THE THTR-300 COOLANT GAS ACTIVITY, AN INDICATOR OF FUEL PERFORMANCE

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Abstract

During incommissioning and the following 423 full power days (fpd) of THTR operation, the activities of 9 noble fission gas nuclides in the primary coolant have been measured quasi continuously. The radioactive decay times of these nuclides cover a range of more than 3 orders of magnitude. The sources and the mechanisms of the fission gas release can be derived from the dependence of the steady-state release fractions (R/B-values) on the half life times t1/2 of the various nuclides. Thus, the assessment of the fuel performance under operational conditions is primarily based on the routine measurements of the coolant gas activity.

During the first 100 fpd of the THTR-operation the slope s of the curve, R/B-values of noble gas nuclides versus t 1/2 in double logarithmic scales, was measured to be s = o.3. This result is in accordance with the design model, where the dominant release source is given by the manufacture induced uranium contamination of the graphitic matrix material. The relatively low slope-value of o.3 is caused by the superposition of the release fractions from two material components with different diffusion constants.

Within the subsequent operation the coolant gas activity increased by a factor of about two. However, the maximum did not exceed 4 % of the design value of the THTR coolant gas activity. The slope s decreased to 0.2. This change can be explained by the contribution of fission product atoms directly recoiled from the surface of damaged fuel elements into the coolant. In fact, a small fraction of the fuel elements have been mechanically damaged by the frequent insertions of control rods into the pebble bed core under the specific conditions of the THTR incommissioning procedure.

From the measured coolant gas activity data a fraction of coated particle failures less than 8 10-5 due to external forces was evaluated. In-service failures of particles embedded in the matrix can be neglected. This evaluation demonstrates its capability to identify specific fuel failure modes during operation. In this way, the coolant gas activity can be used as an indicator of the fuel performance.

1. Introduction

The radiation protection scheme of any gas cooled reactor requires the regular and reliable measurement of the coolant gas activity during all operational periods. The noble gas activities, predominantly detected in the coolant, determine an important source term for the radiological assessment of the plant during normal and upset conditions. In the case of the THTR-300, the steady-state noble gas activities in the coolant together with the iodine concentrations also constitute the leading sources for the maximum radiological design accident, the total loss of coolant. For the safety analysis of this accident the iodine release from the fuel elements is evaluated by the known relation between iodine and noble gas release.

Apart from the above safety aspects, the measurement of the coolant gas activity provides also a means to asses the actual quality of the fuel elements in the core. In particular, incipient fuel failures can be identified, so that adequate countermeasures can be developed in due time. In this context it is noteworthy, that the commercial guarantee of the in-service quality of the THTR fuel elements (burnup guarantee), which was contracted between the fuel manufacturer and the utility, was based on a certain value of the coolant gas activity (well below the design limit). Thus, the coolant gas activity can be used as an indicator not only of the fuel performance but also of potential financial obligations.

At any rate, the reliable interpretation of the coolant gas activity in relation to the fuel performance necessitates a sound knowledge of the fuel element material properties and the fission gas release mechanisms. This will be given in the following sections in advance of the actual THTR measurements and their evaluation.

2. THTR Fuel Elements

The THTR fuel elements are designed for the high enriched thorium / uranium cycle. The inner fuel zone of the spherical fuel element contains about 38000 $(Th,U)O_2$ kernels coated with pyrocarbon layers (BISO particles). The outer pressure holding layer of the coated particle is derived from methane at deposition temperatures up to 2100°C (HTI-layer). This extreme temperature causes a relatively high uranium contamination in the outer HTI layer. During the final heat treatment of the integral fuel element at 1950°C part of this uranium migrates into the surrounding matrix causing a finely dispersed uranium contamination in the matrix material. This contamination is the prime source for the fission gas and iodine release from the fuel elements during normal and accident conditions of the THTR. The effect of potential manufacture induced coating defects is negligible compared with the contamination.

The frequency plot in Fig. 1 summarizes the quality control data for the uranium contamination of the fuel elements for the initial THTR core (identical product specification and manufacture for initial core and reload fuel elements). The mean value of this most important property amounts to $3.0 \cdot 10^{-4}$ with a standard deviation of $7 \cdot 10^{-5}$ for the single values (3 measurements per batch, maximum batch size : 2000 fuel elements).

