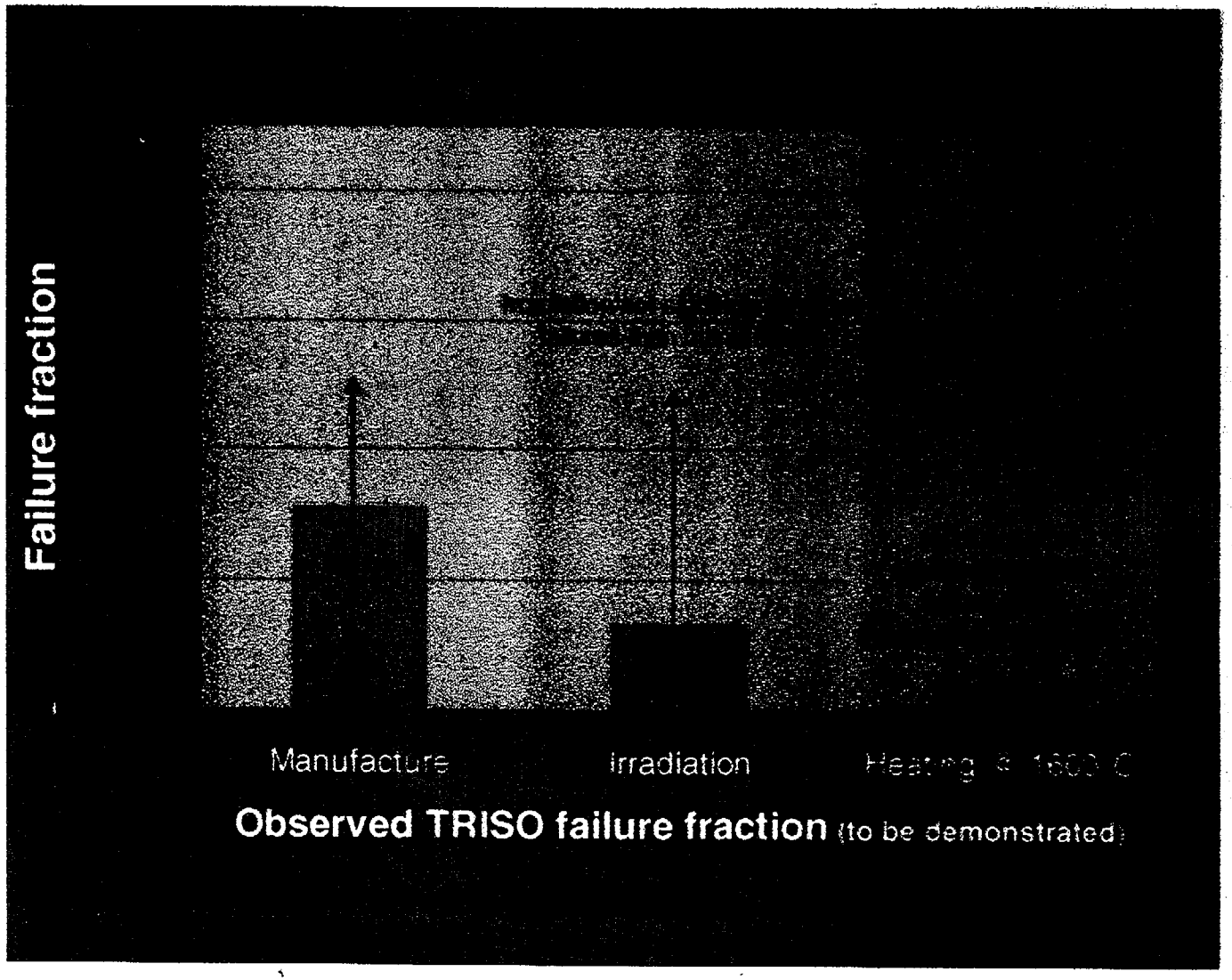
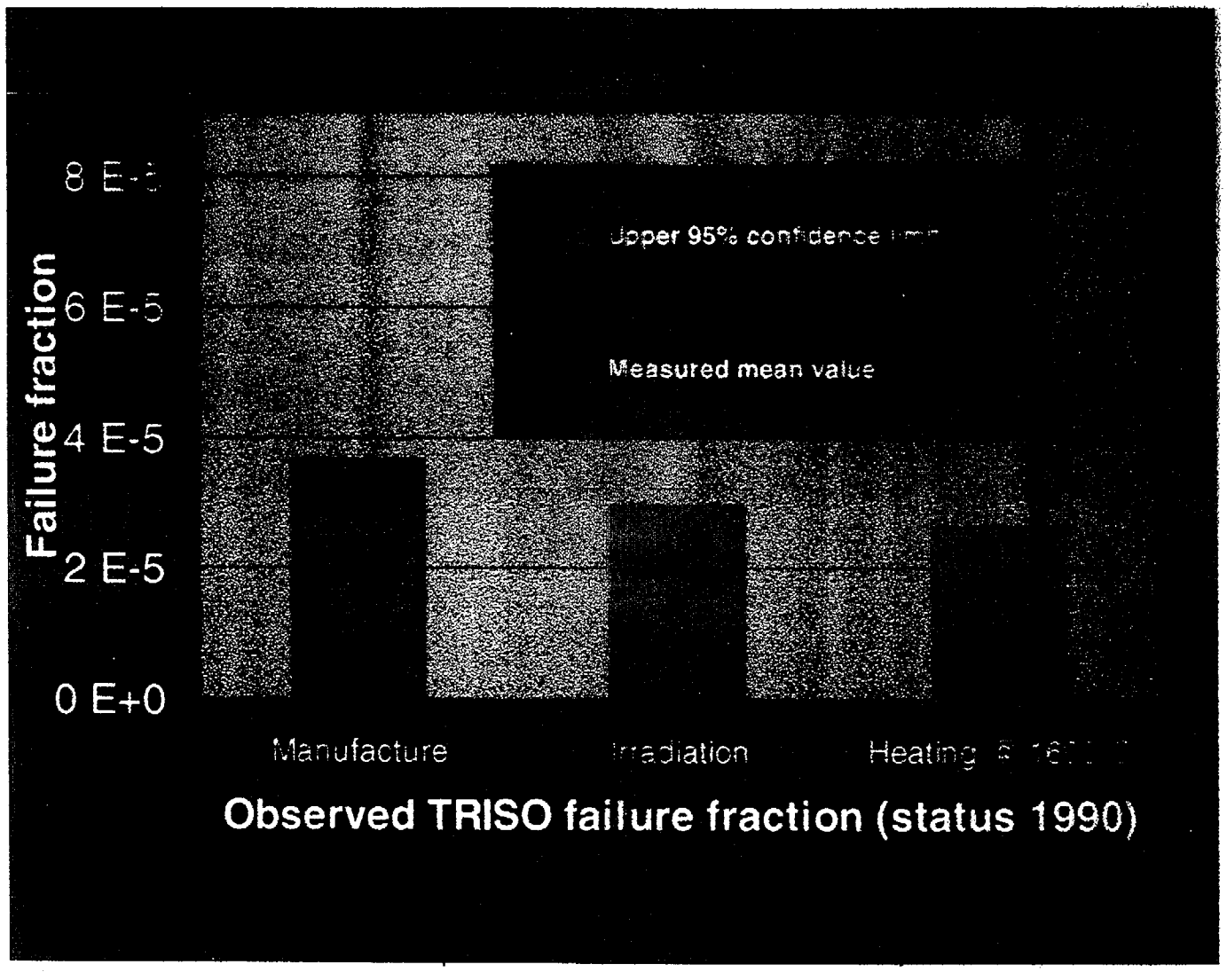
Target philosophy



Desirable properties have been achieved and have been demonstrated
(here HEU and LEU TRISO up to 1990)



Observed TRISO failure fraction (status 1990)

HTR fuel: main criteria quantified ...

Manufacture

burn-leach on particles $< 1 \times 10^{-6}$ *

burn-leach on fuel body $< 6 \times 10^{-5}$

Irradiation

in-pile R/B $\ll 1 \times 10^{-6}$ *

PIE shows $F(^{137}Cs) < 2 \times 10^{-5}$

... and ... $F(^{110m}Ag) < 2 \times 10^{-3}$

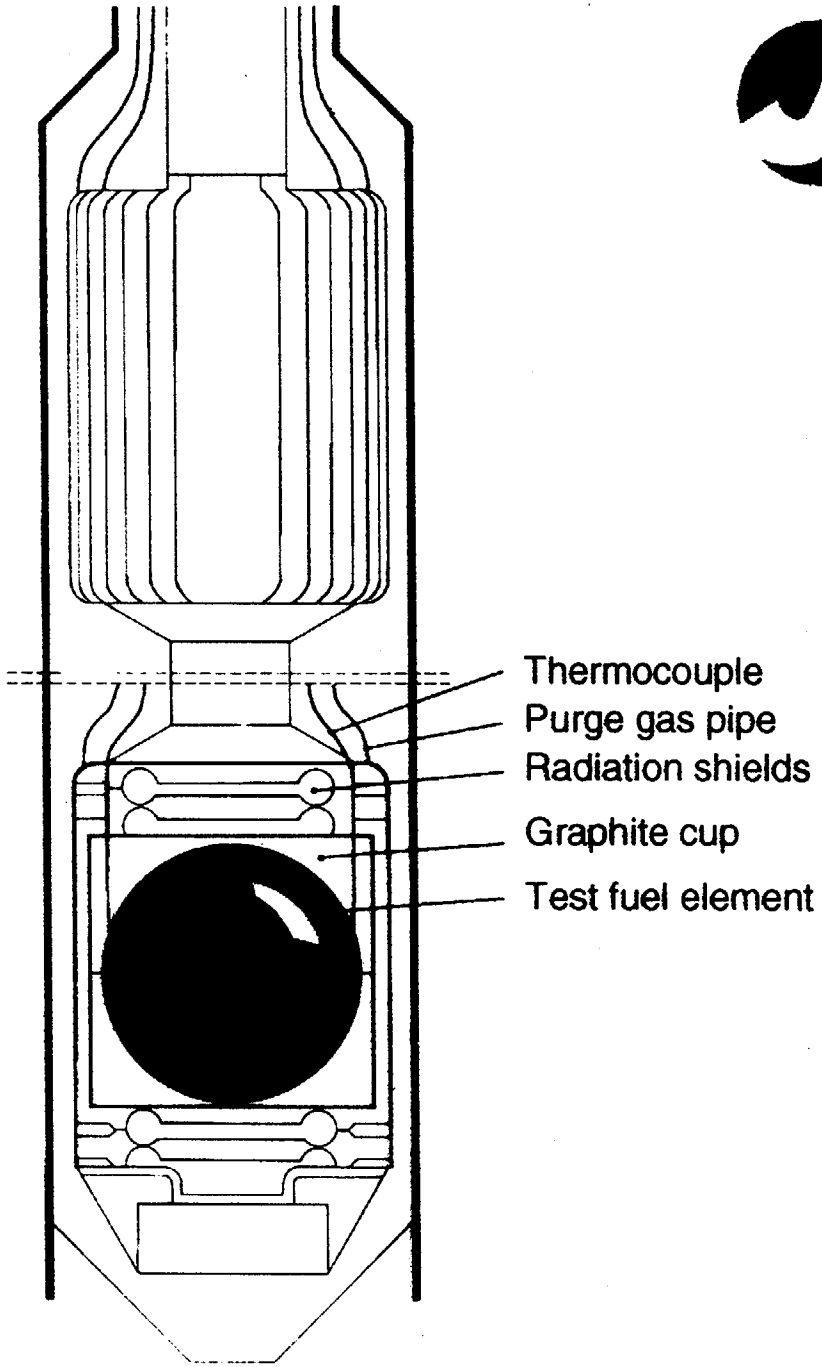
Heating

shows $F(^{85}Kr) < 2 \times 10^{-6}$ *

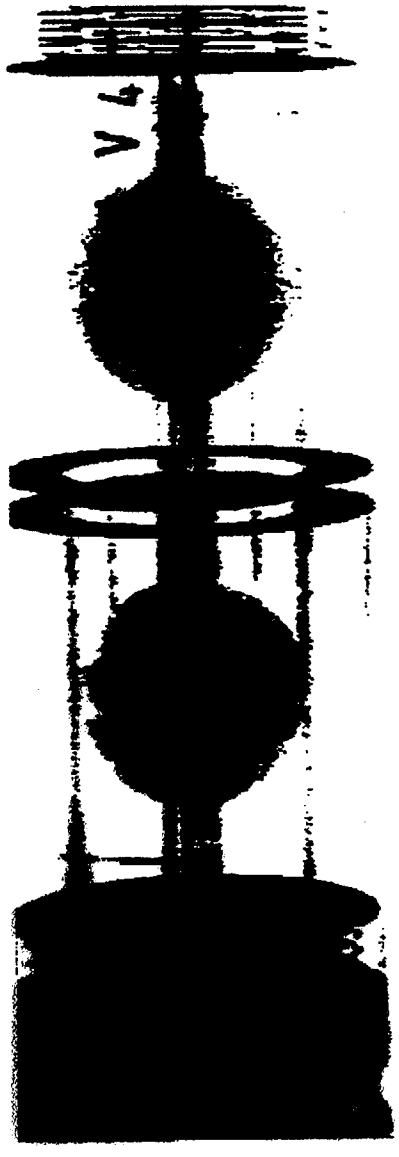
and $F(^{137}Cs) < 2 \times 10^{-4}$

* Necessary, but not sufficient

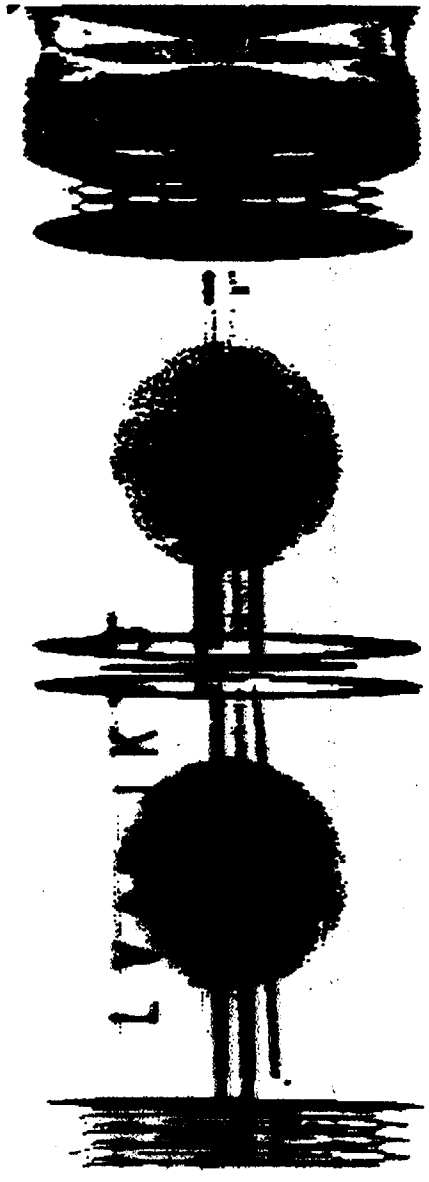
Every fuel element in an irradiation experiment is contained in an isolated cell. Temperatures are closely controlled by adjusting the He/Ne ratio in the purge gas which also allows the on-line determination of the fission gas release rate.



