Accident Simulation Testing with Spherical HTR Fuel Elements

W. Schenk and H. Nabielek,
Kernforschungsanlage Jülich GmbH, Federal Republic of Germany

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.

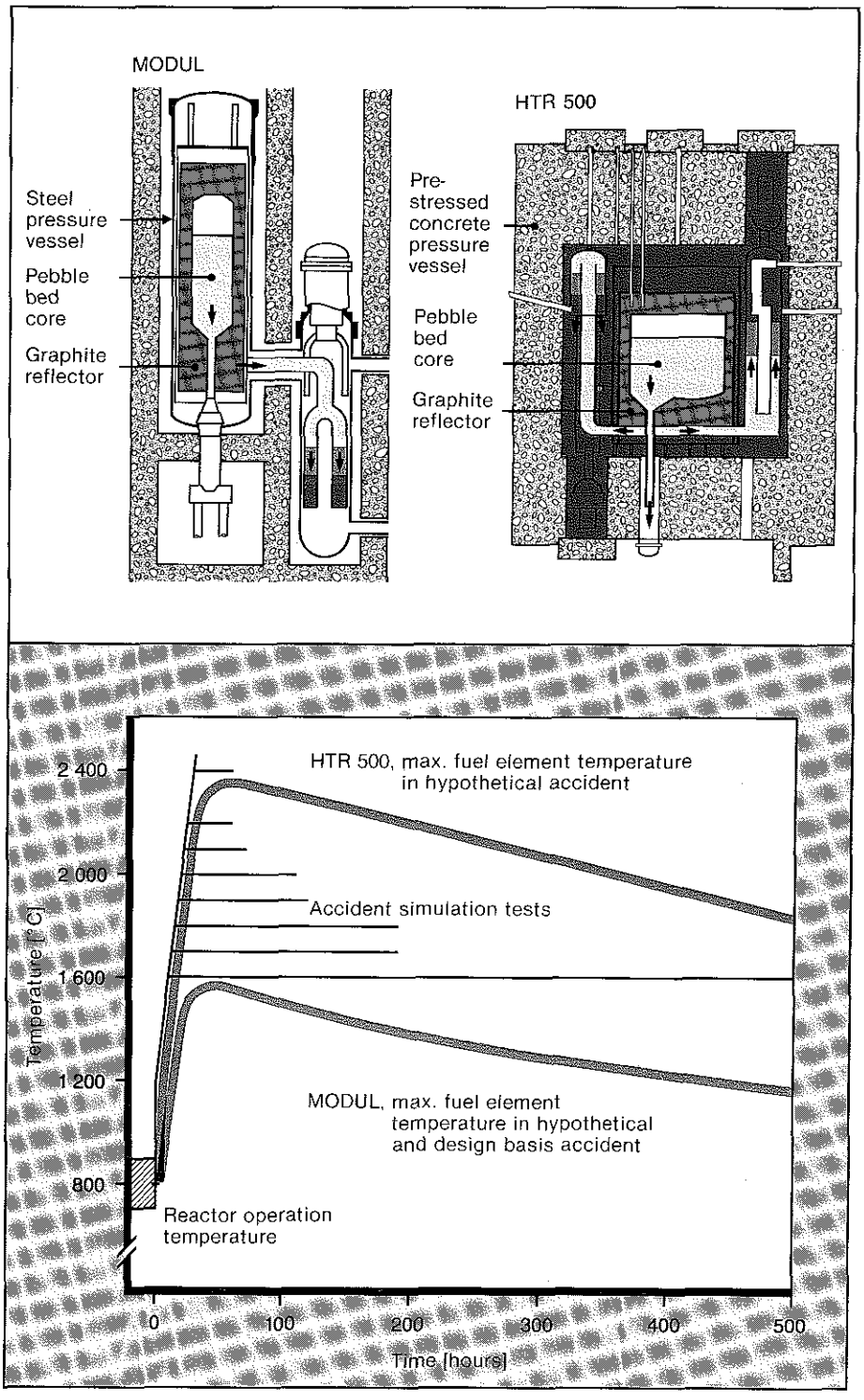

Figure 1: Two of the High Temperature Reactors presently planned are MODUL and HTR-500. Depending on power, power density and geometry of the reactor core, temperatures in HTR-500 would be much higher than in the MODUL in a depressurisation accident. However, the safety philosophy is different between 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 1 250 °C, because redundant cooling systems prevent any further core heatup. Temperatures of 2 300 °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 1 500 °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 technology much more simple. Design and safety concept of this reactor are based on the results of accident simulation tests.

Table 1: High Temperature Reactors with spherical fuel elements

Reactor	AVR	THTR	MODUL	HTR 100	HTR 500
Start of operation	1967	1986	Future concepts		
Thermal power, MW_{th}	46	750	200	258	1250
Electrical power, MW_e	15	308	80	100	500

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.

Table 2: The fission products caesium, strontium and iodine are radiologically significant because, unlike the noble fission gases, they can be incorporated in the human body.

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

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 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-