The key design data of the THTR fuel elements are given in Tab. 1.

During operation a small fraction of the coated particles is expected to fail. Based on irradiation testing of THTR fuel elements, the in-service failure fraction at the end of the residence time in the core (fast neutron fluence: $4.5 \cdot 10^{21}$ cm^{-2}, E > 0,1 MeV, burnup: 11.5 % fima) is estimated to be $2 \cdot 10^{-3}$. These defectives contribute only < 10 % to the total fission gas release from the THTR core

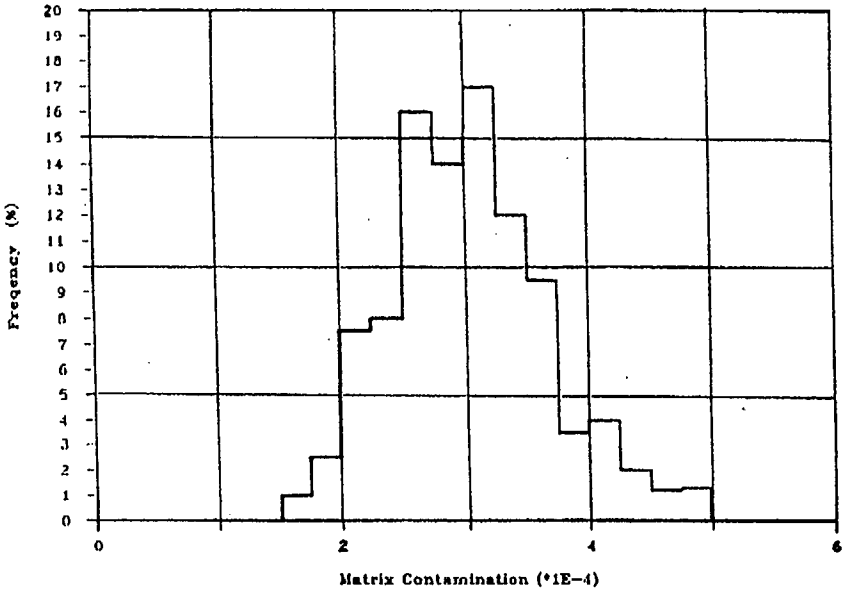

Fig. 1 Distribution of the uranium contamination data for the fuel elements of the THTR initial core (n = 770)

(exception: long-lived Kr 85). Nevertheless, the fission gas release mechanisms must be known equally well for both, the matrix contamination and the failed particles, if unexpected changes of the coolant gas activity are to be explained.

3. Fission Gas Release Models

3.1 Uranium Contamination of Matrix Material

The steady-state fission gas release from THTR fuel elements is calculated with a model which was derived from in-pile measured xenon and krypton release data of a series of irradiation experiments with THTR prototype fuel elements.

Table 1 Design Data of Spherical THTR Fuel Elements

Fuel Element		
Matrix Material		A3-3
Outer Diameter	mm	60
Diameter of Fuel Zone	mm	46
Thorium Loading	g/FE	10.2
Uranium Loading	g/FE	0.96
U-235 Enrichment	%	93
Free Uranium Fraction a)	-	$3.0 \cdot 10^{-4}$
Coated Fuel Particle		
Type		HTI - BISO
Fuel Composition		(Th, U) O_2
Kernel Density	g/cm^3	9.9
Dimensions		
Kernel Diameter	µm	400
Porous PyC	µm	80
Sealing PyC	µm	30
Outer PyC	µm	80
Uranium Contamination of Outer PyC	-	$1.0 \cdot 10^{-3}$

a) weight of uranium outside intact coated particles divided by total uranium loading of the fuel element

This model was successfully submitted to the THTR licencing procedure. Details of the model are given in /1/, so that only a qualitative survey is needed here.

The graphitic matrix material of the fuel element is treated as a two-component system. Component 1 may be attributed to the graphite grains of the raw material, and component 2 to the amorphous, non graphitized binder coke between the grains.

