at up to 1 800 °C can be determined quantitatively with high precision. At temperatures above 2 000 °C, determination of the caesium loss from an element can be accomplished via the differential method, i.e. by means of measurements before and after the test.

|   | Cold finger test rig<br>(since 1984)           | Heatup test rig<br>(since 1977) |
|---|--|---------------------------------|
| Maximum temperature   | 1 800 °C                                       | 2 500 °C                        |
| Experimental range  | Design basis accidents                         | Hypothetical accidents          |
| Fission gas measurement   | Krypton release profile                        |                                 |
| Measurement of solid fission products                                   | Caesium, strontium and iodine release profiles | Overall release of caesium      |
| Sensitivity of release measurement (Fraction of fuel element inventory) | very high<br>(10 <sup>-8</sup> )               | low<br>(10 <sup>-2</sup> )      |

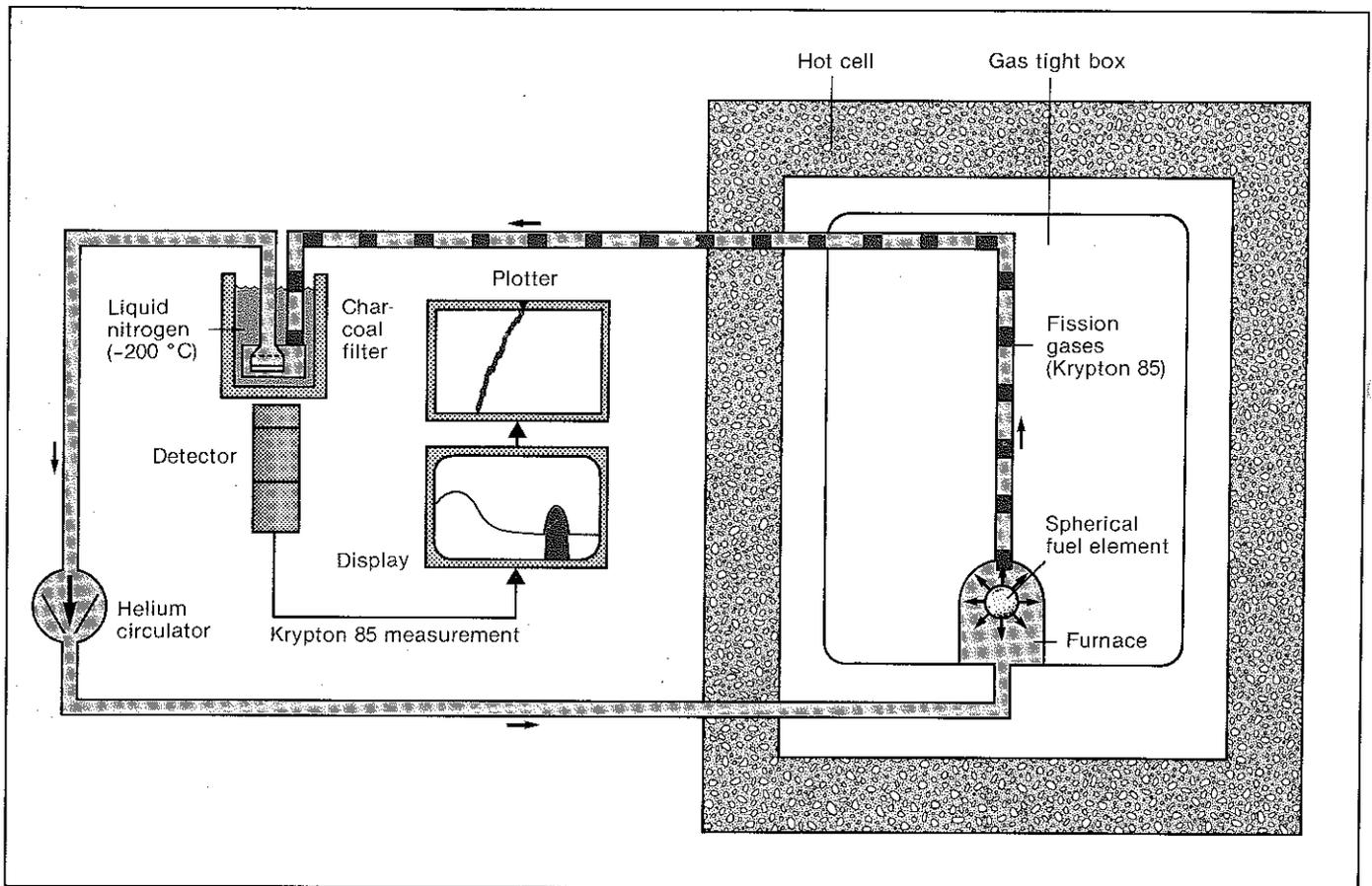


Figure 3: Gaseous fission products like Kr 85 released from a fuel element during the heating test are swept out of the Hot Cell by the helium loop. Measurements of krypton are performed in a cooled charcoal filter.

## MEASUREMENT OF FISSION GASES

The measurement of fission gases under accident conditions is important because the release curves indicate individual particle failures in the fuel element which lead to gas leakage. Also, conclusions can be drawn about iodine release which is difficult to measure directly, but which is released to the same extent as krypton. Whereas the two test rigs differ in temperature range and the ability to determine solid fission products, the method for determining fission gas release is the same for both.

For this purpose, the furnaces installed in a gas-tight box in the Hot Cell are incorporated in helium circuits (figure 3). A compressor circulates the helium in

the circuit. Whilst the solid fission products released from a fuel element in a heating test are largely deposited in the furnace, the remainder is retained in the cell by means of absolute filters in the "hot" section of the rig. The fission gases are taken with the sweep gas to the measuring trap outside the Hot Cell. The released fission gases are adsorbed in the active charcoal filter at liquid nitrogen temperature. The activity in the measuring trap is determined by on-line gamma spectrometry throughout the whole test. In general, only long-lived krypton 85 can be detected. Provided the cooling period of the fuel elements, i.e. the time between discharge from the reactor and start-up of the test, is less than 4 to 8 weeks, measurements of xenon 133 are possible as well.

## MEASUREMENT OF SOLID FISSION PRODUCTS

The determination of solid fission products is more complicated than that of the chemically inert fission gases. At high temperatures they can get into the coolant gas by desorption from the surface of the sphere, changing from the solid to the gaseous phase. Conversely, such fission products are re-deposited by adsorption on cooler surfaces, and this deposition mechanism is exploited for trapping solid fission products in the cold finger test rig.

The fuel element is supported by three pins in the centre of a tantalum tube in the furnace; helium flows through the tube from bottom to top (figure 4). The tantalum tube and fuel element are heated by an electrical resistance