Capsule 1



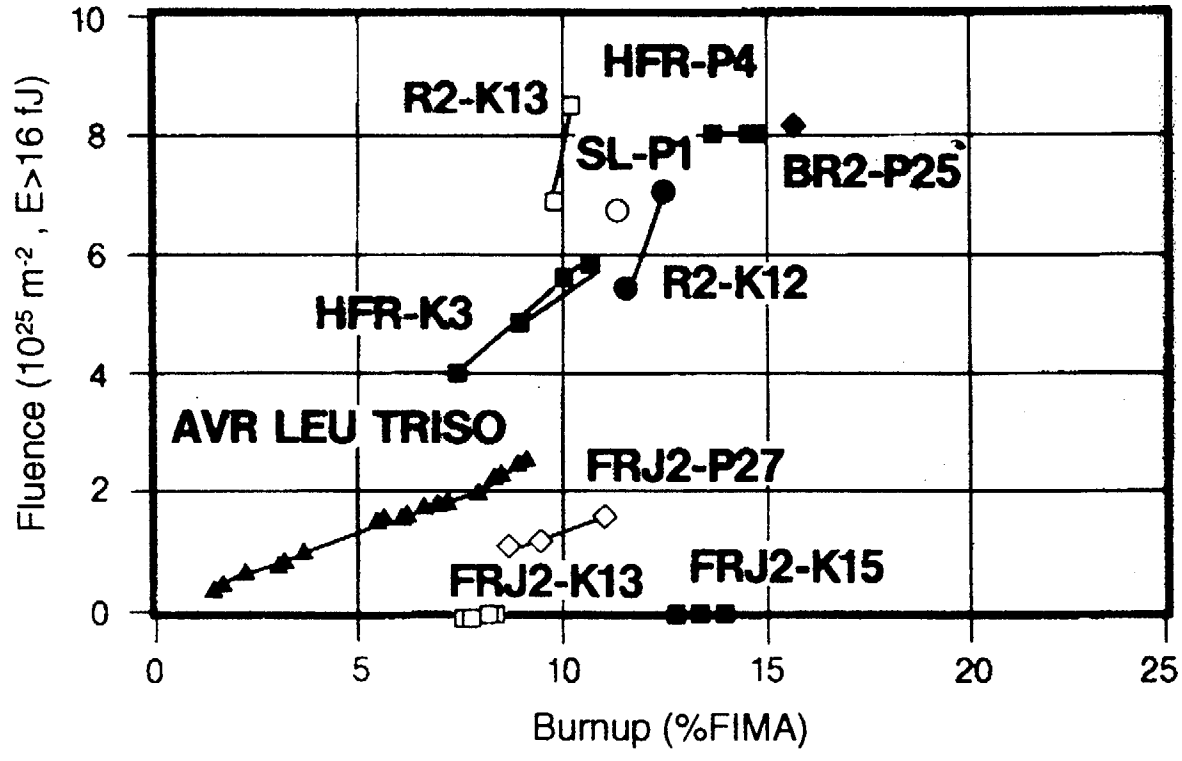
Capsule 2



X-ray photograph from the assembled capsules 1 and 2 for experiment FRJ2-K13

German Irradiation Experiments with HTR Fuel

RDD program	Old LEU 1972-1976	HEU Program for PNP and HHT 1977-1981			HEU Program 1982-1986
Coated Particle	UO_2 TRISO UO_2 BISO	Variant 1 $(Th,U)O_2$ BISO	Variant 2 $(Th,U)O_2$ Triso	Variant 3 UC_2 Triso ThO_2 Triso	UO_2 TRISO
Test Goal					
Particle Performance	HFR-M5 DR-S6	BR2-P24	BR2-P25	BR2-P26	HFR-P4 SL-1
FP Transport in Intact Particles	DR-S4	FRJ2-P22	FRJ2-P23	FRJ2-P24	FRJ2-P27
Release from Kernel	-	FRJ2-P25	FRJ2-P25	FRJ2-P26	FRJ2-P28
Chemical Effects	FRJ2-P16	-	-	HFR-P3	HFR-P5
Fuel Element Tests					
Fuel Element Performance	DR-K5	HFR-K1	R2-K12 R2-K13	R2-K12	HFR-K3
FE fission product transp.	-	-	FRJ1-K11	FRJ2-K10	FRJ2-K13 FRJ2-K15
Large Scale Demonstration	AVR 6	AVR 14	AVR 15	AVR 13	AVR 19 AVR 21 AVR 21-2
Proof Tests					HFR-K5 HFR-K6

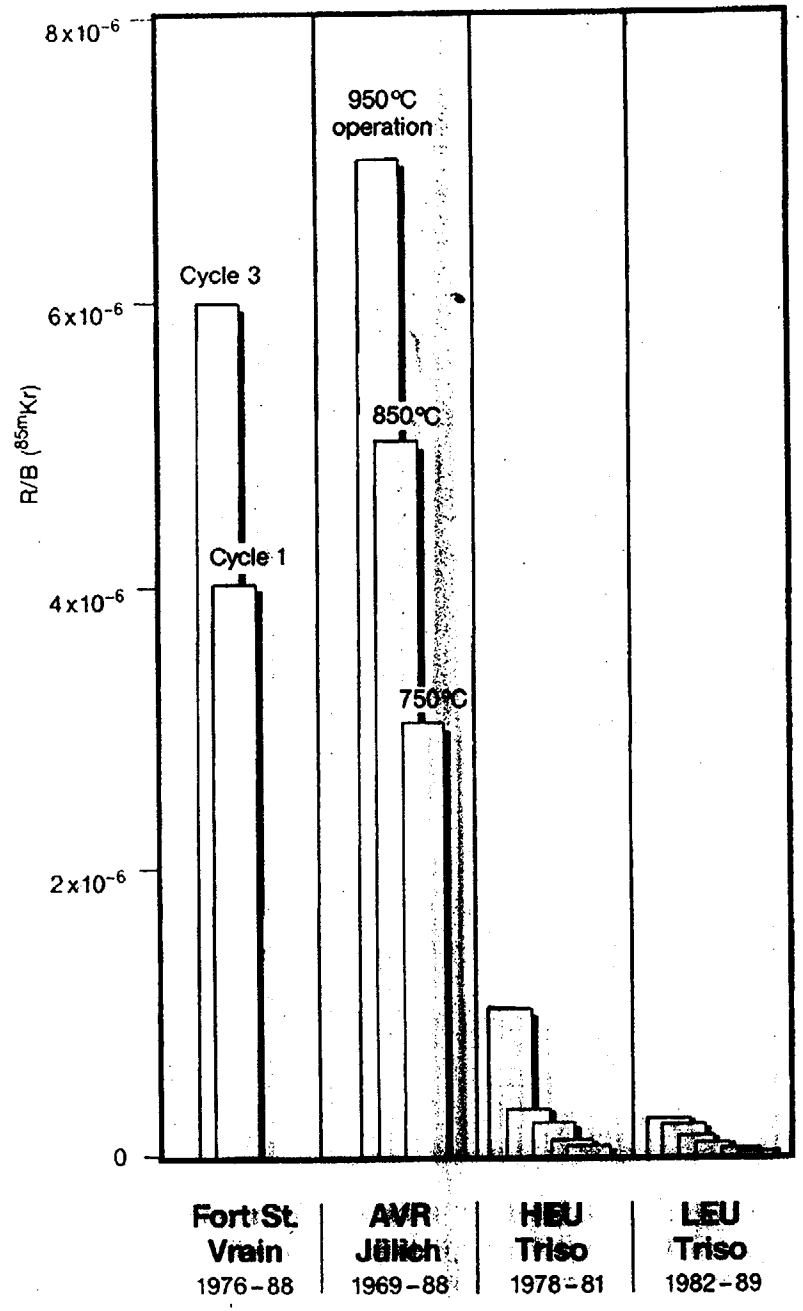


The main parameters in fuel irradiation testing are heavy metal burnup and accumulated fluence of fast neutrons. Irradiation temperature, perhaps the most important parameter of all, is usually around 1000° in the tests shown here.

Criteria for irradiation testing in order of relevance

- Temperature
- Burnup
- Fluence
- Power/ temperature gradients
- Transients
- Real time

The level of fission gas is determined by the uncovered uranium in the fuel. Here, the release rate over birth rate of ^{85m}Kr is shown for Fort St. Vrain, AVR and irradiation tests with modern TRISO fuels.



Source terms

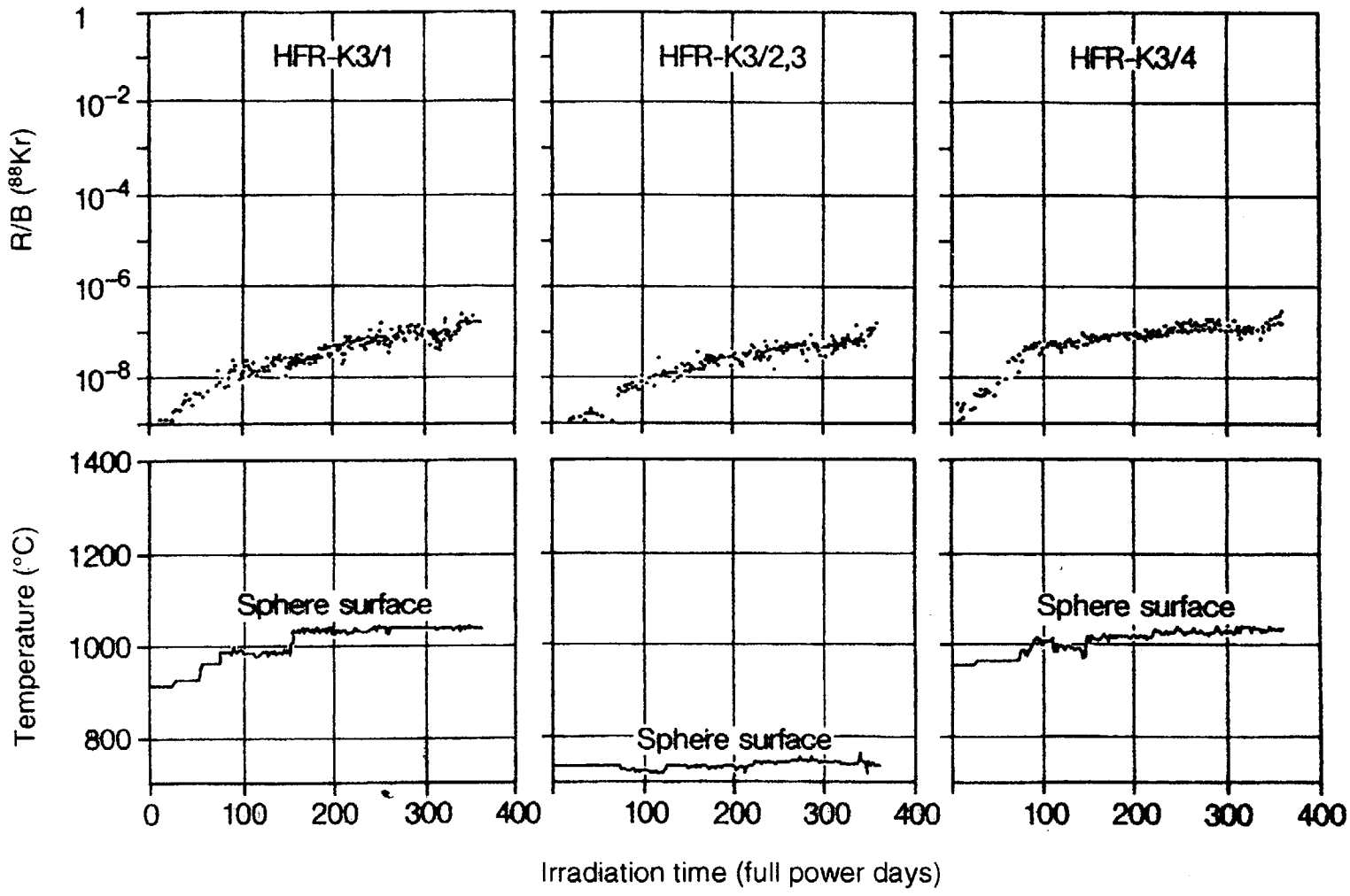
for fission products into the primary circuit of an HTR are:

(i) heavy-metal contamination;
(ii) particle defect and/ or failure;
(iii) release from intact particles.

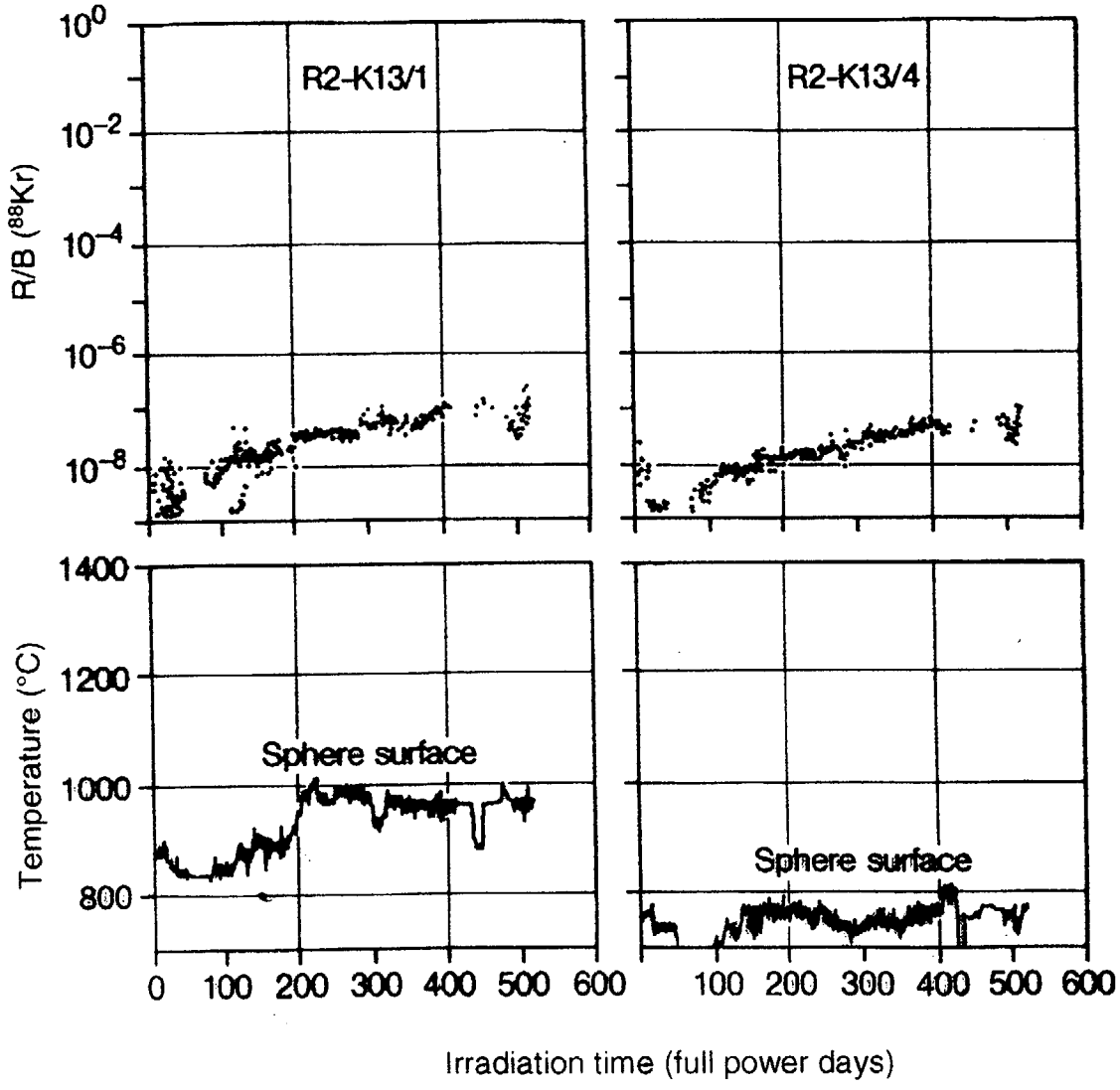
Sequence of release is

^{110m}Ag, ^{137}Cs, ^{134}Cs, ^{85}Kr, ^{90}Sr, ^{106}Ru, ^{95}Zr

Observations: / Measurement Technologies	Contribution to Fission Product Source Terms			
	Free heavy metal fraction	Φ broken particle fraction	Φ' defect SiC/ weak particle fraction	Diffusive release contribution
Manufacture	Acid leach, weak irradiation, TRIGA furnace		Burn-leach hot chlorination	—
Irradiation: in-pile	R/B (Kr, Xe isotopes)		—	—
Irradiation: PIE	cold gas release ?		F(Cs,...), hot Cl	F(Ag, ...)
Accident condition testing	F(Cs)	F(Kr)	F(Cs, Sr,..)	F(Ag) F(Kr) delayed by diffusion through PyC