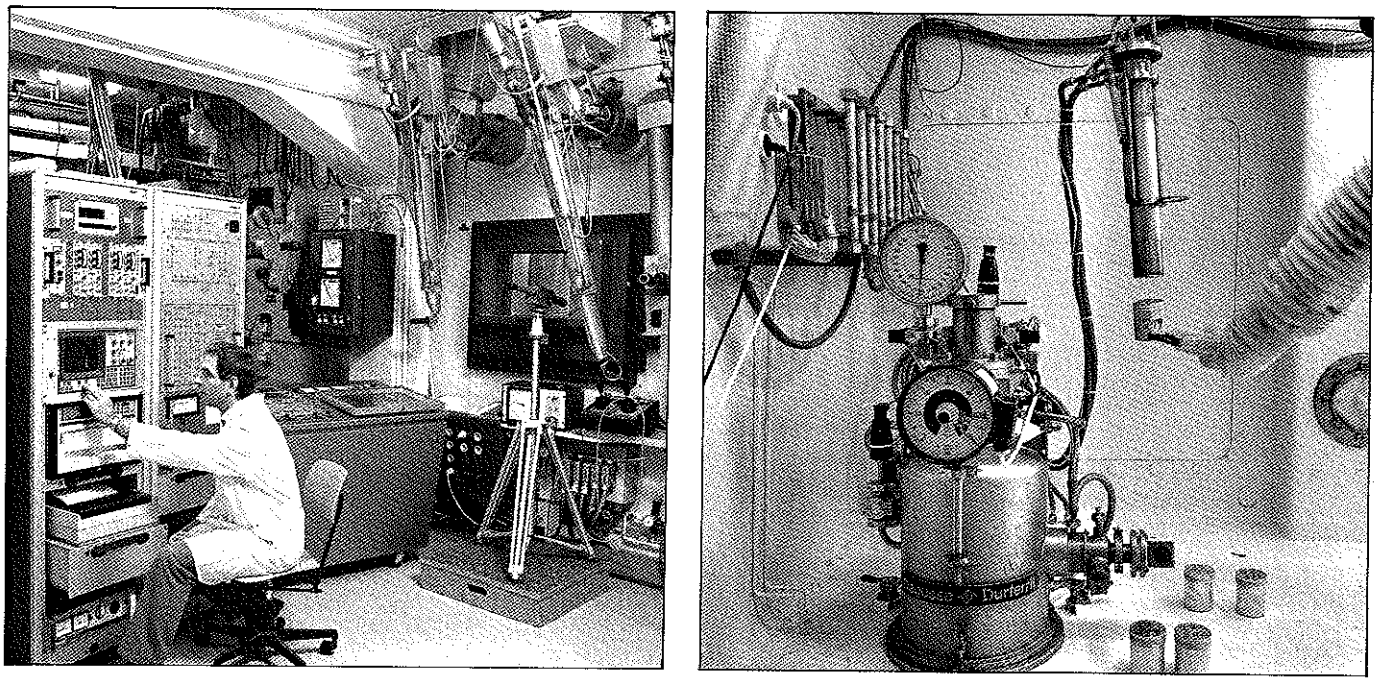

Figure 2: The photograph on the left shows the operating equipment for the cold finger apparatus, viz. electronic controls for fission gas measurements, control panels for the sweep gas circuit and furnace, and hot cell window with manipulators. The right-hand side photograph shows the cold finger furnace 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 (since 1984)	Heatup test rig (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 (10^{-8})	low (10^{-2})