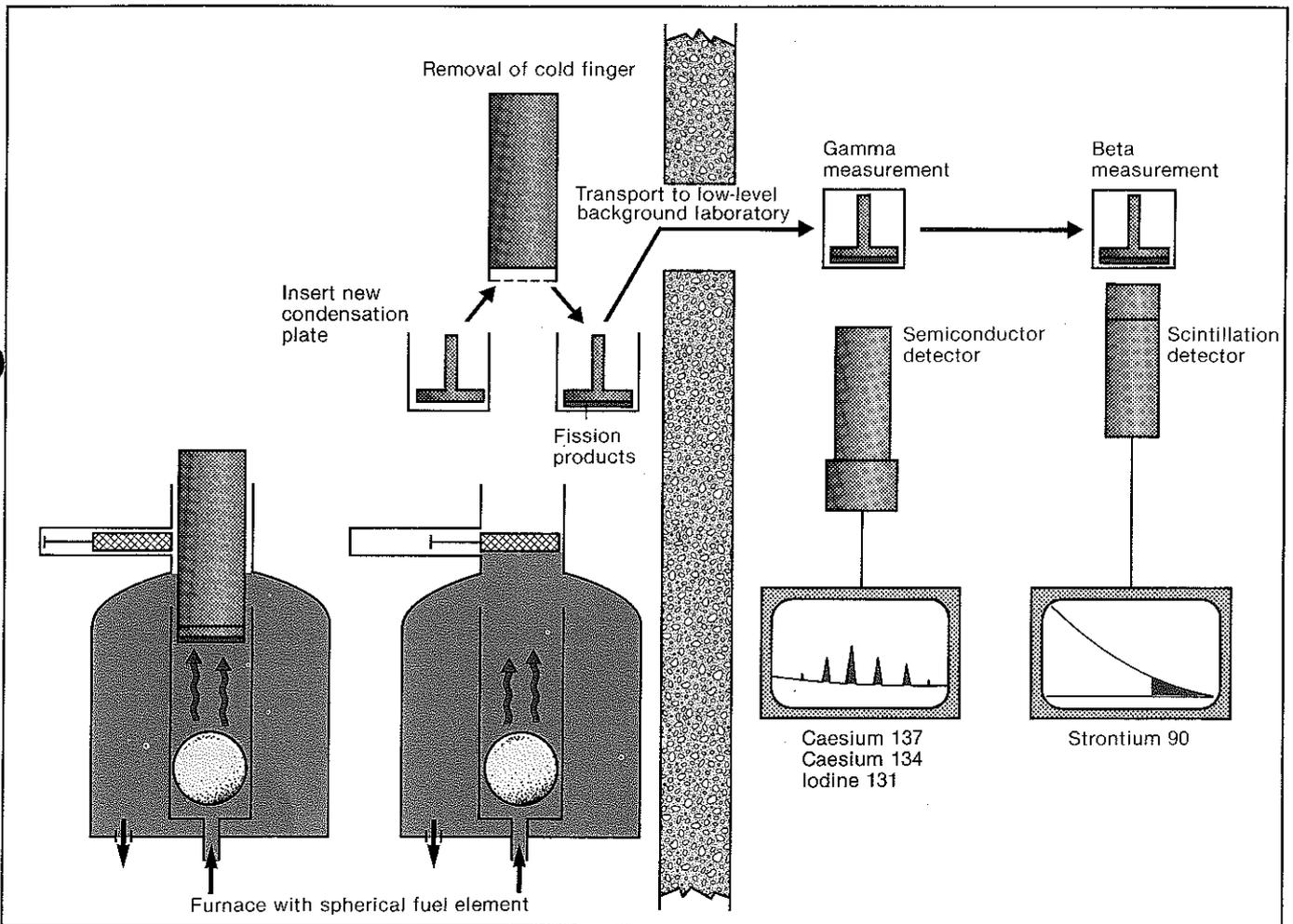


Figure 4: Solid fission products released from a spherical fuel element during the heating test are deposited on the condensation plate which can be exchanged during heating. These plates are measured by beta- and gamma-spectrometry outside the Hot Cells.

heater which likewise consists of tantalum. Into the hot tantalum tube protrudes a water-cooled cold finger, at the end of which a replaceable condensation plate is held. The solid fission products released from the fuel element are deposited on this plate, which has a temperature of less than 100°C with a sphere temperature of 1 600—1 800°C.

During the test, the cold finger can be removed from the furnace via an air-lock system without cooling down the sphere. After replacing the condensation plate, the cold finger is returned

to the furnace again. The plate is normally changed once or twice a day, but this can be done more often since the procedure only takes a few minutes.

The used plates are taken out of the Hot Cell to be measured by gamma- and betaspectrometry in a low-background laboratory. Measurement of gamma emitters such as caesium 137 and iodine 131 is relatively simple, because they can be identified by their individual energy lines. Strontium 90, a nuclide which emits only beta radiation, has to be separated chemically from other fission products for meas-

urement. Since strontium emits beta rays with the highest energy of the deposited fission products, the activity can also be estimated with a scintillation counter after a special calibration procedure.