The primary fission products are distributed homogeneously in both components by direct recoil. The gas atoms diffuse from the recoil sites to the open porosity of the fuel element. This volume diffusion in both components is described with Booth's "equivalent sphere" model yielding equation (1) for the fractional release of a contamination-born fission gas nuclide from component m:

$$(1) \qquad R_m / B = 3 K_m F_m \cdot (\coth K_m^{-1} - K_m)$$

$$\text{where } K_m = \sqrt{D_m / (r_m^2 \lambda)}$$

- R_m - release rate from component m into the open porosity
- B - contamination induced birth rate in the fuel element
- F_m - fraction of recoil sites in component m
- D_m - diffusion constant of xenon or krypton in component m
- r_m - equivalent sphere radius of component m
- λ - decay constant of a given xenon or krypton isotope.

The two diffusion processes working in parallel in both components are followed by gas phase transport trough the open porosity of the fuel element into the coolant. This process is also described by equation (1) where F_m is given by the sum of the R_m/B-values of both matrix components and r_m equals the radius of the fuel element.

The application of this model with optimized material parameters to the xenon release data measured in THTR irradiation experiments (T = 700°C, p = 1 bar) is shown in Fig. 2. The superposition of the contributions from both components leads to a mean slope of the R/B versus λ-curve of -0.33 in double logarithmic scales.

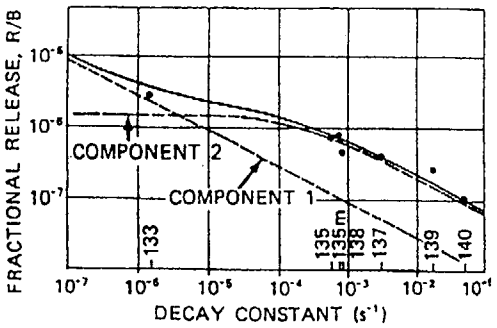

Fig. 2 Calculated and measured xenon release from THTR fuel elements in irradiation experiments at 700 °C, 1 bar (uranium contamination: $3.0 \cdot 10^{-4}$)

The model /1/ was improved by taking into account the fraction of direct recoil atoms which are stopped in the open porosity of the fuel element. This effects only short-lived isotopes at high pressures. Under THTR conditions (700°C, 38 bar) the release of Xe 137 e.g. is increased by about 20 %.

The flow pattern of this enlarged model for the release of contamination induced noble gases is shown in Fig. 3a.

3.2 Failed Fuel Particles

The gas release model for failed fuel particles embedded in the matrix was developed on the base of an irradiation experiment with known contents of artificial defect (Th, U)O_2-particles (laser drilled coatings). The dependence of the in-pile measured gas release data on the radioactive decay constant and on temperature (780°C - 1100°C) is described by a quasi three-component system consisting of

- kernel grains,
- "grains" of the porous buffer layer,
- gas filled, open pores in the kernel and in the porous buffer layer.