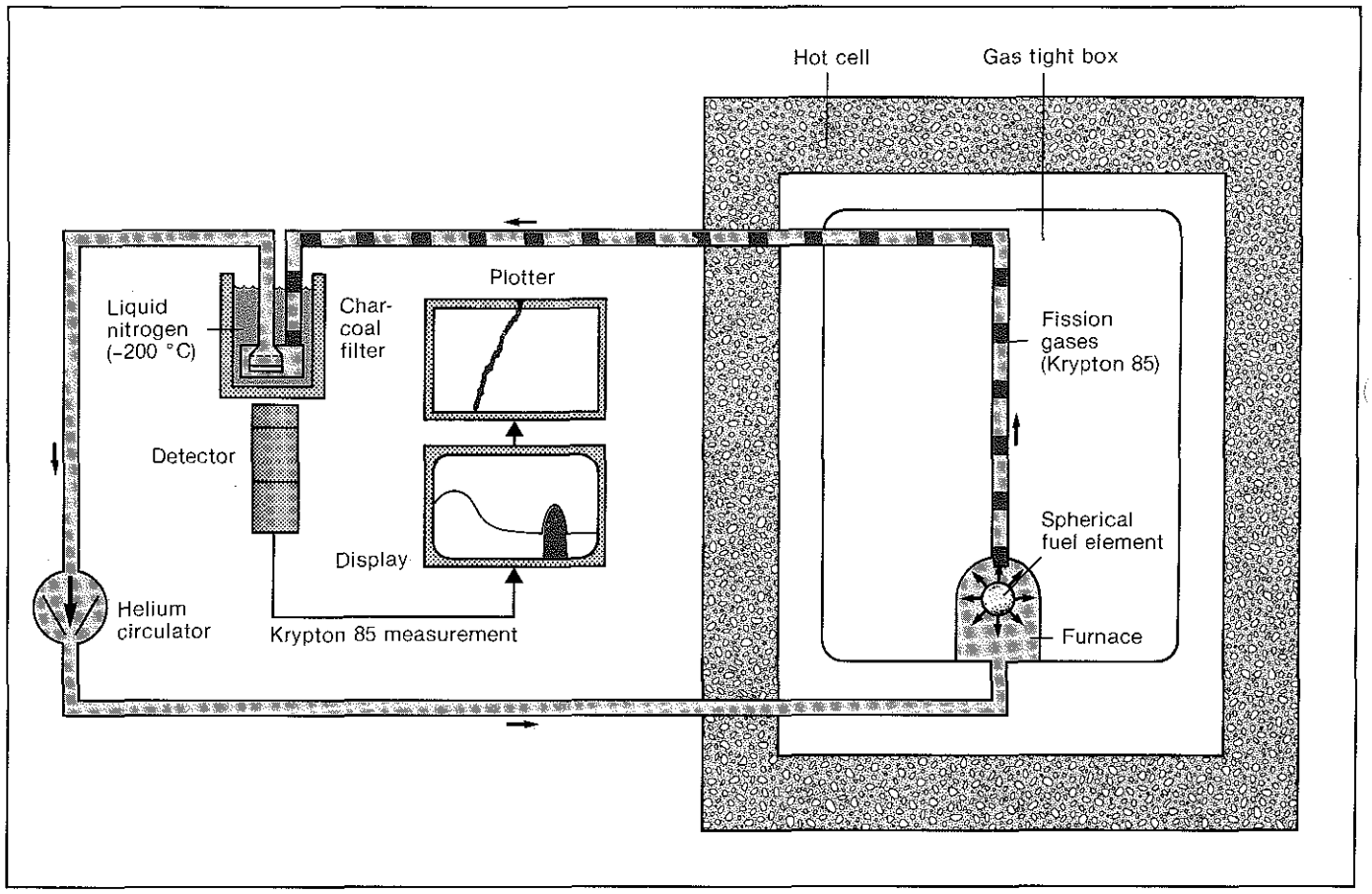

Figure 3: Gaseous fission products like Kr 85 released from a fuel element during the heating test are swept out of the Hot Cell by the helium loop. Measurements of krypton are performed in a cooled charcoal filter.

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 gammaspectrometry 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 redeposited 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