The results plotted in figure 6 illustrate the great advance achieved with the new rig in the determination of fission product release. The two upper curves were determined from a large number of heating tests, which individually gave one measurement point for which the detection limit lay at 1% (10<sup>-2</sup>). In the heatup rig it would have been im-

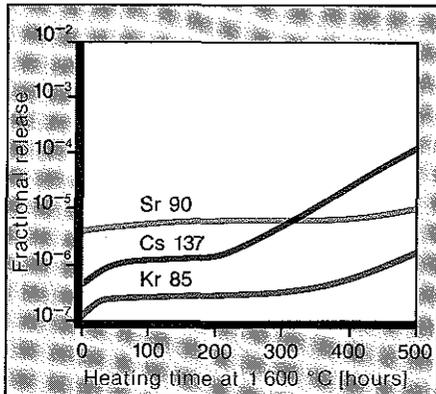


Figure 5: The release of key fission products caesium, strontium and iodine (similar to krypton) is small at 1600°C ( $10^{-6}$  = 1 millionth of the amount available in the fuel element).

possible to determine the variation in release from fuel particles with a SiC intermediate coating, since it is too low. Using the cold finger test rig, even the smallest fractional release down  $10^{-8}$  (ten billionths) can now be measured.

## ACCIDENT SIMULATION TESTING

The highest temperatures in the reactor core occur in depressurisation events with loss of all cooling systems. Since the chain reaction has then ended, only the comparatively low afterheat is still being generated due to radioactive decay. This leads to a constant temperature profile over the fuel element. Gas pressure and temperature distribution can be simulated very accurately in the heating tests. Since at the start of an accident, the fuel elements in the reactor heat up from the operating temperature, in the standardised accident simulation tests, following a purification bake-out at 300°C, the fuel element is held at elevated operating temperatures of 1050°C and 1250°C. This is primarily intended to set up steady state conditions to create the same circumstances as at the start of an accident in the reactor. Only then does the actual accident simulation begin and heating

takes place at the rate of 50°C per hour up to the target temperature. When reached, this temperature is held constant for a maximum of 1000 hours (figure 1).

## FISSION PRODUCT SOURCES

The most important results in accident simulation tests with spherical fuel elements are the values of fission product release [5, 6]. Fission products can originate from different sources in the fuel element. The first branch in the fission product release curve is usually due to matrix graphite contamination, either due to manufacture or received in the reactor. In the case of modern fuel elements, the fraction of heavy metal contamination due to manufacture is extremely small, namely less than  $5 \times 10^{-5}$  (five hundred-thousandths) of the uranium present.

In general, fission products are released during accident simulation testing when particle coatings fail. The 0.035 mm thick silicon carbide (SiC) intermediate coating is the most important fission product barrier. Two mechanisms may lead to coating damage:

- *corrosion of SiC* due to chemical reaction with fission products begins below 2000°C and takes place relatively slowly,
- *thermal decomposition of SiC*, disintegration of its structure as a result of high temperature, predominates above 2000°C and can take place relatively quickly as the temperature increases.

## FISSION PRODUCT RELEASE AT 1600°C

Typical fission product release curves for a fuel element heated at 1600°C are shown in figure 5. The fractional releases are very low and over the first 200 hours entirely due to contamination in the graphite. After 300 hours, the caesium release increases more steeply, which is attributed to the start of SiC degradation. The krypton and

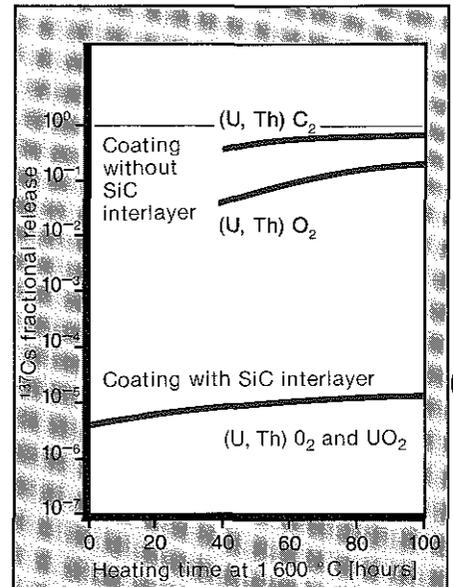


Figure 6: Measurements of caesium release from several fuel elements at 1600°C show the good retention potential of the SiC intermediate layer of particles in modern fuel elements.

strontium releases also increase, but with a time shift and to a lesser extent than of caesium. Strontium diffuses more slowly through the fuel kernel and matrix graphite, whereas noble gases are retained better than caesium by the pyrocarbon layers which still remain intact.