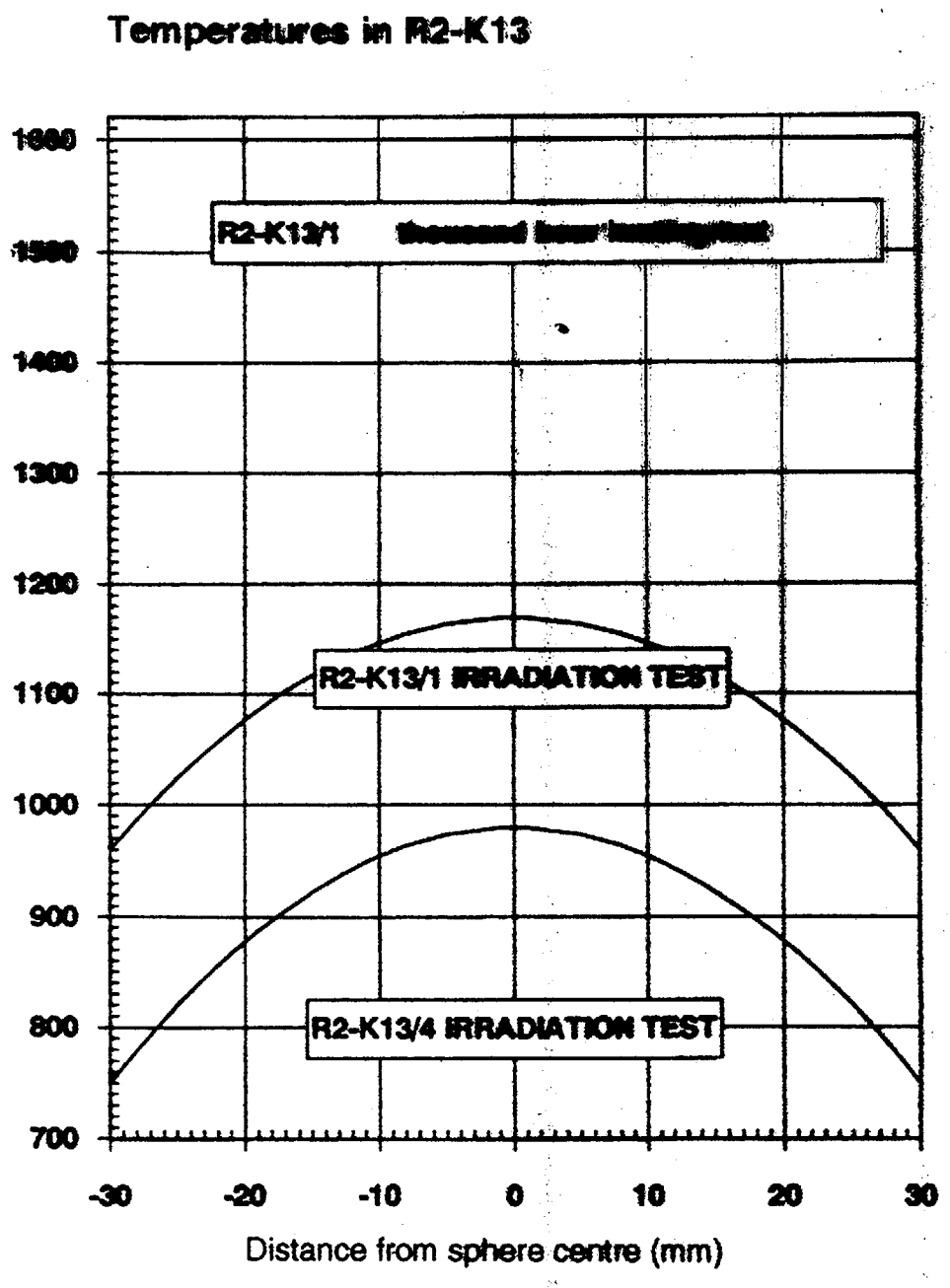


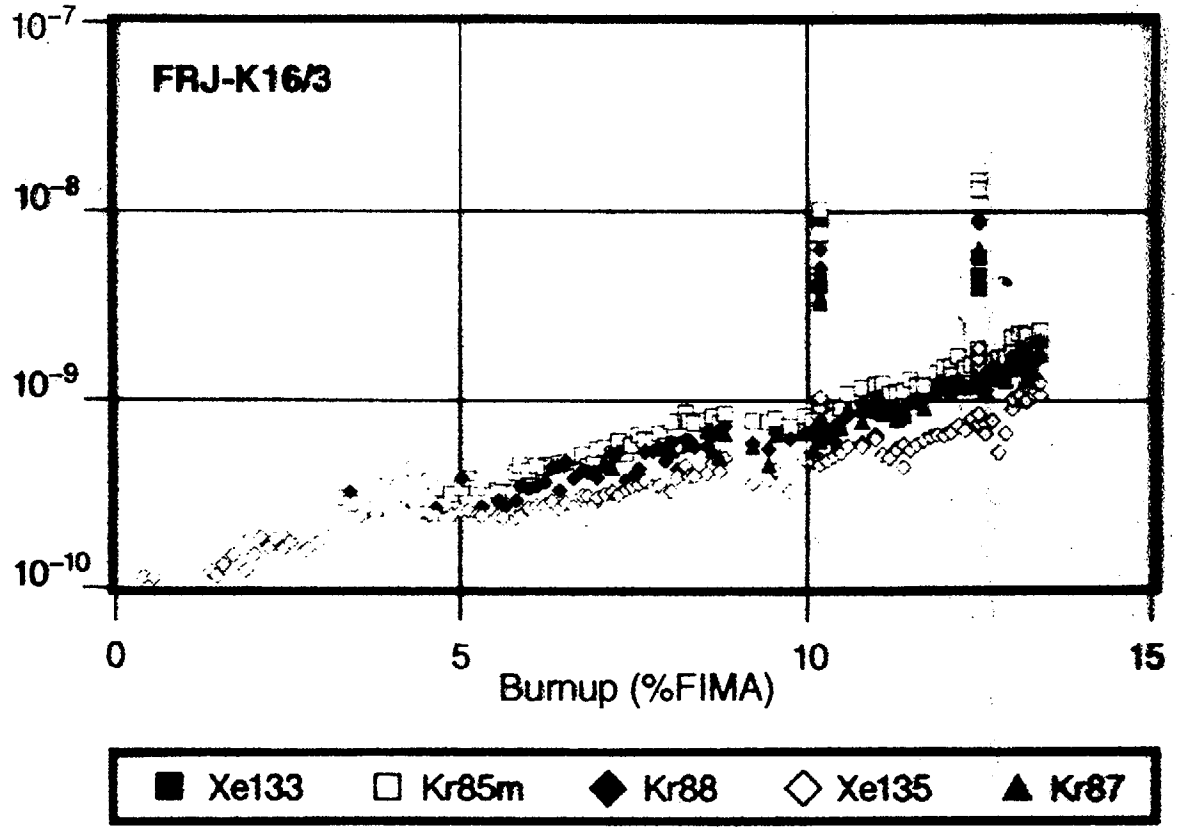
Gas release and temperature during irradiation of experiment HFR-K3



Gas release and surface temperatures in experiment R2-K13

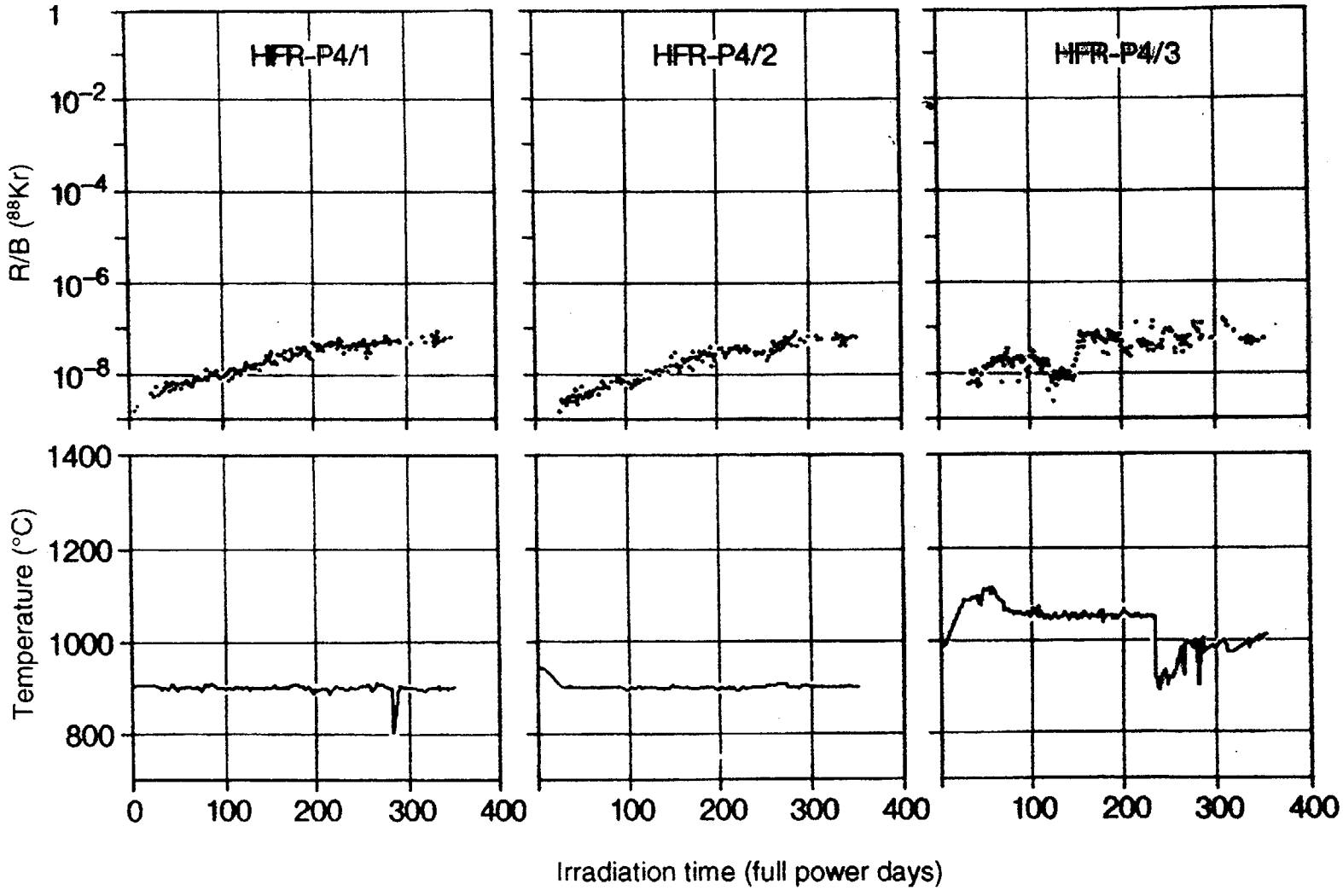
Radial distribution of irradiation temperatures (time average) of R2-K13 test and during 1600 °C heating test



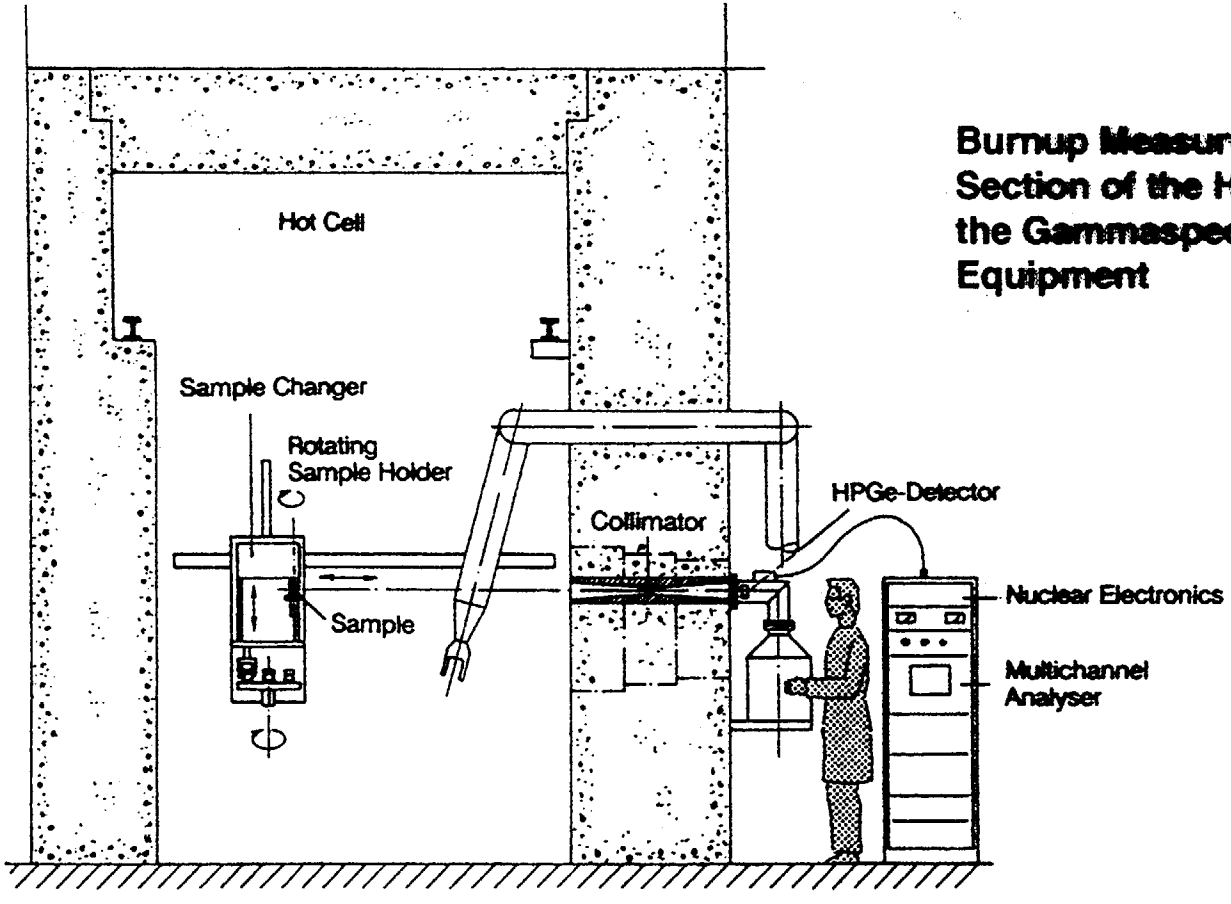


The release rate of short-lived fission gases, R/B, as a function of burnup in an ongoing irradiation experiment in the Jülich DIDO reactor (FRJ2-K15, status from June 1989). The spikes in R/B have been measured during +200°C temperature transients. The slow increase in R/B is an artifact of the birthrate calculation for 16.7% enriched uranium, while the actual release is from uranium contamination of natural enrichment.