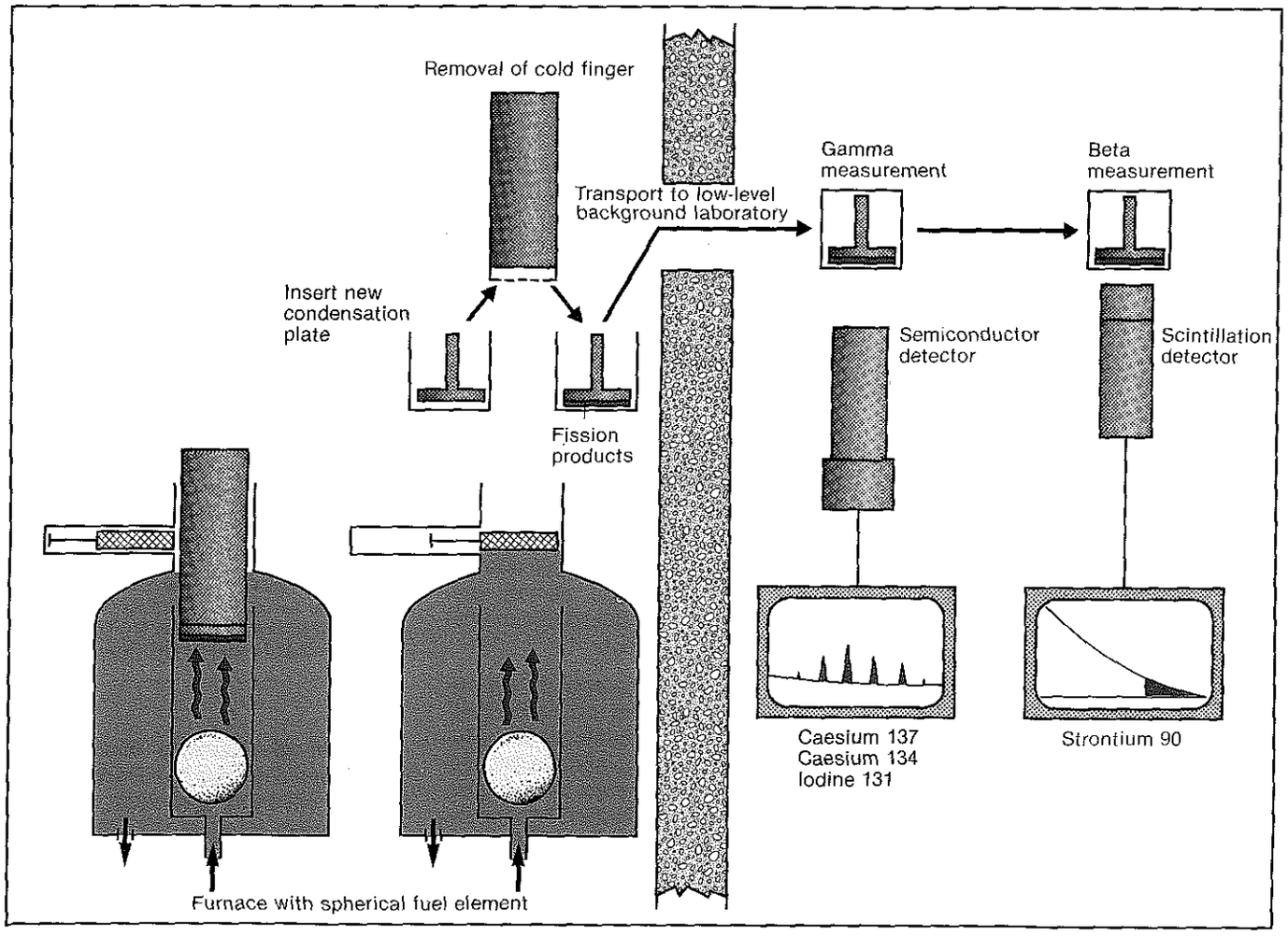

Figure 4: Solid fission products released from a spherical fuel element during the heating test are deposited on the condensation plate which can be exchanged during heating. These plates are measured by beta- and gamma-spectrometry outside the Hot Cells.

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 airlock 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 measurement. 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^{-2}). In the heatup rig it would have been im-

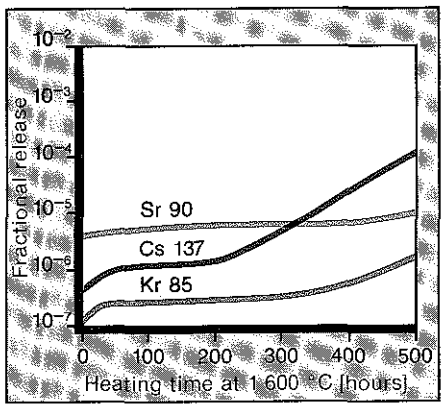

Figure 5: The release of key fission products caesium, strontium and iodine (similar to krypton) is small at 1 600 °C (10^{-6} = 1 millionth of the amount available in the fuel element).

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 1 050 °C and 1 250 °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 1 000 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 5×10^{-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 2 000 °C and takes place relatively slowly,