The Cs 137 releases at 1600°C from different types of fuel elements are compared in figure 6. Whereas intact SiC coatings represent an effective barrier for caesium for the first 100 hours at this temperature, caesium diffuses relatively quickly through a coating of pyrolytic carbon under the same conditions. On the other hand, fission gases and iodine are completely retained in all types of particles at 1600°C.

## FISSION PRODUCT RELEASE AT 1800°C

While at 1600°C the SiC intermediate coating of the particles of modern HTR fuel elements is only damaged

after more than 200 hours and initially only to a slight extent, this period reduces with increasing temperature. At 1800 °C, the increasing caesium release shows silicon carbide degradation already after 20 hours. For the same reasons as at 1600 °C, the strontium, fission gas and iodine releases are lower than that of caesium. However, the point in time and extent of SiC degradation in an accident simulation test are not dependent solely on temperature and time, but also on the irradiation conditions in the reactor. SiC damage occurs more quickly with increasing burnup (fissions of heavy metal atoms), fluence (irradiation by high energy neutrons) and higher irradiation temperature.

## FISSION PRODUCT RELEASE ABOVE 2 000 °C

Above 2 000 °C, silicon carbide is damaged by thermal decomposition relatively quickly. The SiC coating of particles in the fuel elements studied are destroyed to an increasing extent when heated up at 50 °C per hour from 2 200 °C. This can be clearly seen in figure 7 from the variation in Kr 85 release from a number of fuel elements. When heating up from 1 250 °C to 2 200 °C, still no damage occurs in this time period. At 2 200 °C, thermal decomposition then leads relatively quickly to SiC failure and the coating becomes ineffective as a barrier for strontium and caesium. The pyrolytic carbon layers remain intact which prevents complete release of fission gases above 2 200 °C.

## CERAMOGRAPHY OF FAILED SiC

Damage to the SiC coating can be made visible by means of ceramographic examinations (figure 8). Particles taken from heated fuel elements are first polished to the centre and then photographed through a microscope. The