Irradiation to near 15 % FIMA



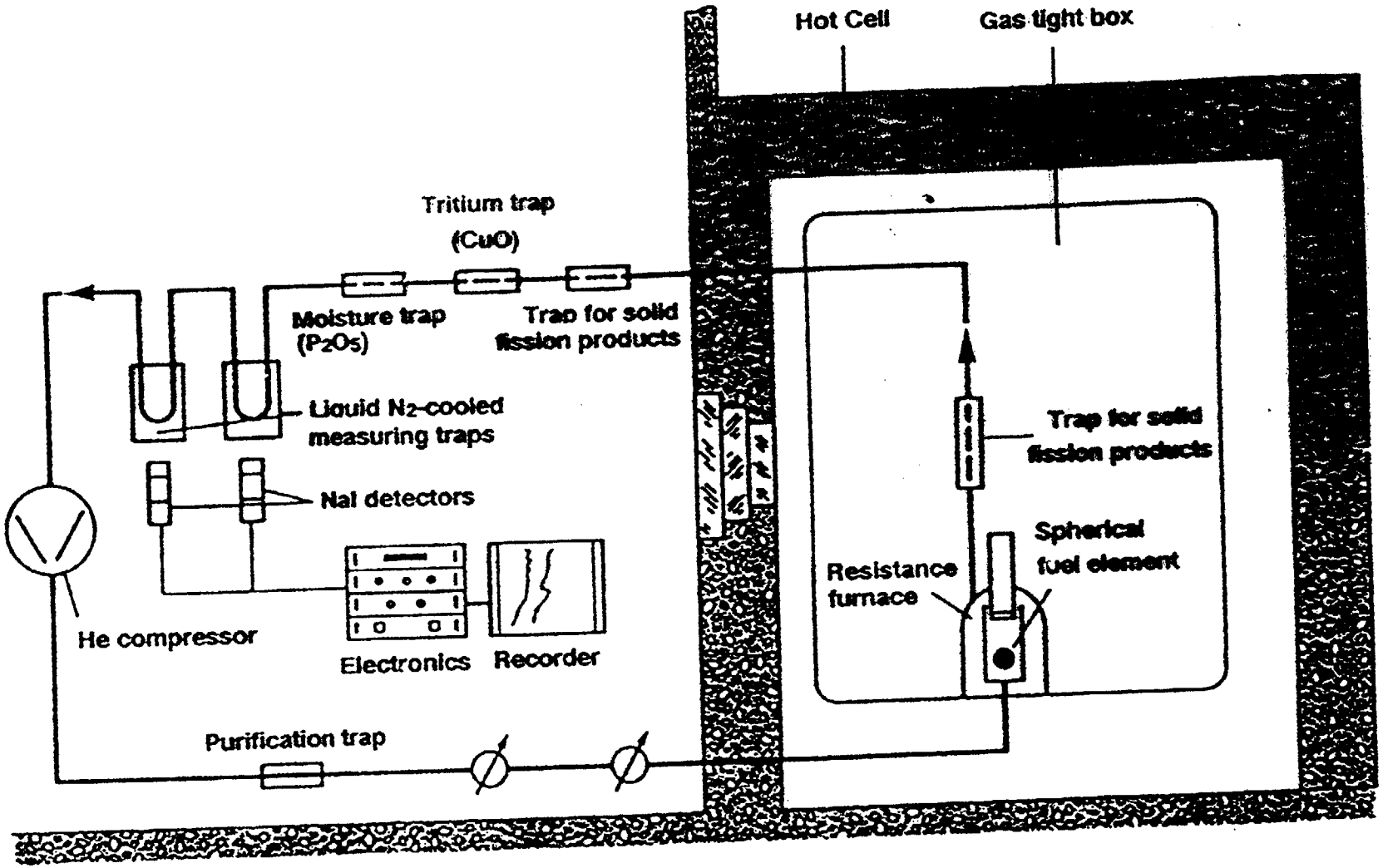
Processing of fuel elements in the Hot Cells, e.g. during gamma spectrometry for burnup determination



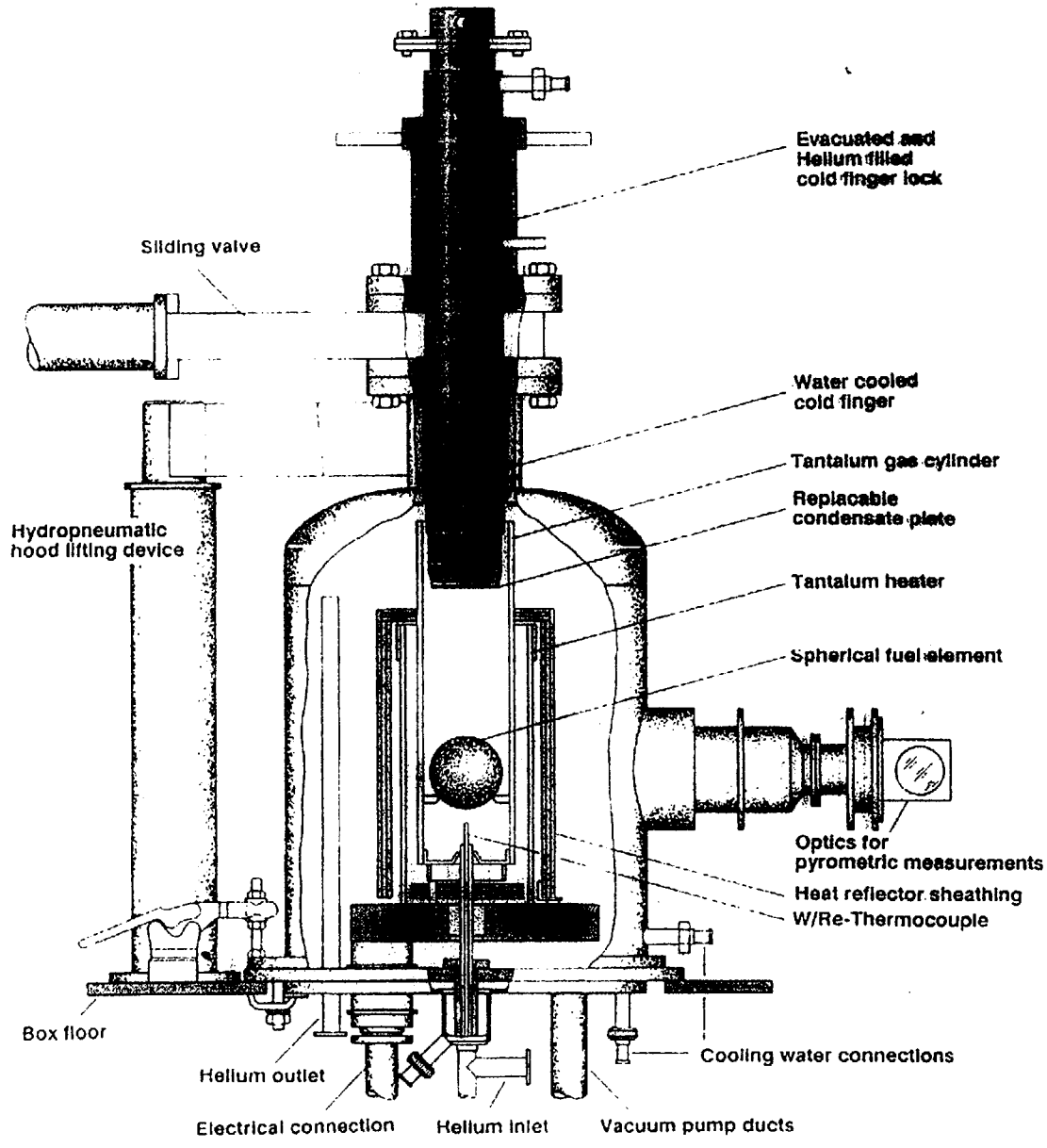
Burnup Measurements Cross-Section of the Hot Cell with the Gammaspectrometric Equipment

Heatup Testing

- Test rigs
- Near complete release up to 2500°C
- Cold finger KÜFA 1600-1800°C
- Measurement technology for fuel criteria

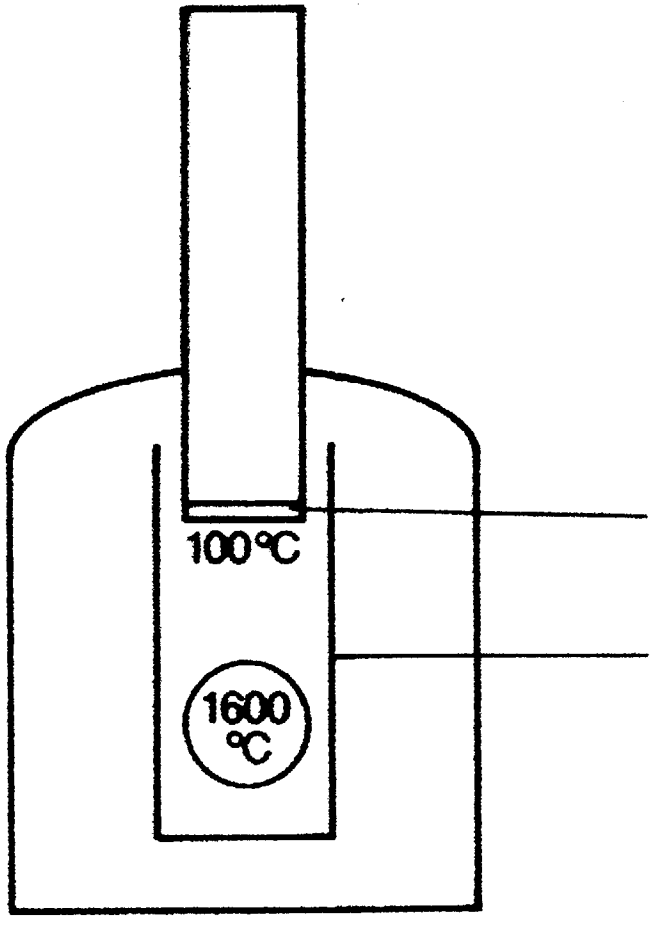


Sweep gas circuit of cold finger apparatus (KÜFA).



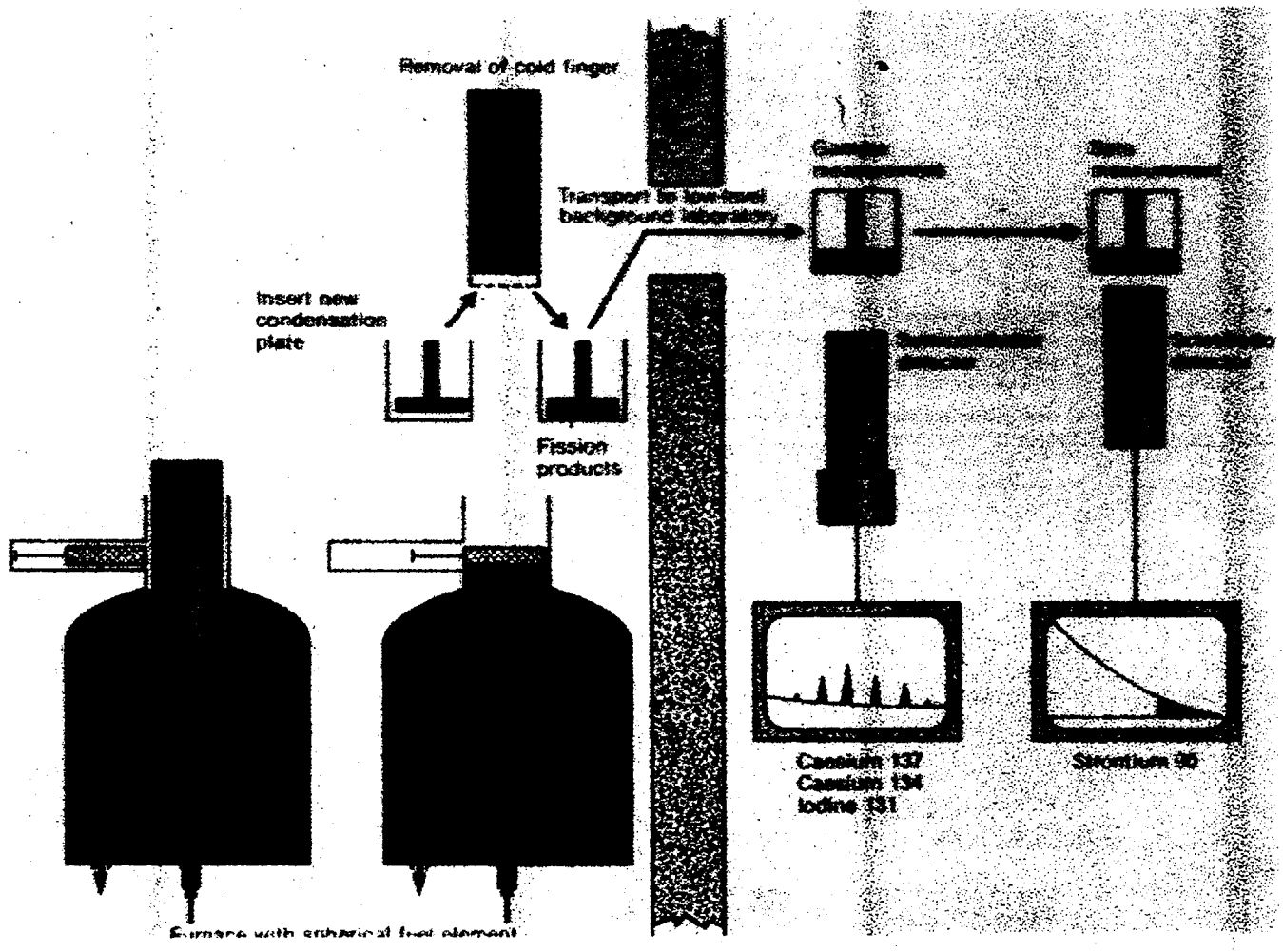
Heating furnace used in accident simulation tests with irradiated spherical fuel elements (KÜFA).

Plate-out fractions of KÜFA furnace cold finger plate



	Released Fraction	
	Cs, I, Ag	Sr
plate	0.7	0.2
Ta-tube	0.02	0.8
Furnace	0.3	-

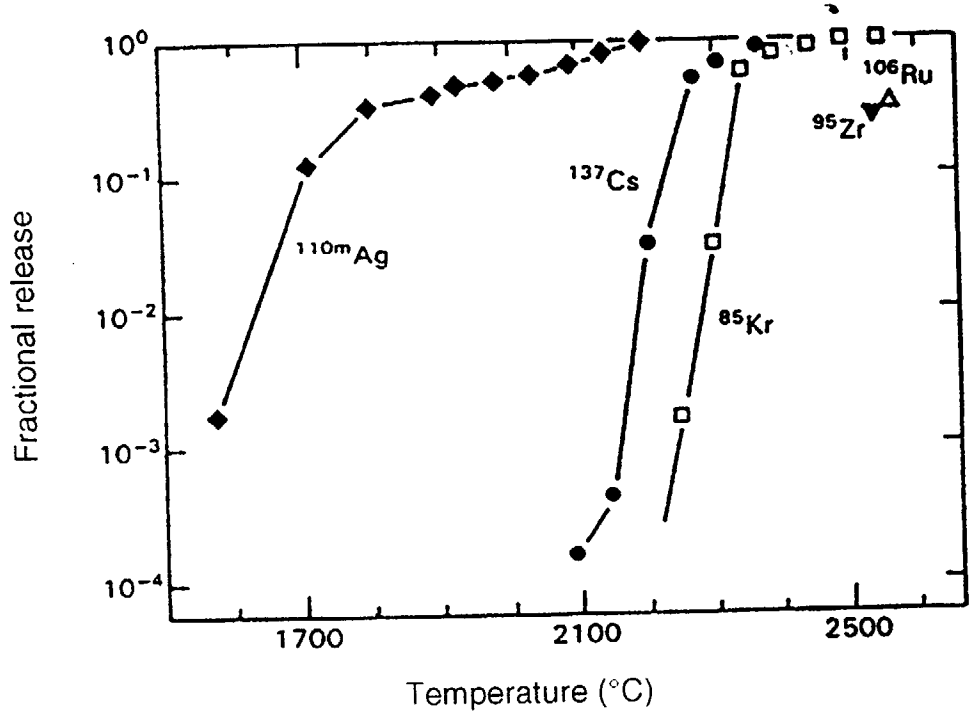
Schema of KÜFA solid fission product measurements



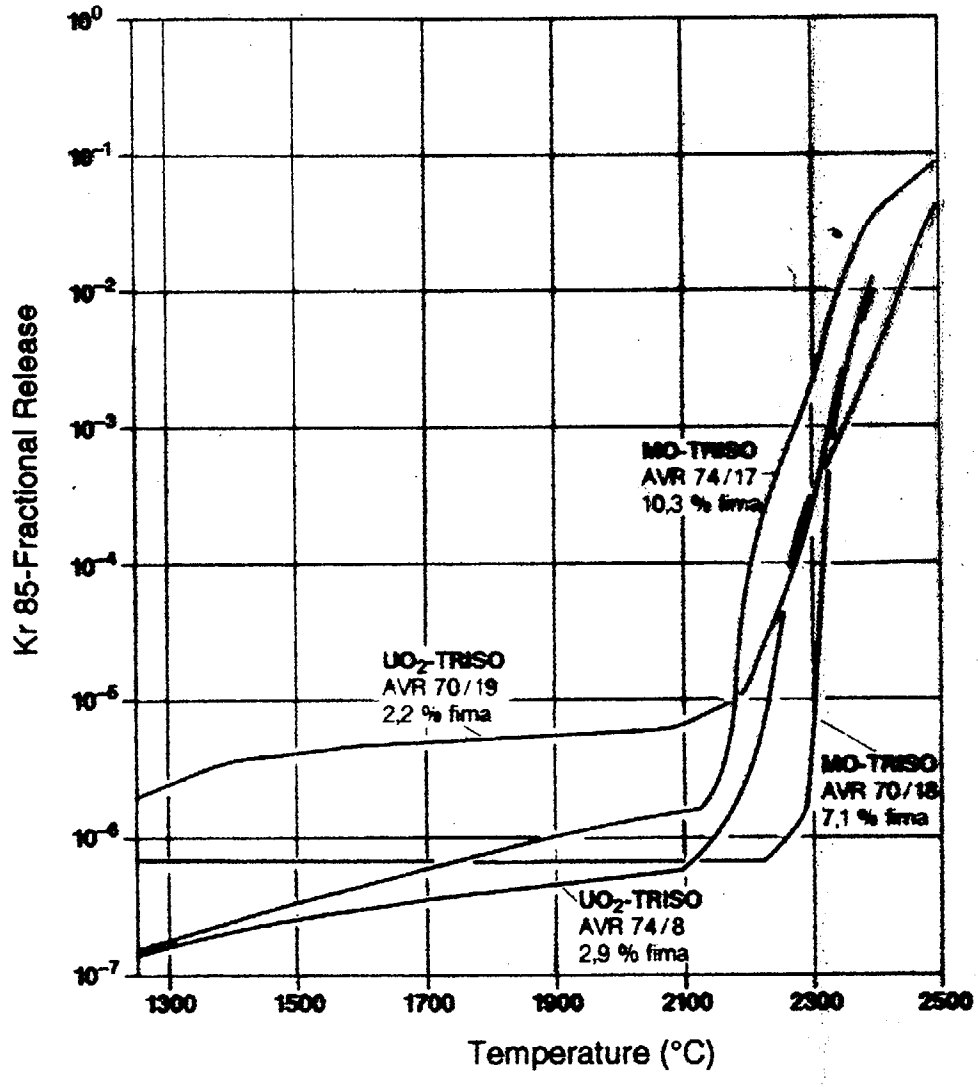
Fission products Cs, Sr, I are radiologically significant because— unlike the noble fission gases — they can be incorporated in the human body.