- *thermal decomposition of SiC,* disintegration of its structure as a result *of high temperature, predominates* above 2 000 °C and can take place relatively quickly as the temperature increases.

FISSION PRODUCT RELEASE AT 1 600 °C

Typical fission product release curves for a fuel element heated at 1 600 °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

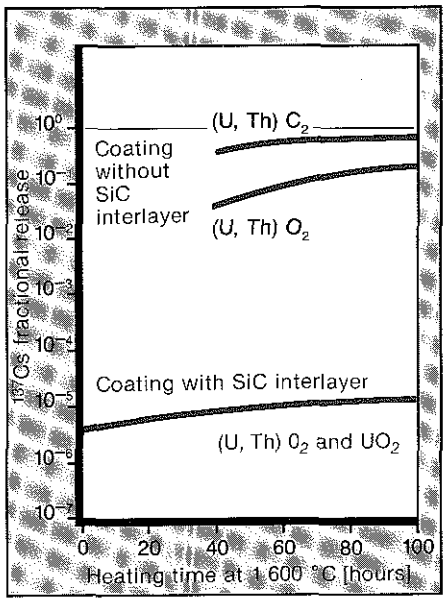

Figure 6: Measurements of caesium release from several fuel elements at 1 600 °C show the *good retention potential of the SiC intermediate* layer of particles in modern fuel elements.

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 1 600 °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 pyrolitic carbon under the same conditions. On the other hand, fission gases and iodine are completely retained in all types of particles at 1 600 °C.