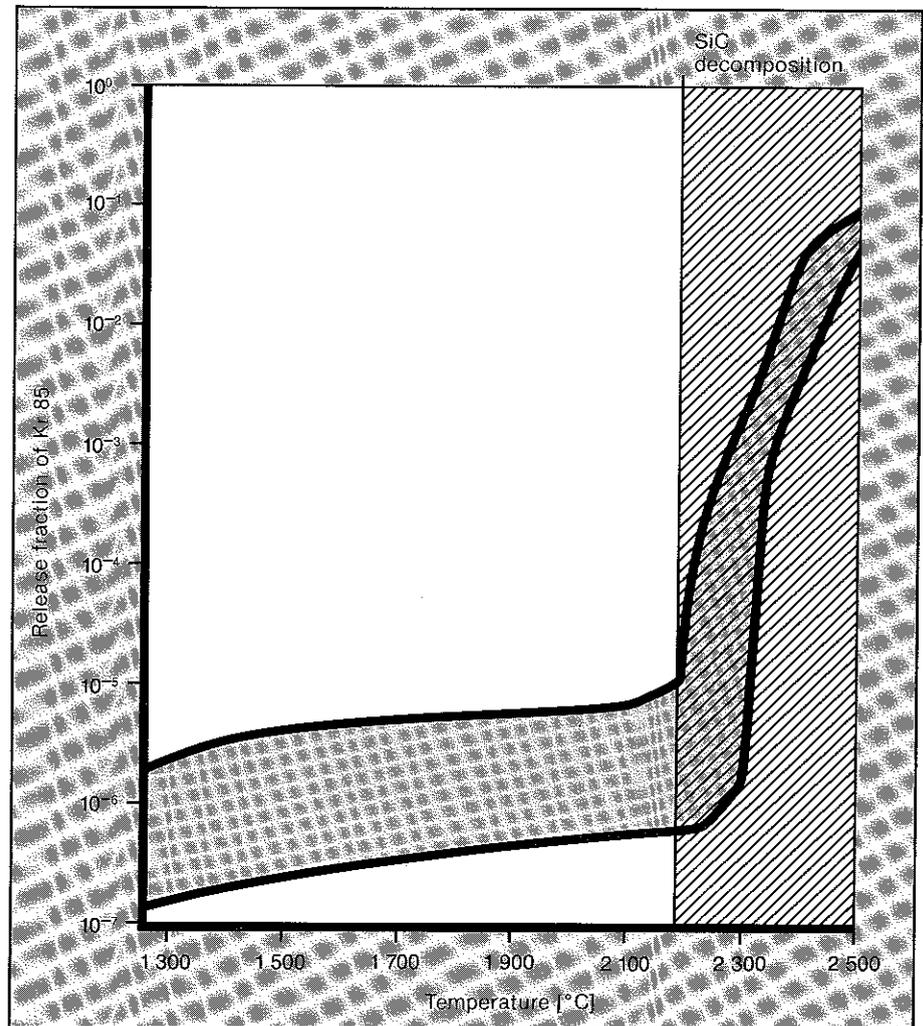


Figure 7: The release of Kr 85 from spherical fuel elements during temperature ramps to 2 500 °C shows that failure of particles with SiC coating occurs above 2 200 °C.

white SiC coating layer of the particle from the fuel element tested for 160 hours at 1 600 °C still shows no change in comparison with an unheated particle which is also confirmed by the fission product release curve remaining low during the accident simulation test. Damage to the SiC can be seen in the particle exposed to 1 700 °C for 185 hours.

At 2 400 °C (figure 8 bottom right), decomposition of the SiC coating is clearly visible, whereas the pyrocarbon lay-