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

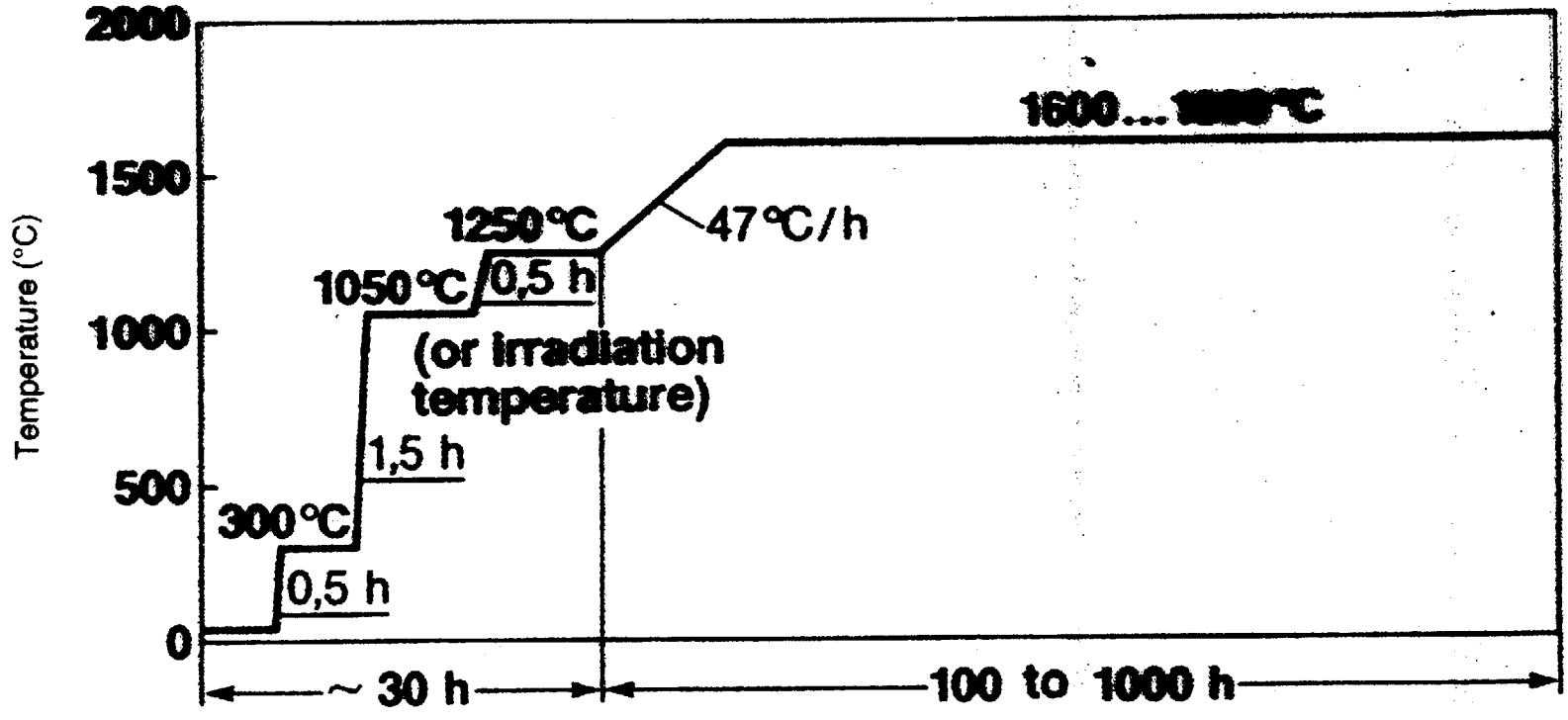
Sequence of fission product release is ^{110m}Ag, ^{137}Cs, ^{134}Cs, ^{85}Kr, ^{90}Sr, ^{106}Ru, ^{95}Zr



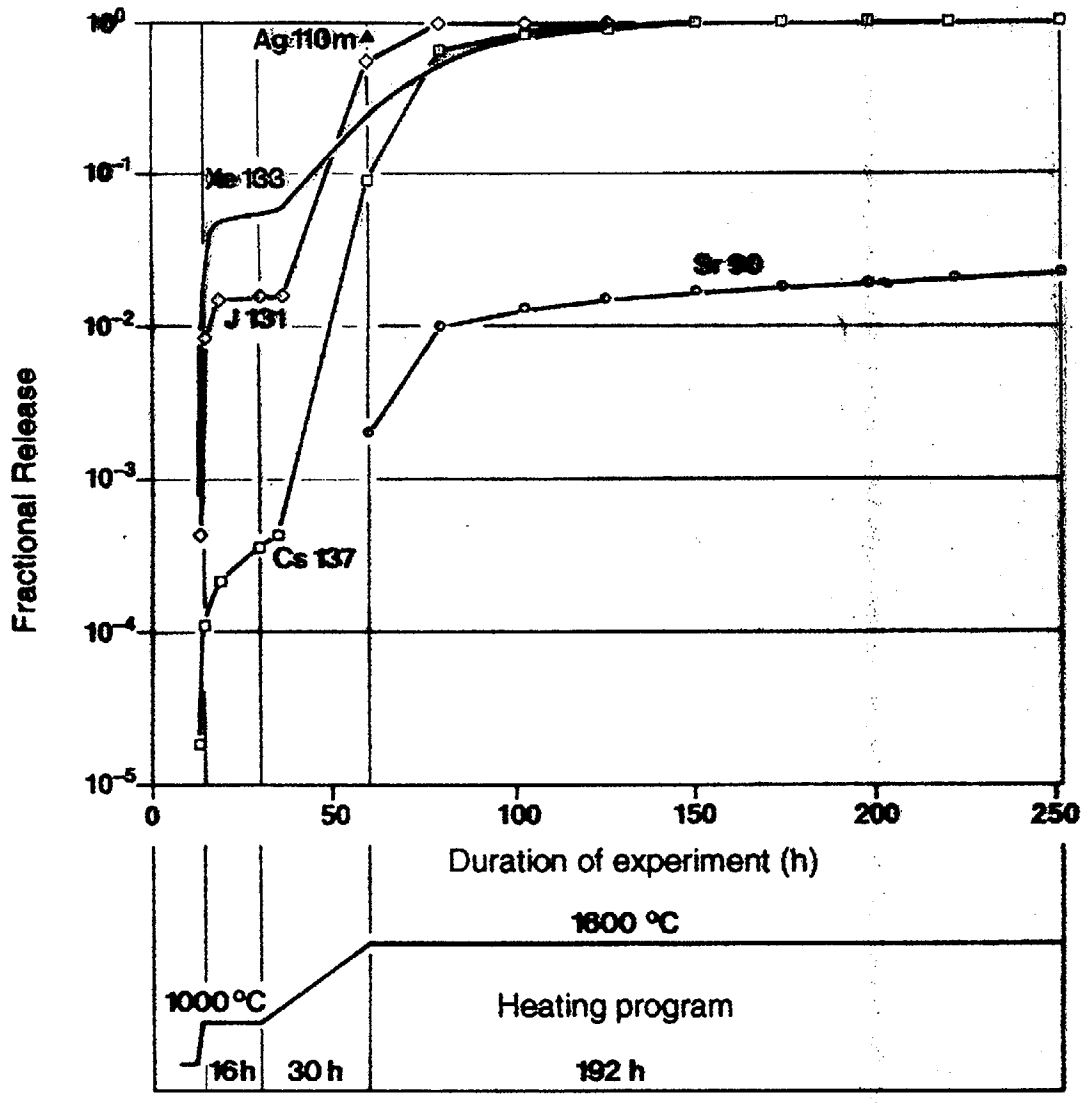
Here the fractional release during temperature ramp to 2600°C [GA data, from Fig 4-39, page 192 of IAEA-TECDOC-978]



Gas release during temperature ramp to 2500°C.

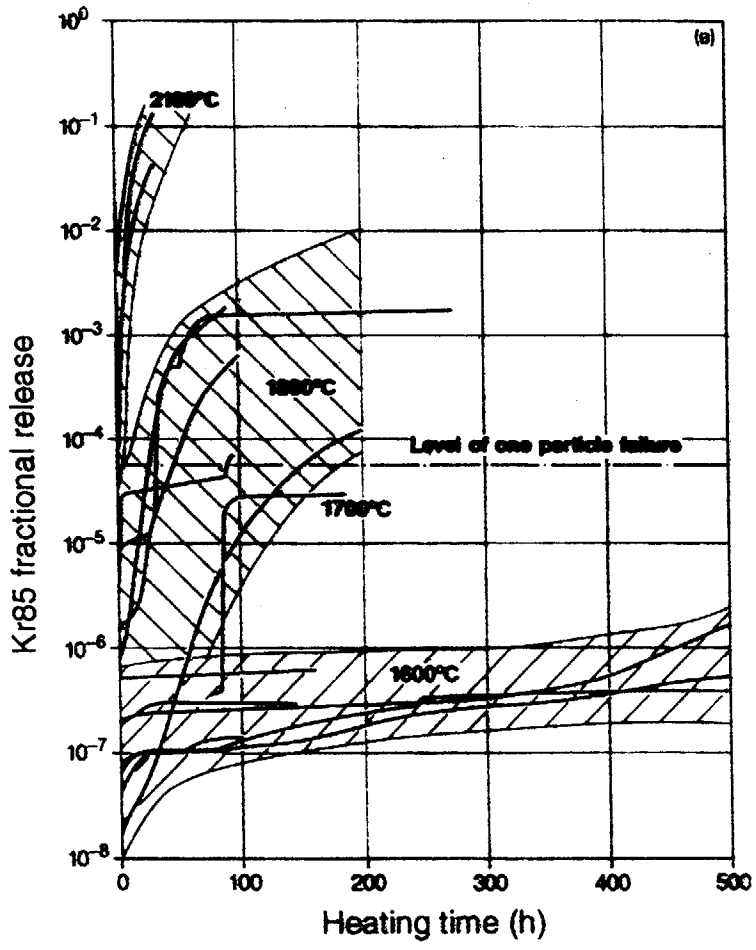


Temperature / time correlation in standard isothermal tests.

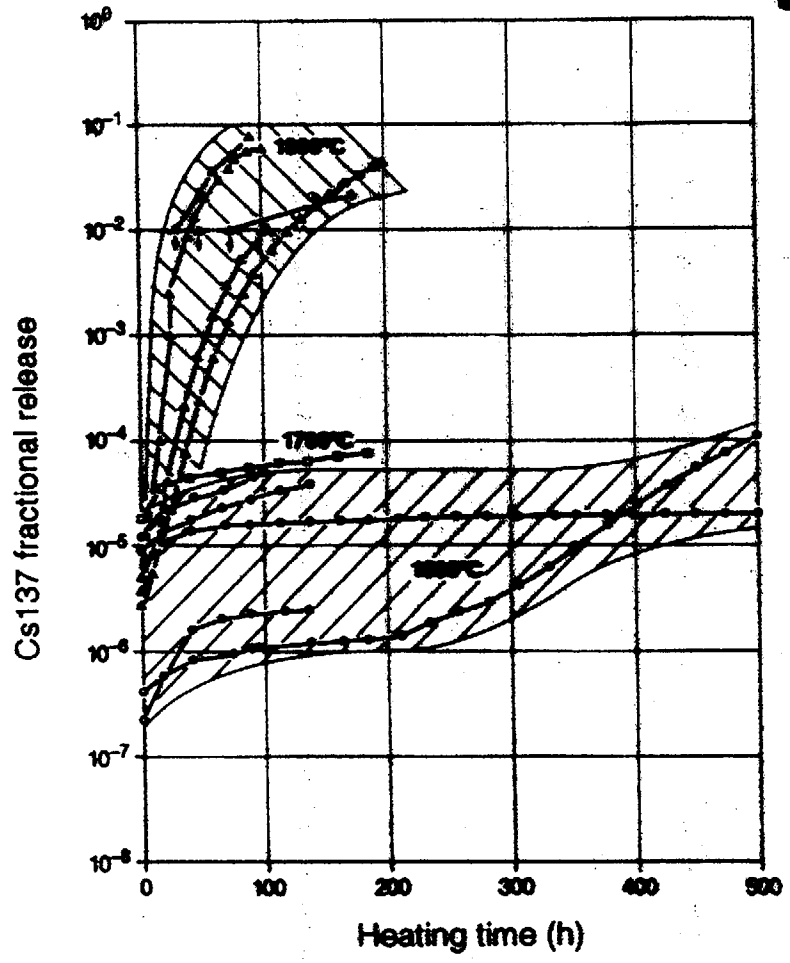


Fission product release from UO_2 kernels (FRJ2-P28/C6) during heating test at 1600 °C.