FISSION PRODUCT RELEASE AT 1 800 °C

While at 1 600 °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 1 800 °C, the increasing caesium release shows silicon carbide degradation already after 20 hours. For the same reasons as at 1 600 °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 pyrolitic carbon layers remain intact which prevents complete release of fission gases above 2 200 °C.

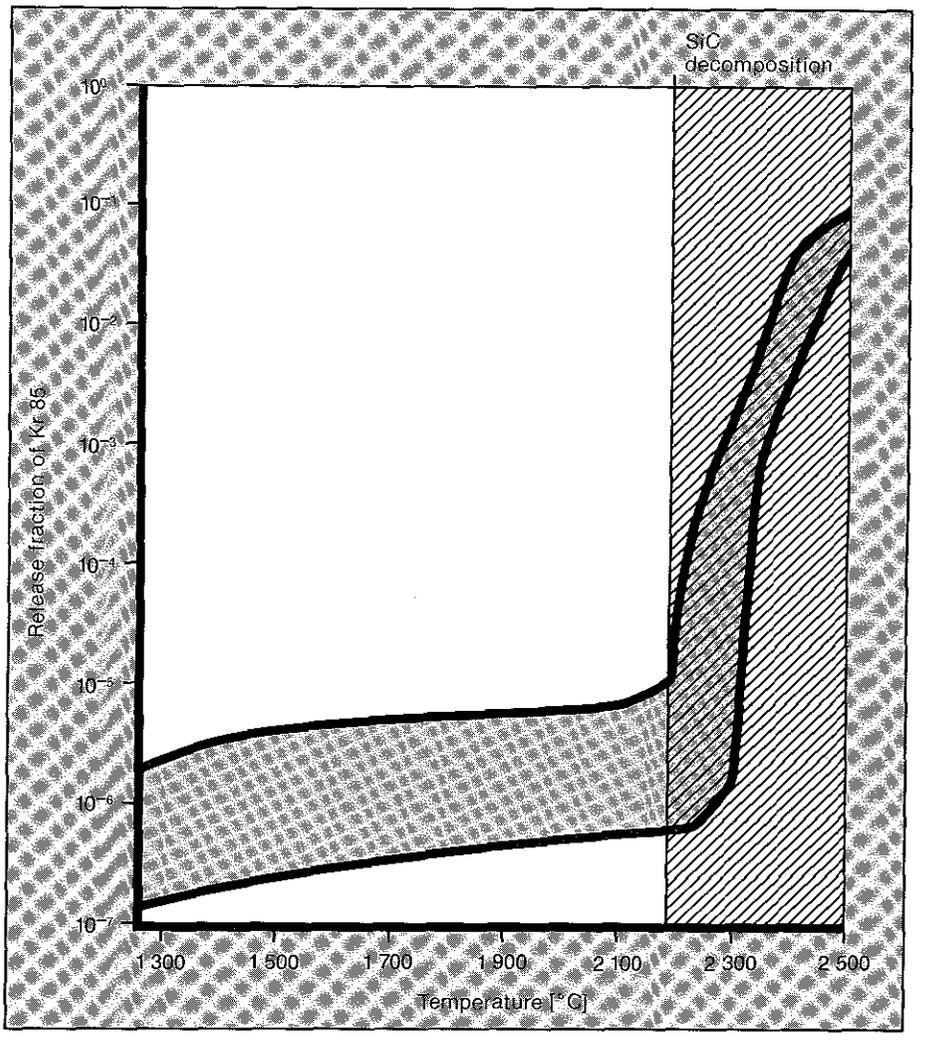

Figure 7: The release of Kr 85 from spherical fuel elements during temperature ramps to 2 500 °C shows that failure of particles with SiC coating occurs above 2 200 °C.