ers surrounding the silicon carbide have remained intact.

## SUMMARY OF RESULTS

Figure 9 shows a highly simplified summary of the fission product release measurements obtained so far from modern spherical fuel elements. Here a heating period of 100 hours is conservatively taken into account for the temperature regime at 1 600 °C impor-

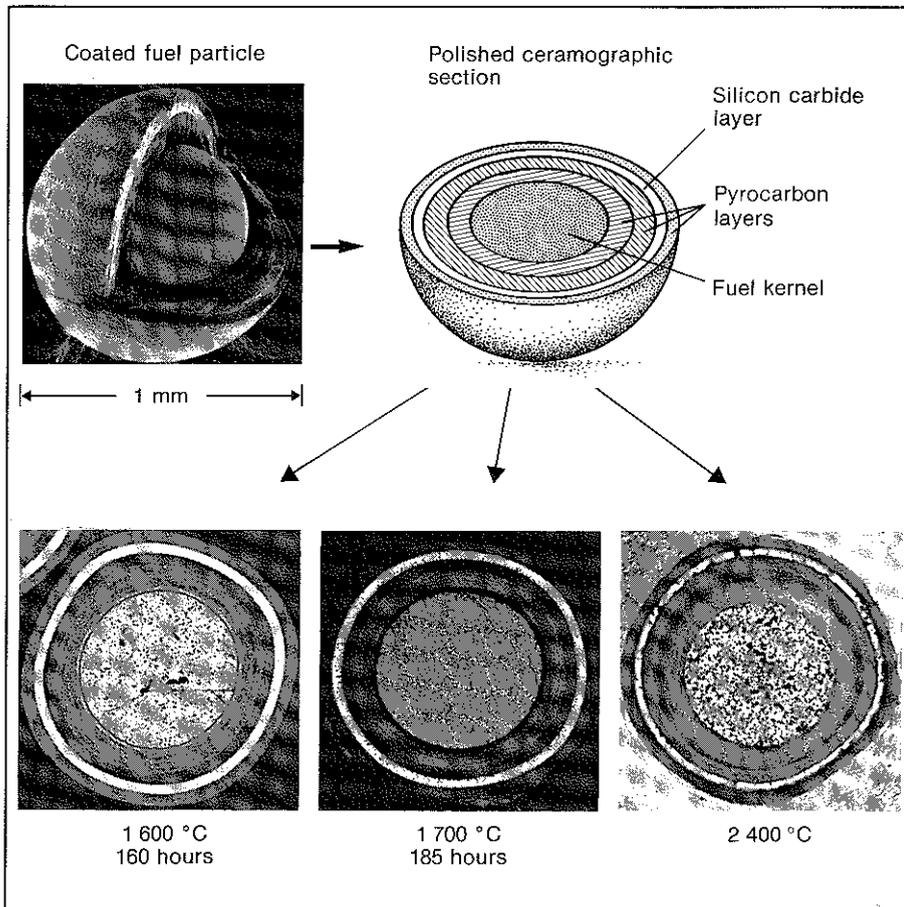


Figure 8: Polished ceramographic sections through heated particles show the degradation of the silicon carbide layer at very high temperatures. This SiC degradation leads to fission product release.