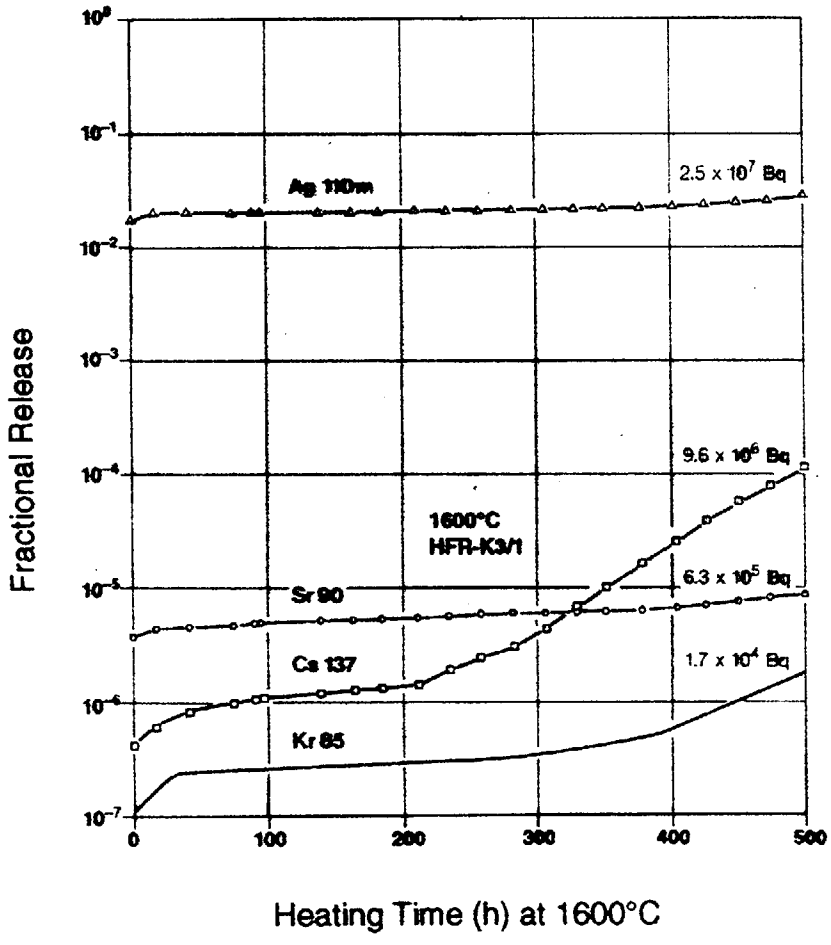
Heating tests at 1600-2100 °C



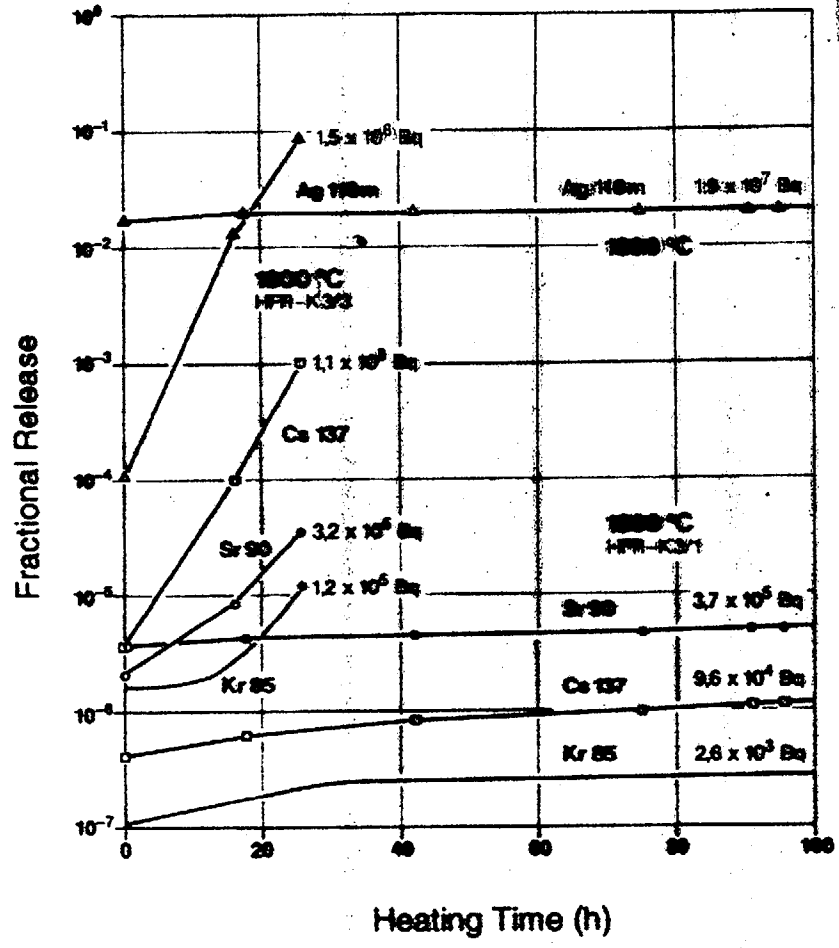
Krypton release during tests with irradiated spherical fuel elements at 1600 to 2100 °C.



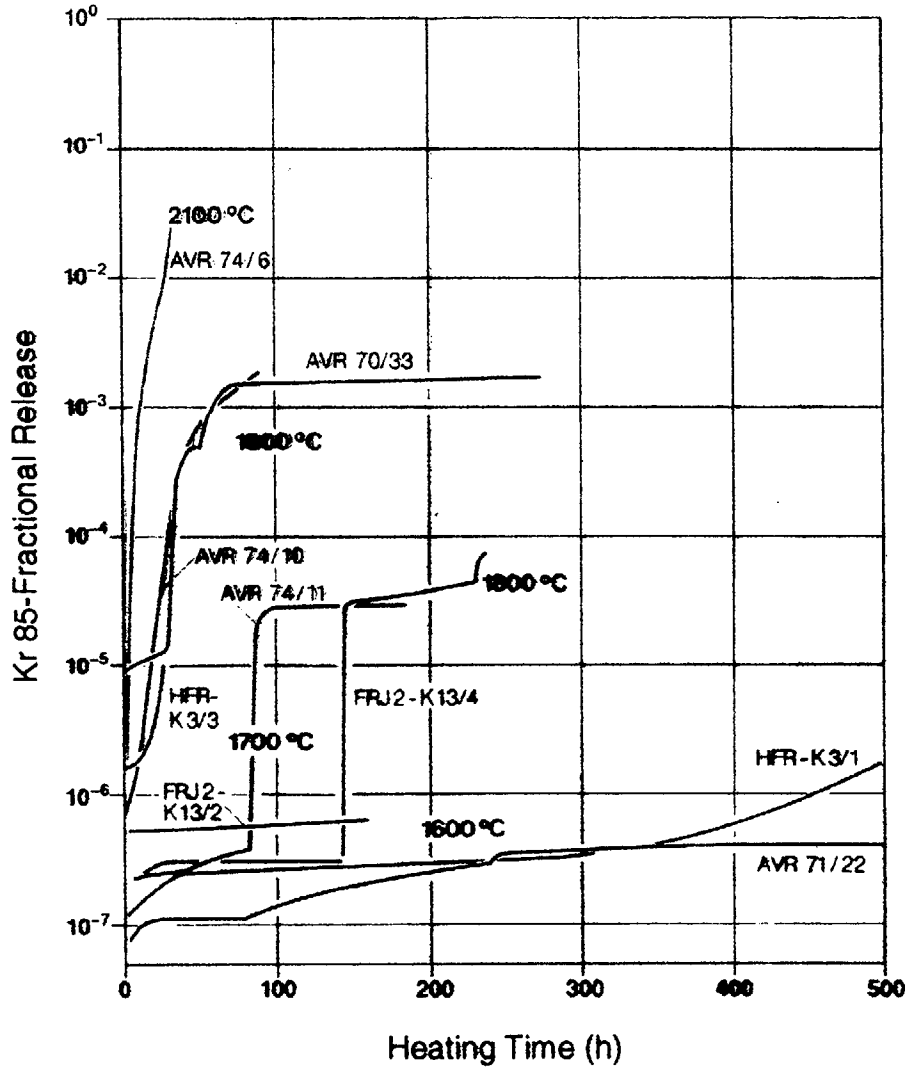
Caesium release from heated spheres as a function of heating times up to 500 hours.



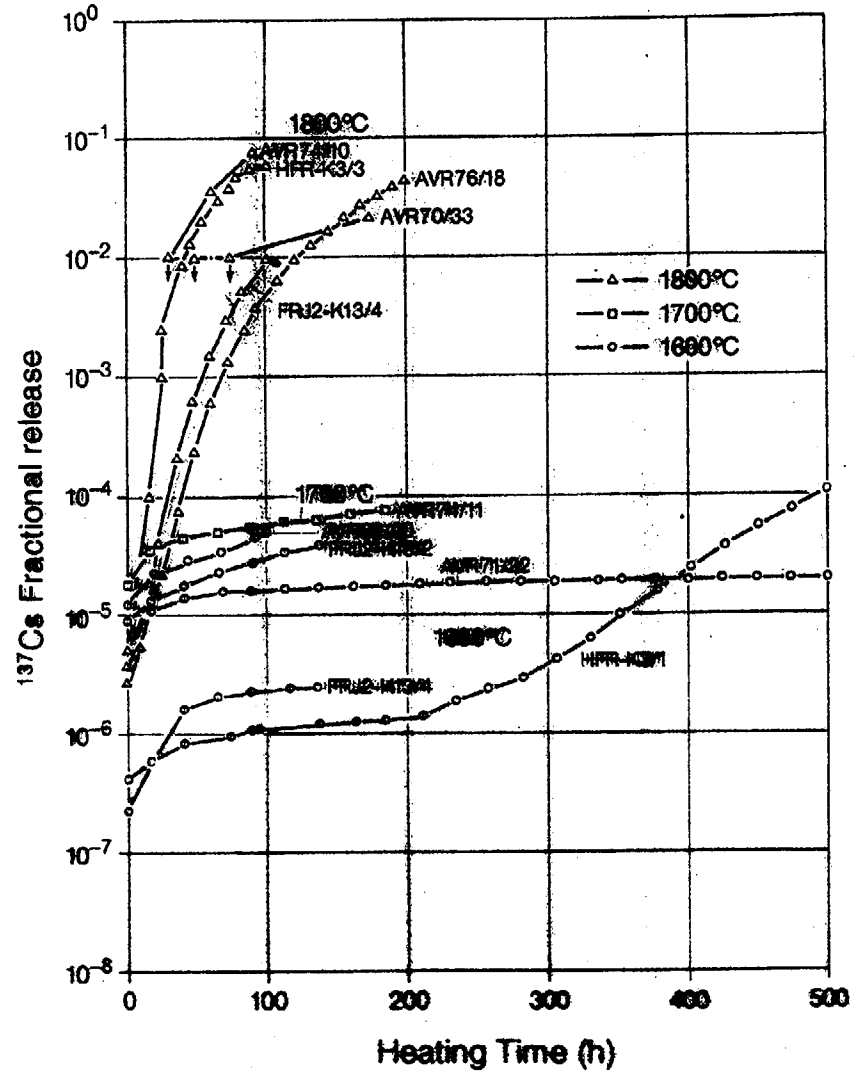
Fission product release from fuel element HFR-K3/1.



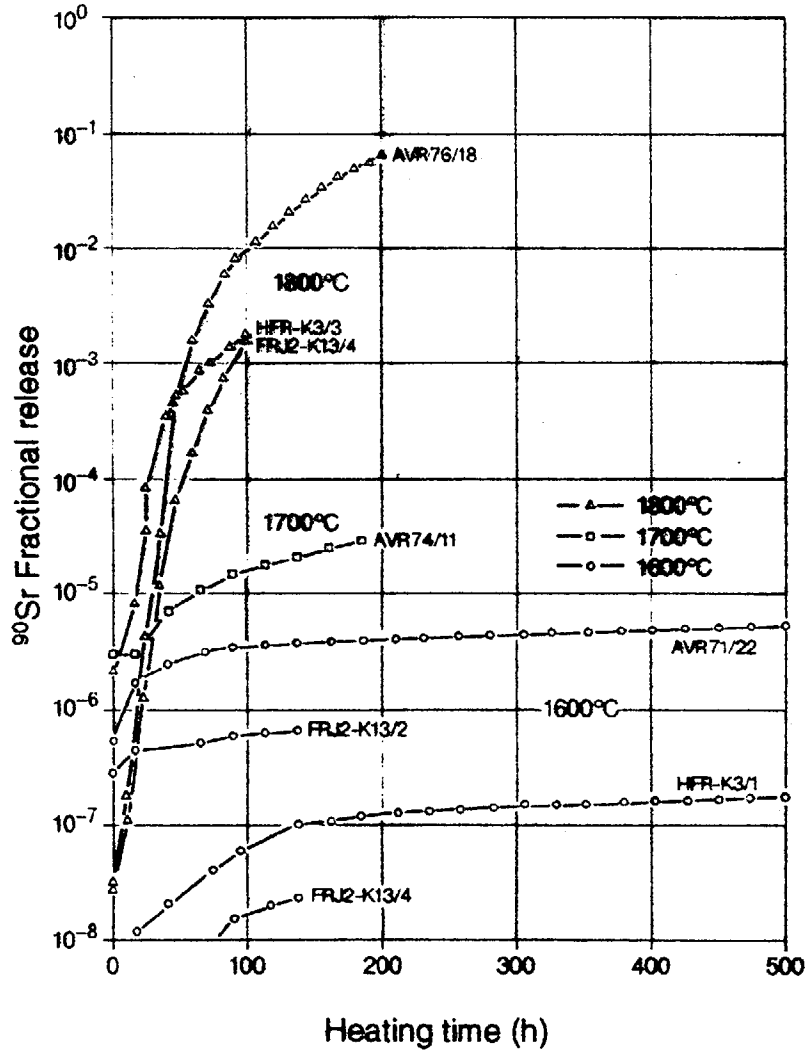
Fission product release at 1600°C and 1800°C.



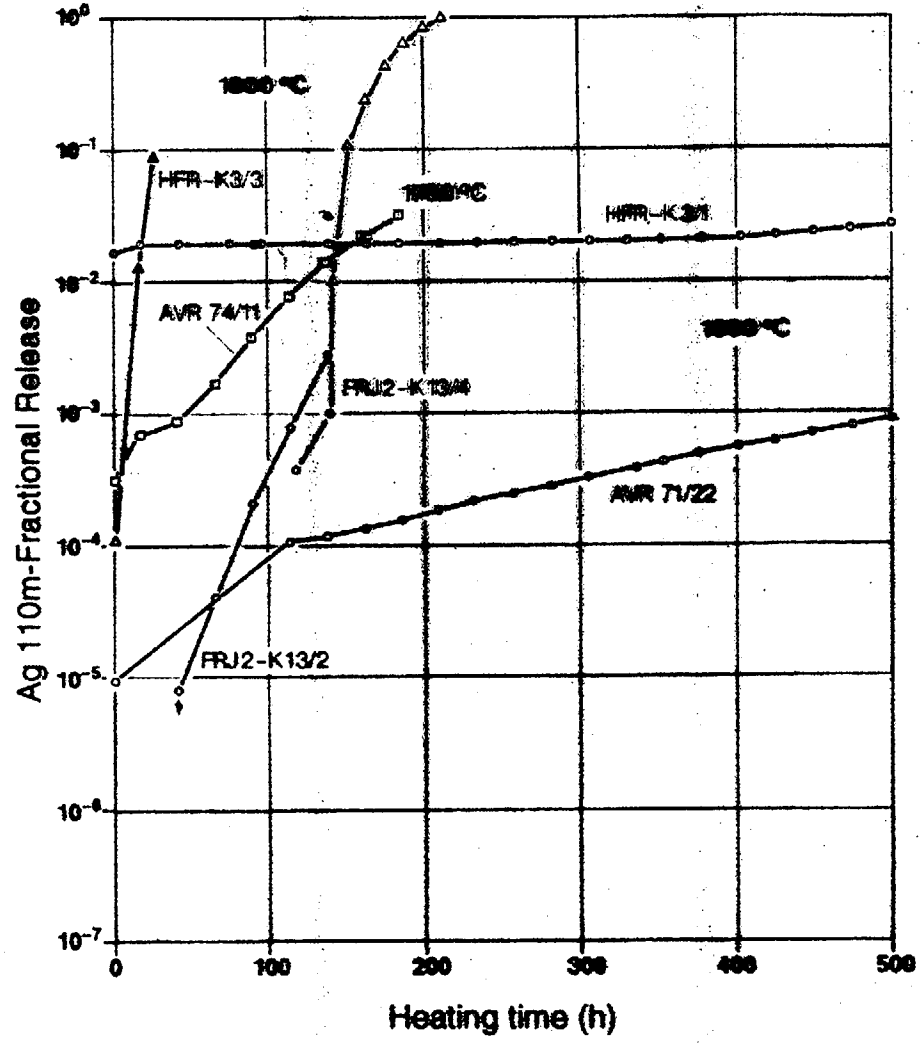
Krypton release in isothermal heating tests.