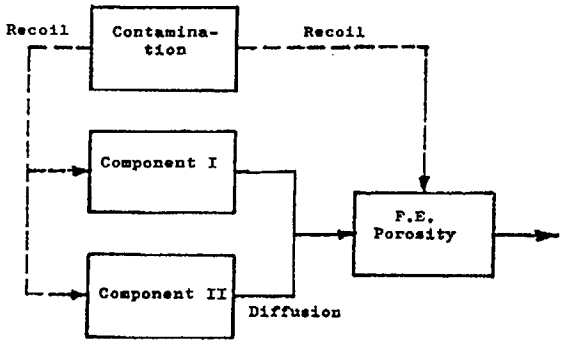

Fig. 3a Gas release from uranium contamination of matrix material

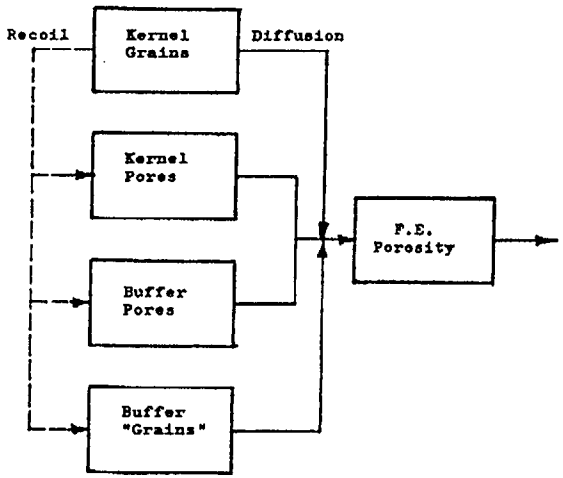

Fig. 3b Gas release from failed fuel particles

The birth rates of primary fission products in these components are calculated with the known relations for the recoil stopping ranges (Bragg-Kleemann Rule). It is assumed that the open porosity of the kernel and the buffer layer is filled with helium at system pressure. In addition to the direct recoil fraction from the kernel into the buffer layer (2.8 % for Xe, 3.7 % for Kr from 400 µm kernels) the manufacture induced uranium contamination of this layer (3 %) is taken into account.

The fractional noble gas releases from the two grain components are treated in the same way as for the contamination-born release using equation (1). The retention capabilities of the open porosity of the kernel and the porous layer as well as the eventual delay of the transport through the coating defects (e.g. hairline cracks) are neglected. The release from the failed particles is followed by the gas phase transport through the open porosity of the fuel elements.

The flow scheme of this release model is depicted in Fig. 3b. Fig. 4 shows the break-down of the release fractions from failed particles (400 µm (Th, U) O_2 kernels) calculated with the above model using optimized material parameters for the krypton isotopes at typical THTR conditions (700°C, 38 bar). At the relatively low temperature of 700°C the diffusional release from the kernel grains (activation energy of the diffusion constant: 310 kJ/mole) is negligible. The kernel release becomes significant only at temperatures above 1000°C.

In preparation of the following discussion of the THTR coolant gas activity, in Fig. 5 the mean slopes of the fractional release curves versus the radioactive half lives $t_{1/2}$ of the various nuclides (from Kr 89 with $t_{1/2}$ = 190s up to Xe 133 with $t_{1/2}$ = 5.3d) in double logarithmic scales are plotted against temperature. It is distinguished be-

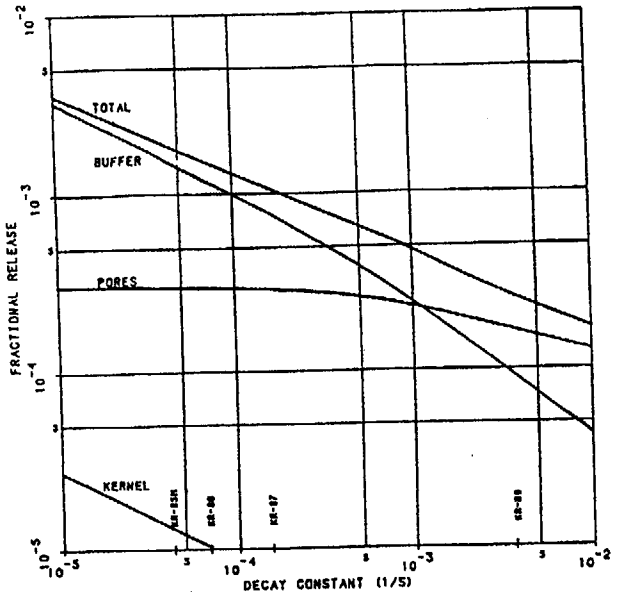

Fig. 4 Calculated krypton release from failed particles in THTR fuel elements at 700 °C, 38 bar

tween pressures at 1 bar (full lines) and 38 bar (broken lines). Both models lead to rather similiar mean log R/B-log $t_{1/2}$-slopes. Neither temperatures, nor pressures influence these slopes significantly.

4. THTR-300 Coolant Gas Activity

4.1 Measurements

The coolant gas activity of the THTR was continuously monitored by β-counting devices. This measurement enables immediate detection of relative changes, but at relatively high uncertainties of the absolute values. Therefore, the