tant to the design and licensing of small HTRs. Figure 1 shows that the calculated maximum temperatures in the core centre of a modular HTR are in the region of 1 500 °C for a short time only. The heating tests at 1 600 °C showed that even beyond 100 hours, the fractional fission product release due to an accident is negligibly small. Only at approximately 1 800 °C does the onset of damage in the SiC layer lead to an obvious rise. However, fission product release at 1800 °C is still small after 30 hours.

Beyond 2 000 °C, SiC failure leads to noticeable fission product release, an effect which is increasing with higher temperatures. Those conditions are representative for hypothetical, i.e. extremely unlikely, accidents of a medium-sized HTR. However, activity release to the environment would still be minute, because nearly all of the activity released from the fuel will be deposited in the cooler parts of the reactor. These effects are also being studied in the KFA.

## FUTURE PROSPECTS

Six accident simulation tests with fuel elements intended for future HTRs have so far been carried out in the cold finger test rig at 1 600 to 1 800 °C and a further seven in the heatup test rig at 1 800 to 2 500 °C [6]. The very accurate fission product release data obtained for the first time on complete spherical fuel elements and the associated findings on individual particle failures at 1 600 to 1 800 °C led to a reassessment of modern fuel elements in this temperature range — a reassessment which is extremely important for the safety concept of small HTRs. It is now necessary to validate and extend

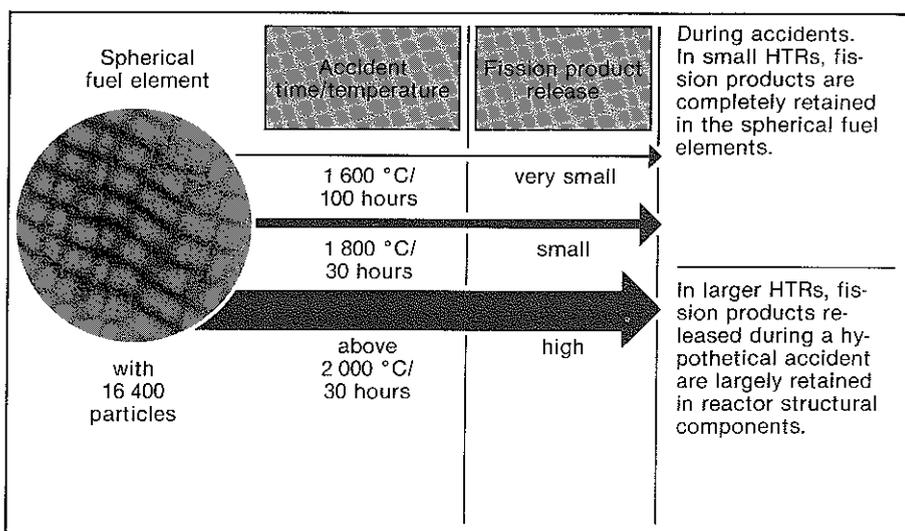


Figure 9: The release of fission products relevant to accident consequences — caesium 137, iodine 131 and strontium 90 — from modern HTR fuel elements at 1 600 °C is practically zero. At 1 800 °C the release is increasing due to changes in the particle coating. Above 2 000 °C, coating failure and increased diffusion through intact coatings lead to high fission product release from spherical fuel elements.

the findings by further investigations. Priorities in future programmes will be:

- tests on high burnup modern fuel elements available from AVR in 1988,
- improvement of the statistics from the 1 600 to 1 800 °C heating tests to substantiate extrapolations to the 500,000 fuel elements in an HTR core.

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