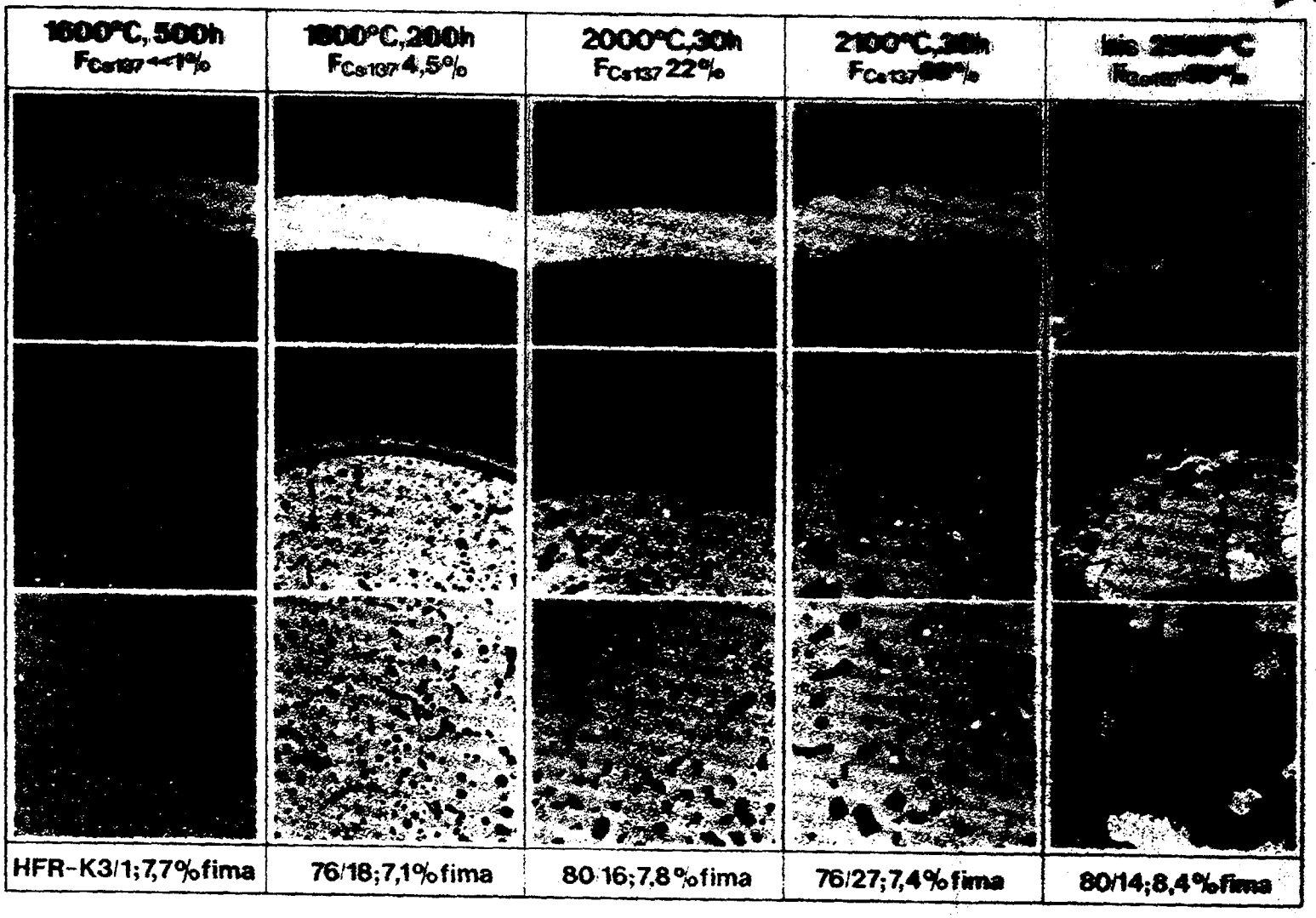
Cesium release in isothermal heating tests.



Strontium release in isothermal heating tests.

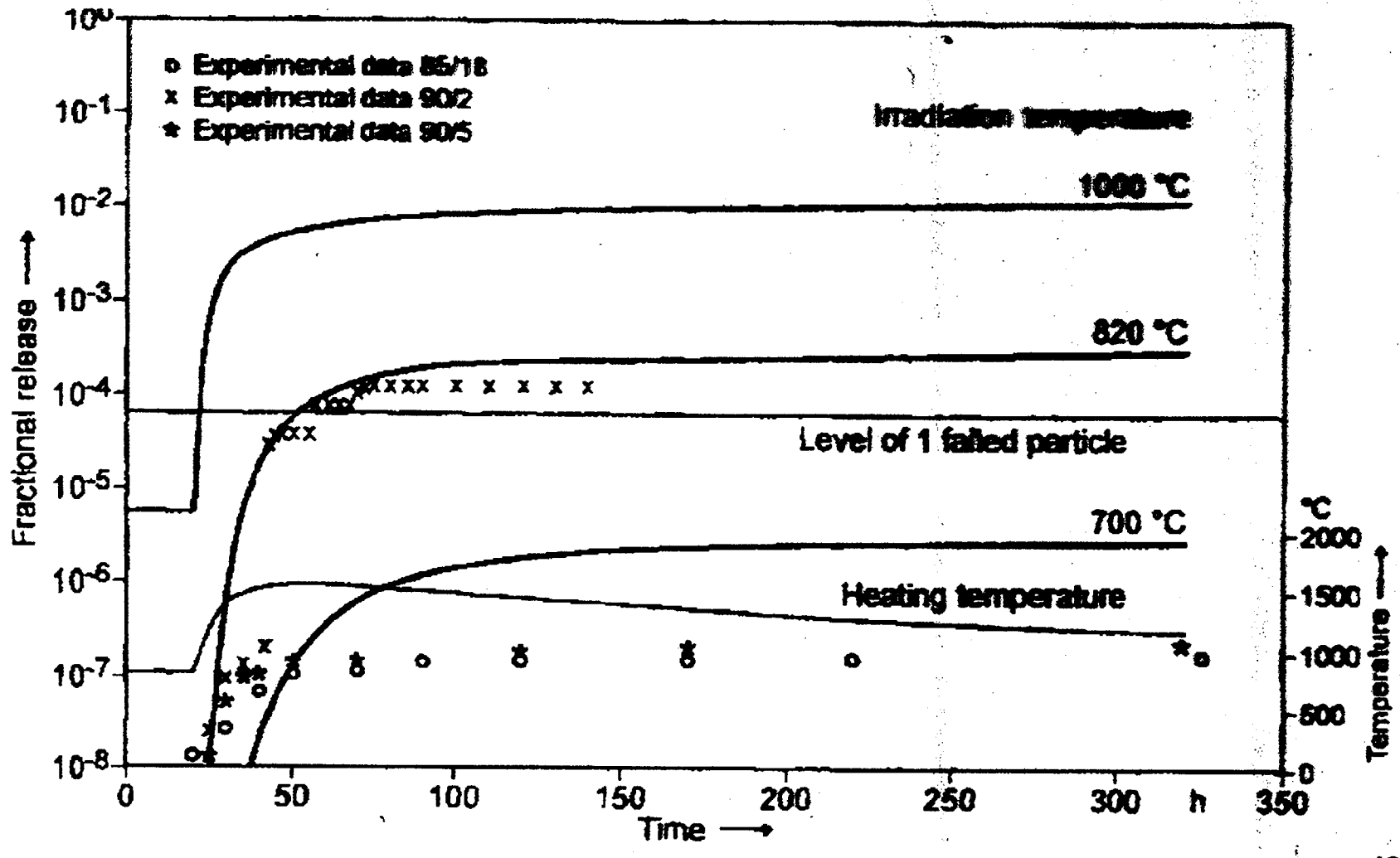


Ag 110m release from fuel elements with UO_2 TRISO particles.

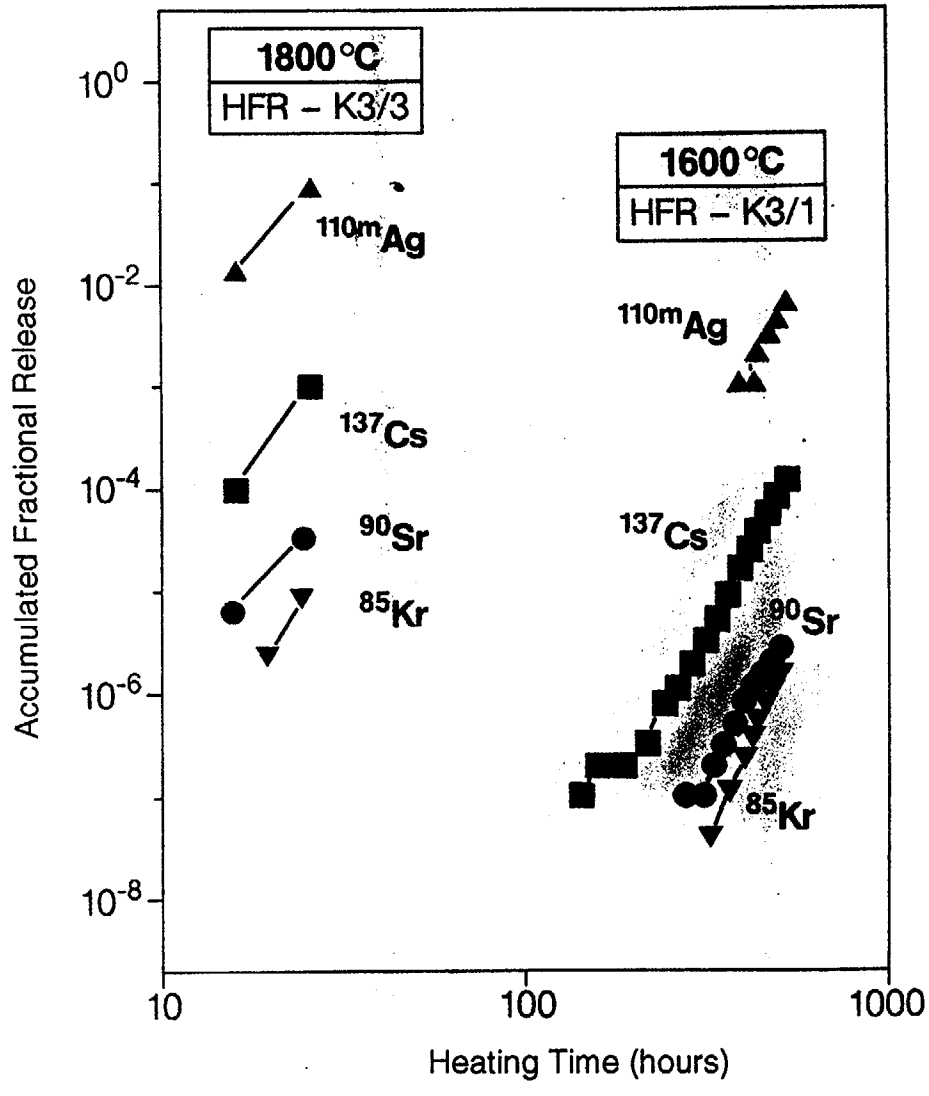


Ceramographic sections through UO_2 TRISO particles.

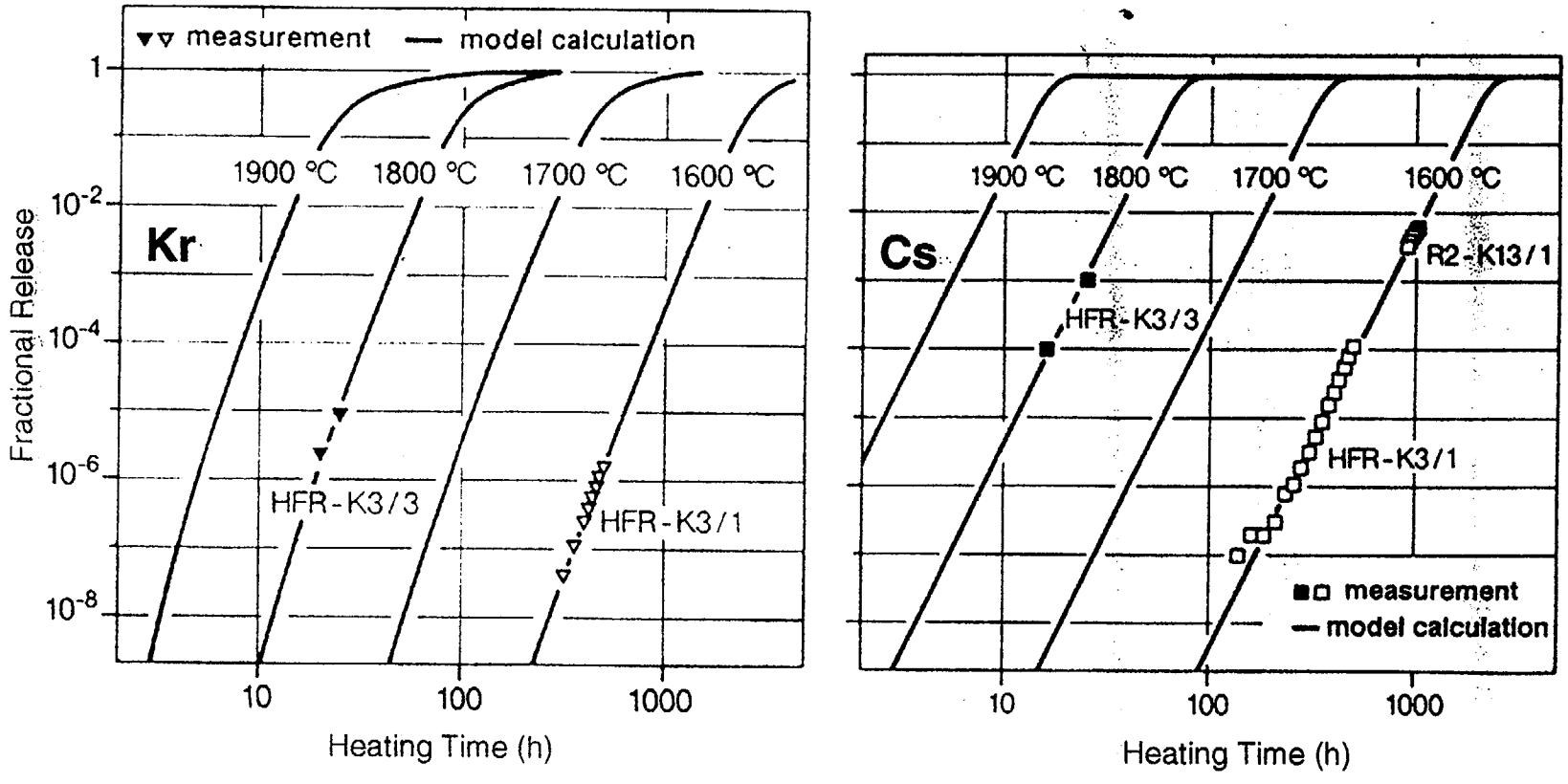
Measured krypton releases from AVR fuel elements [illegible], [illegible] and [illegible] during accident scenario heatup to 1620°C and three PANSIMA predictions using various assumed passages through AVR core:



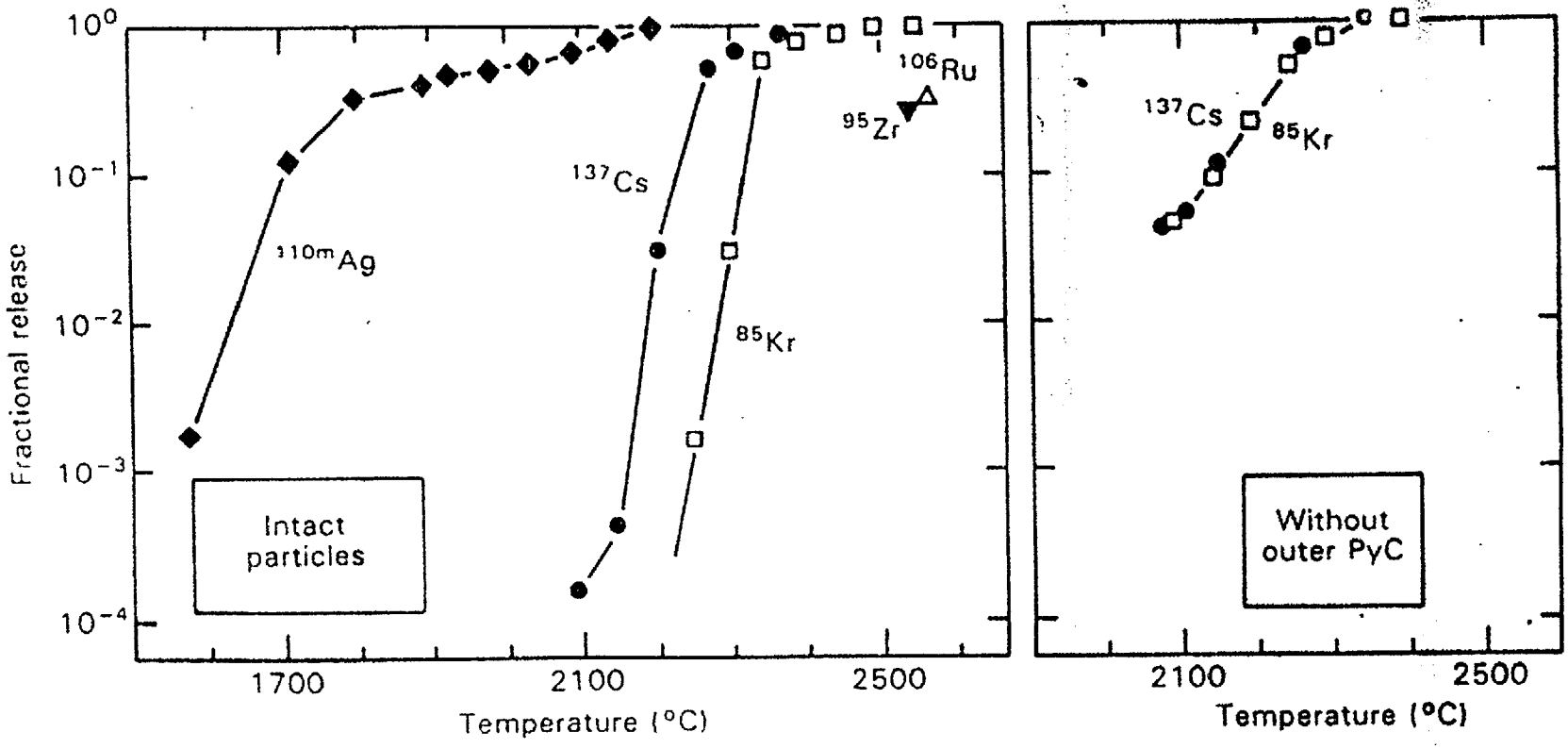
After subtraction of contamination components, fission product release curves increase systematically indicating the progress of SiC deterioration during heating. This observation led to the development of the Goodin-Nabielek, abandoning the classical approach of pressure vessel failure modeling/ diffusional release prediction.