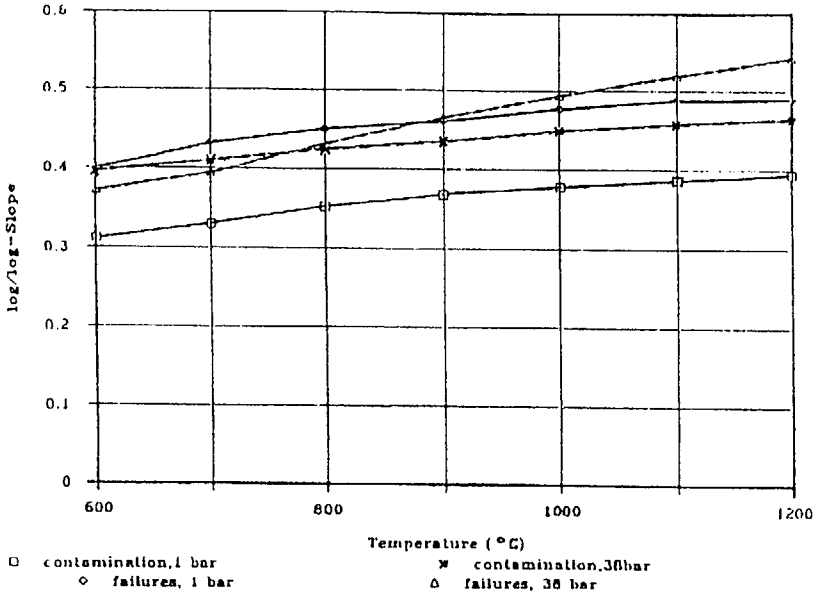

Fig. 5 Slope of log R/B versus log $t_{1/2}$ of the release models for contamination and failed particles (averaged for all nuclides between $t_{1/2}$ = 190 s and $t_{1/2}$ = 6.3 d)

detailed evaluation was entirely based on the γ-spectrometric analysis of isolated gas samples. At regular intervals the activities of 4 krypton and 5 xenon isotopes were measured at power levels from 10 % to 100 % of rated power.

The sum of these activities, covering about 90 % of the total coolant gas activity, is drawn in Fig. 6 against the THTR operation time of 423 fpd (full power days at 762 MW_{th}). For comparison sake, the measurements are related to the actual thermal reactor power (Bq/MW) and to the rated mass flow through the gas purification plant (0.15 kg/s). The load factor of the core and extended shutdown events (marked by arrows) are also shown in Fig. 6.

From 70 to 150 fpd of operation, the activity increased by about a factor 3. From 150 to 300 fpd only small changes occurred. After 300 fpd, a continuous long-term decrease of the activity was observed.

During the rise of the total gas activity the spectrum of the different noble gas nuclides in the coolant changed significantly. The short-lived isotopes increased much steeper than the isotopes with long half-lives. This tendency is demonstrated in Fig. 7 showing the measured fractional releases in dependence of the half-lives at 90 and 150 fpd together with the original calculation (contamination release model). The measured log R/B-log-$t_{1/2}$-slope falls from 0.30 at 90 fpd to 0.15 at 150 fpd. All evaluated slope-values are plotted in Fig. 8 against the operation time. The somewhat arbitrarily drawn average curve in Fig. 8 is obviously (negatively) correlated with the development of the coolant gas activity as shown in Fig. 6.

Any interpretation of the changing coolant gas activity in the THTR can only be regarded satisfactory, if the above tendencies of the slopes are explained consistently.

4.2 Evaluation

From the calculated model results as shown in Fig. 5 it is concluded, that increased temperatures and failed particles embedded in the matrix cannot explain the observed slope changes. This conclusion was confirmed by calculations with revised temperature distributions and substantial failed particle fractions. Furthermore, a sensitivity study of the material parameters of the contamination related release model did not provide a realistic explanation. The measured slope decrease could only be reproduced by complete neglection of the gas phase transport in the open porosity and an increase of the diffusion constant in the amorphous component by two orders