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 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 layers 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-

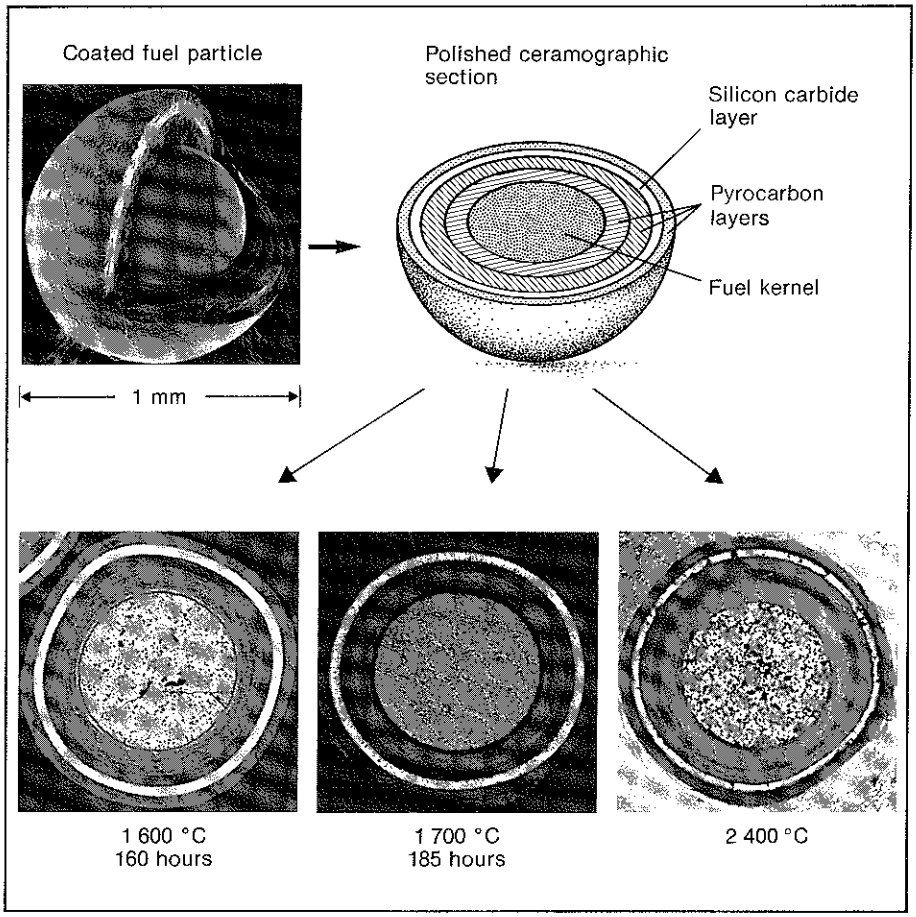

Figure 8: Polished ceramographic sections through heated particles show the degradation of the silicon carbide layer at very high temperatures. This SiC degradation leads to fission product release.

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.