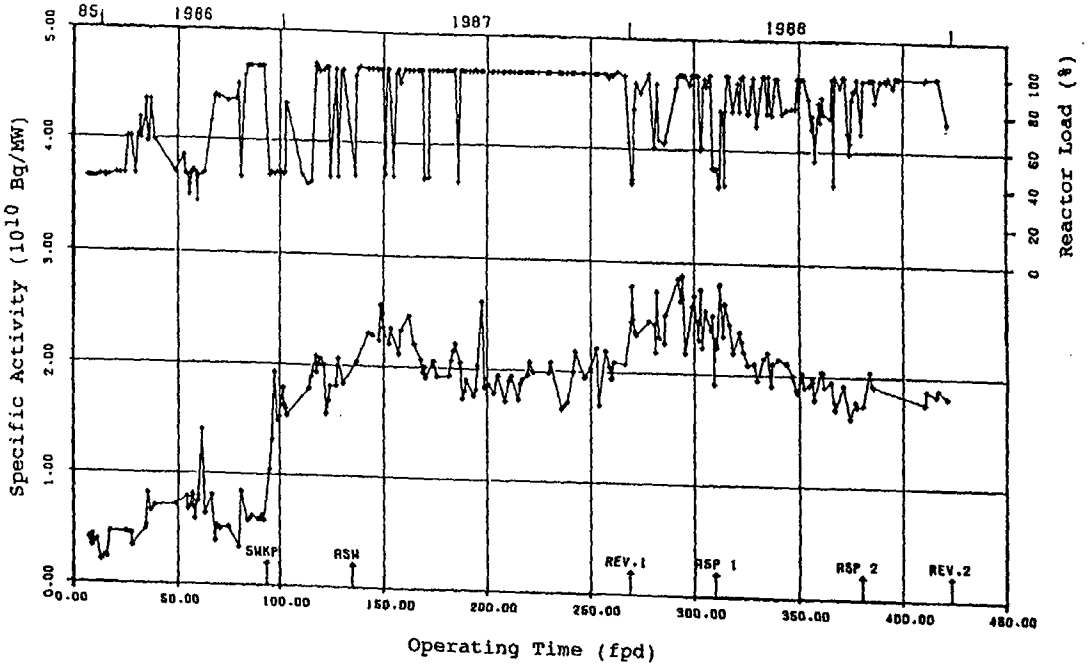

Fig. 6 THTR coolant gas activity
(sum of 9 noble gas nuclides)

of magnitude. However, there is no physical base for this fictitious parameter change. Thus, a new release mechanism had to be postulated.

The background of this model development was the THTR experience that some fuel elements have been damaged by mechanical forces from the control rods inserted into the pebble bed core very frequently under extremely adverse conditions (high sphere loading density in the core) during incommissioning the THTR. The damaged fuel elements (definition: local diameter ≤ 57 mm) are sorted out from recirculation by a special size separator at the core exit. In most of the damaged spheres the fracture surfaces are restricted to the fuel free outer shell. However, some of the damaged fuel elements have fracture surfaces right through the inner fuel zone.

It is assumed that a fraction of the coated particles situated within the fracture surfaces are also mechanically damaged, so that part of their kernel material is exposed

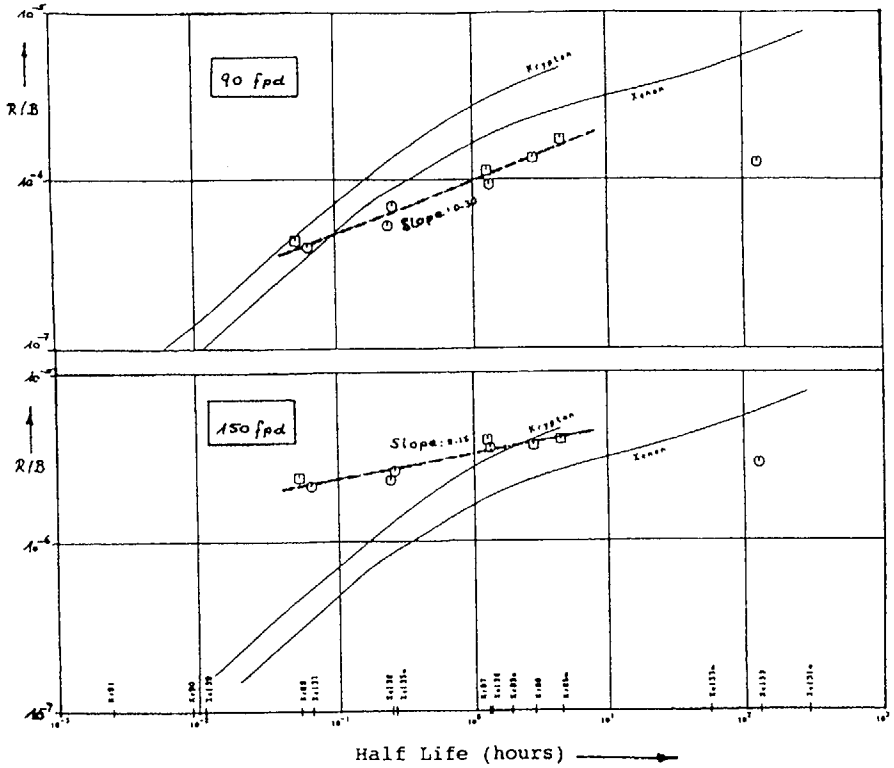

Fig. 7 Measured and calculated release fractions and log R/B-log $t_{1/2}$ -slopes at 90 fpd and 150 fpd

to the coolant. Primary fission products are recoiled into the coolant from these kernels without delay.

Because of the small remaining recoil range in the coolant of about 3 mm (at 38 bar helium), practically all fission products recoiled from the exposed kernel surfaces are stopped in the coolant gas. Thus, an additive release contribution is obtained which is independent of the radioactive half life.

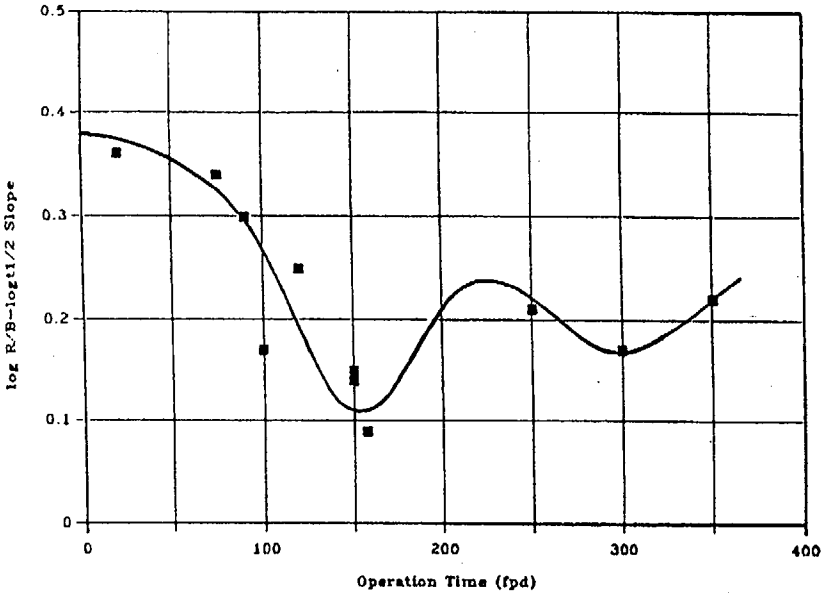

Fig. 8 Development of the log R/B-log $t_{1/2}$ slope during THTR operation

By comparison with the measured activities at 170 fpd a fractional release of $1.4 \cdot 10^{-6}$ from the total core was evaluated for all gas nuclides. Fig. 9 shows the good agreement between measurement and the enlarged model. The above release fraction of $1.4 \cdot 10^{-6}$ leads to an upper estimation of the fraction of exposed failed particles of $8 \cdot 10^{-5}$ in the core, respectively $5 \cdot 10^{-3}$ of the particles in the damaged fuel elements.

The given additive release model was substantiated by the evaluation of the fraction of damaged fuel elements in the circulating flow of spheres at the core exit as measured after the size separator. Fig. 10 shows the development of this fraction in dependence of the accumulated number of circulated spheres. The measured fraction of damaged fuel elements at the