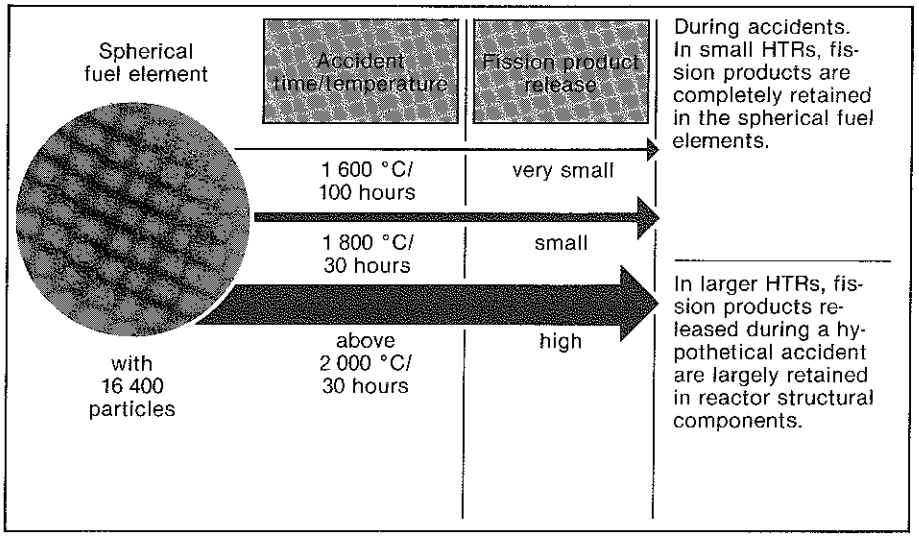

Figure 9: The release of fission products relevant to accident consequences — caesium 137, iodine 131 and strontium 90 — from modern HTR fuel elements at 1 600 °C is practically zero. At 1 800 °C the release is increasing due to changes in the particle coating. Above 2 000 °C, coating failure and increased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

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

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.

LITERATURE

[1] W. Rehm and H. Unger (ed): Safety of High Temperature Reactors, KFA Report, Jül-Conf-53, June 1985

[2] H. Nickel: Long-term testing on HTR fuel elements in the Federal Republic of Germany, KFA Report, Jül-Spez-383, Dec 1986

[3] W. Schenk and A. Naoumidis: Failure mechanisms and fission product release in High Temperature Gas-cooled Reactor fuel under conditions of unrestricted core heatup events, Nuclear Technology, Vol 46, p 228, Dec 1979

[4] H. Nabielek, G. Kaiser, H. Huschka, H. Ragoss, M. Wimmers and W. Theymann: Fuel for pebble-bed HTRs, Nuclear Engineering and Design, 78, p 155, April 1984

[5] W. Schenk: Accident simulation on irradiated spherical fuel elements at temperatures of 1 400 to 2 500 °C, KFA Report, Jül-1883, December 1983

[6] W. Schenk, D. Pitzer and H. Nabielek: Fission product release profiles from spherical HTR fuel elements at accident temperatures, KFA Report, Jül-2091, October 1986

PHENIX

ACTIVITES DES PRODUITS D'ACTIVATION EN GBq PAR cm^3 (acier des gaines et tube hexagonal)

fonctionnement	725 JEPP		
refroidissement	1 an	2 ans	3 ans
activité totale en GBq/cm^3	**41,2**	**26,3**	**20,0**
activité totale en Ci/cm^3	**1,1**	**0,7**	**0,5**

Tableau 1

PHENIX

ACTIVITE DES PRODUITS DE FISSION EN GBq PAR cm^3 DE COMBUSTIBLE (UPu) O_2

activité totale en GBq/cm^3	2708	1410	855
activité totale en Ci/cm^3	73	38	23

Tableau 2

PHENIX

ACTIVITE α DES ATOMES LOURDS COMBUSTIBLE $(UPu)O_2$

	GBq/cm^3 $(UPu)O_2$
activité totale en GBq/cm^3	263
activité totale en Ci/cm^3	7

Tableau 3

HISTOGRAMME DES DOSES REÇUES LORS DE LA DECONTAMINATION

REFECTION DE LA CELLULE 3

dose reçue (m rem)	nombre d'agents
10 à 200	32
200 à 400	28
400 à 600	3
1200	1

Tableau 4

	CELLULE 3 (1986)			CELLULE 9 (1989) (prévision)		
	durée (mois)	dose (rad)	nombre d'opérateurs	durée (mois)	dose (rad)	nb. d'opé.
prédécontamination et/ou vidange cellule	4	4 estimé	3	2	2	3
décontamination	3	12,4	8 permanents 48 temporaires	2	1	6 perm.
rééquipement - hublot - divers	 1 4	 1 2,8	 6 7	3	2	4
total	**12**	**20**		**6**	**5**	

Tableau 5

DUREE DES TRAVAUX EN HOMME-JOUR

	CELLULE 3		CELLULE 9 (prévision)
	permanent	temporaire	permanent
prédécontamination et/ou vidange	240		120
décontamination	480	40	240
total	**760**		**360**

Tableau 1