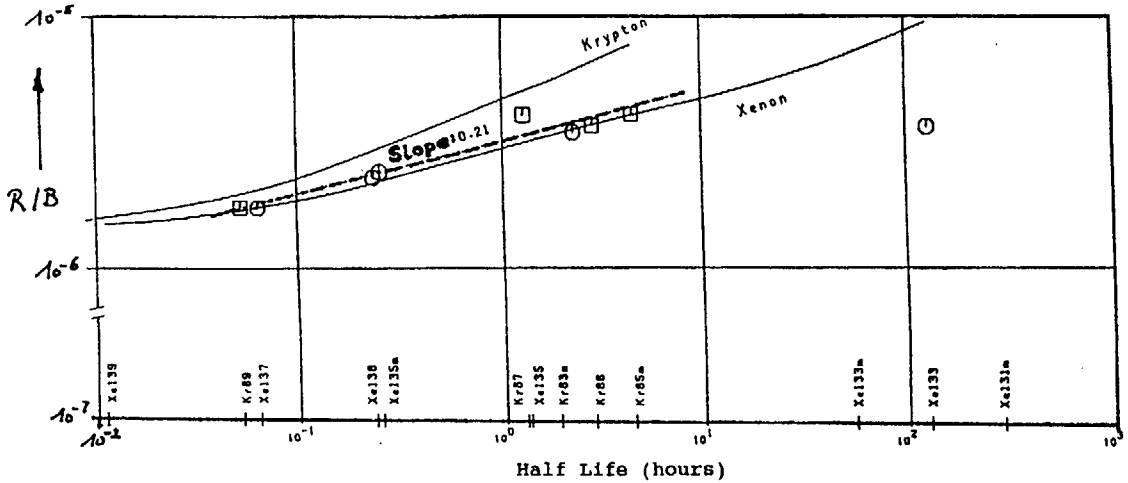

Fig. 9 Measured and calculated release fractions at 170 fpd (enlarged model with constant $1.4 \cdot 10^{-6}$ recoil release)

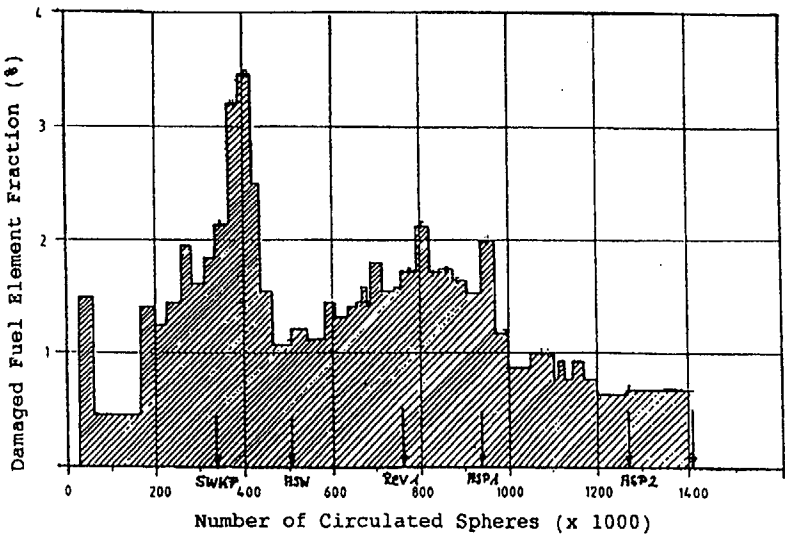

Fig. 10 Fraction of mechanically damaged fuel elements at core exit (measured after size separator)

core exit is positively correlated with the measured coolant gas activity (marked shutdown events in Fig. 6 and in Fig. 10 for orientation). The steepest rise of the activity in Fig. 6 coincides with the maximum damaged fuel element fraction (between SWKP and ASW) in Fig. 10. The long-term decline of the activity is also beginning at the same time as the steady decline of the damage fraction (between Rev. 1 and ASP 1). This correlation proves the presumption that exposed fuel kernels from mechanically damaged fuel elements caused the changes of the THTR coolant gas activity.

5. Conclusions

The model guided analysis of the THTR coolant gas activity enables a comprehensive understanding of the actual status of the fuel element performance. The unexpected activity changes are attributed to defective particles in mechanically damaged

fuel elements which are exposed to the coolant. Thus, new release mechanisms hitherto not investigated in preceding experiments can be revealed by operational measurements.

It should be pointed out,that - inspite of the activity increase - the measured sum activity in the coolant agreed with the licenced expected value within a bandwith of ± 20 % (since 100 fpd). The measured activity amounted only to 4 % of the licenced design value.

Reference

/1/ K. Röllig: Nuclear Technology 35 (1977) p. 516