The Goodin-Nabielek model for Kr and Cs release predictions



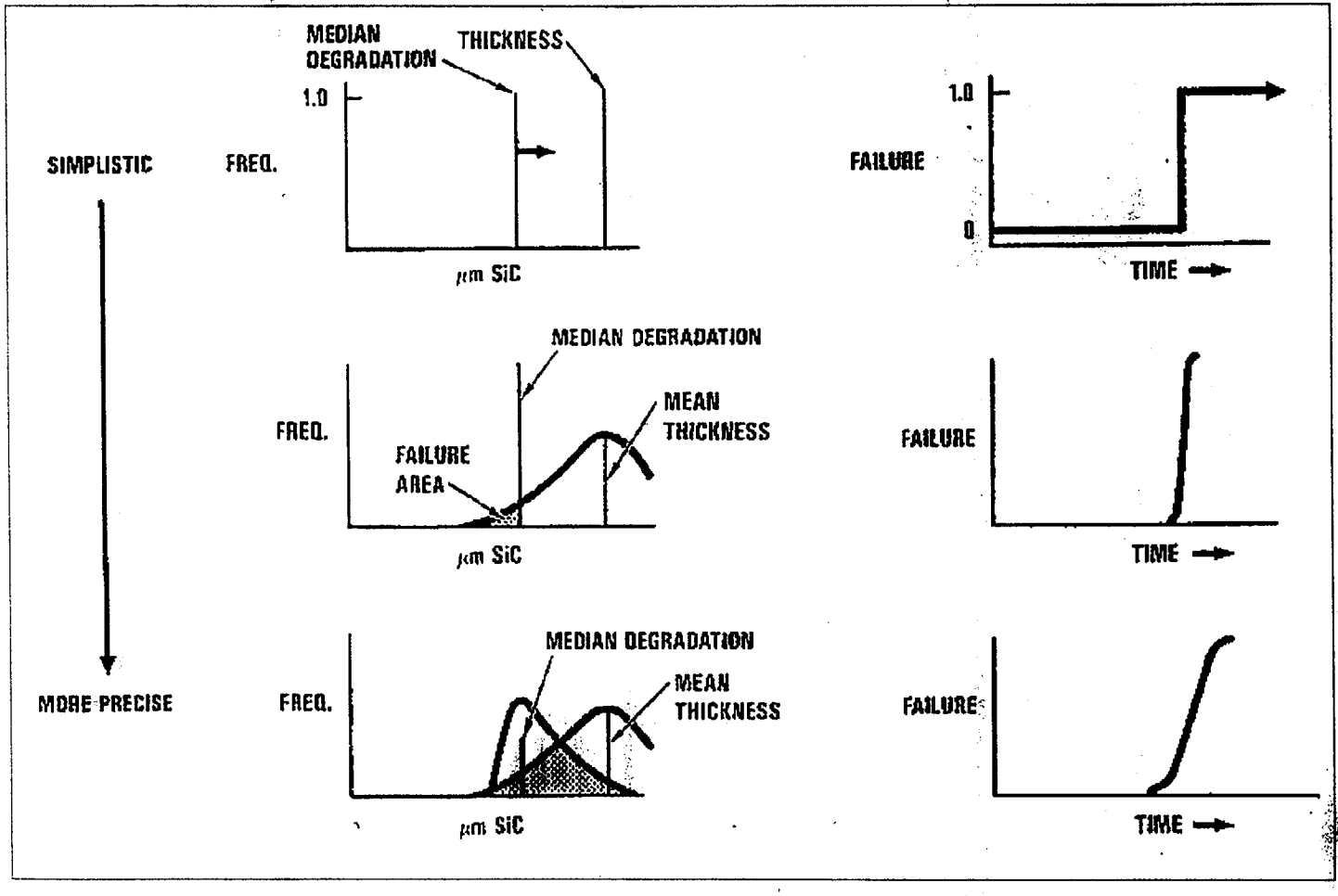
Observed fission product release sequence during ramp



Typical fission product release profiles during linear temperature ramp. In both cases, ~200 irradiated particles were heated to 2500°C. The left diagram shows intact particles, and the right diagram shows particles where the outer PyC layers had been removed.

Forschungszentrum Jülich

The Goodin-Nabielek Modelling Approach combines the statistics of SiC degradation with the variation of SiC thickness (= SiC mass) to predict simultaneously caesium and subsequent krypton release.



Goodin-Nabielek Modelling Approaches

- **Silicon carbide corrosion rate k is temperature dependent**

$$k(T) = k_0 \, e^{-Q/RT}$$

Q... activation energy

R... gas constant

T ... heating temperature

- **The statistics of SiC failure determines Cs release**

$$\Phi(t) = 1 - 2^{-(kt)^m}$$

Φ... SiC failure fraction = Cs release fraction

t ... heating time

m... Weibull modulus

- **Diffusion of Kr through outer PyC after SiC failure**

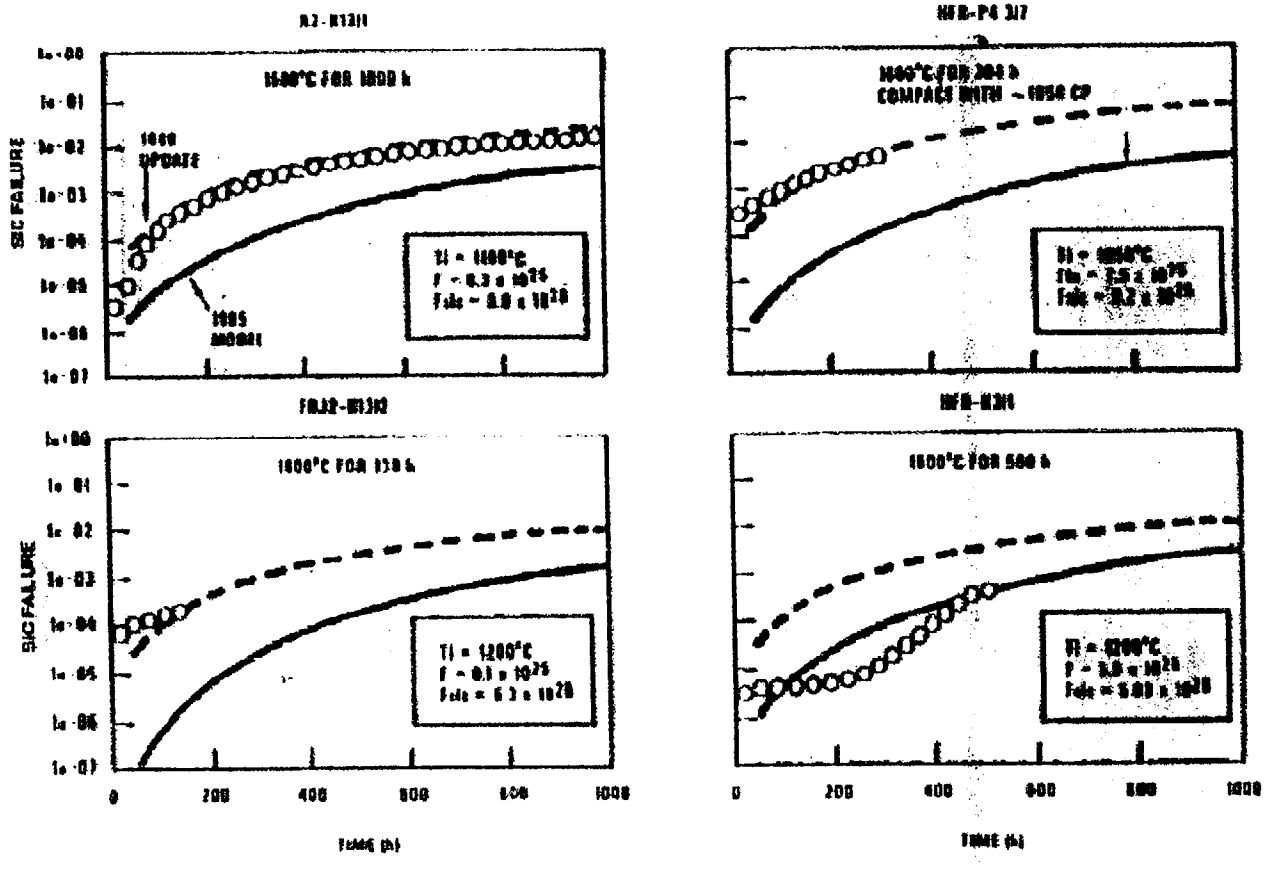
$$F_{Kr}(t) = \int_0^t \frac{d\Phi(t')}{dt'} F_{PyC}(t-t')\, dt'$$

F_{Kr} ... Kr release fraction from particles

F_{PyC}... Kr diffusion fraction through outer PyC

Jülich

1988 MODEL UPDATE CORRELATES WITH THE HEATING DATA AT 1600°C



Forschungszentrum Jülich

Goodin-Nabielek Approach with SiC Degradation 10

A subsequent evaluation of about 20 heating tests by Goodin[84,85] used the degradation rate

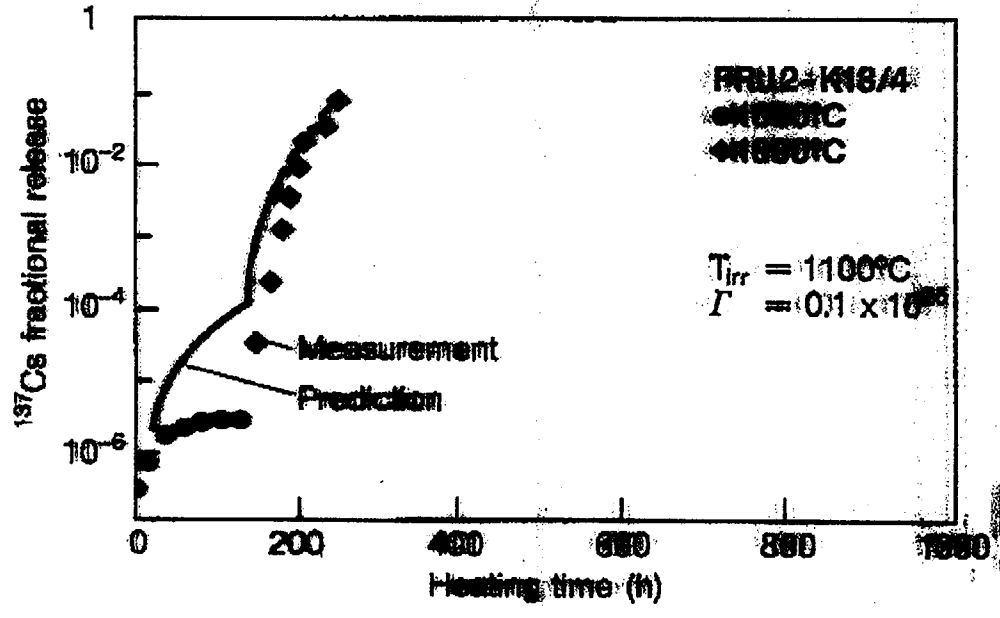
$$k_o(s^{-1}) = 5.03 \cdot 10^{-4} \times \left(\frac{\text{fission density}}{1 \cdot 10^{26}}\right)^{2.09} \times \left(\frac{\text{fast fluence}}{1 \cdot 10^{25}\,m^{-2}}\right)^{.041} \times \left(\frac{\text{irr. temp.}}{K}\right)^{4.14} \qquad (18)$$

Figs. 89 through to 92 show the good agreement with experimentally determined ^{137}Cs release measurements in the range of heating tests from 1600°C to 1800°C.

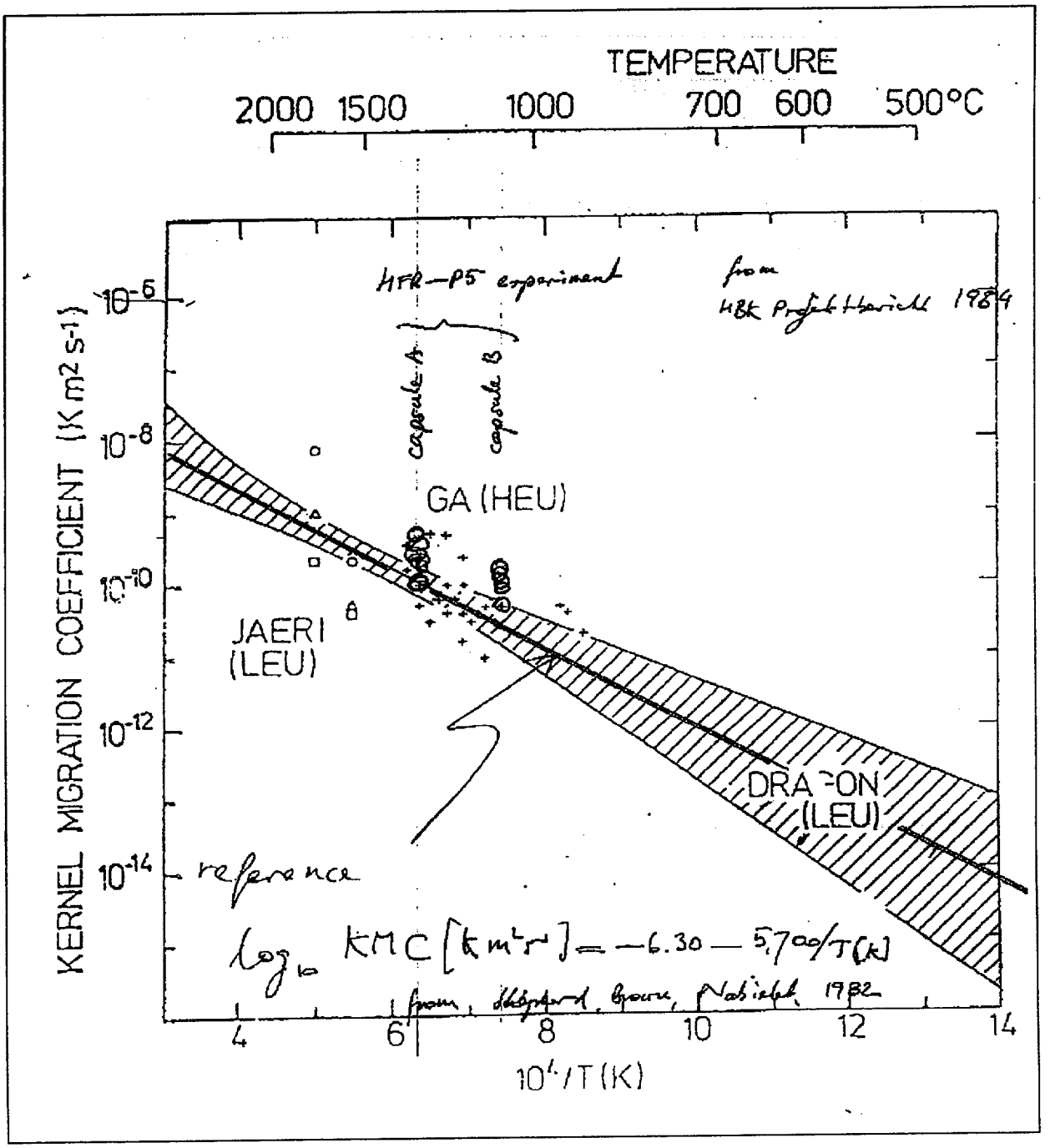
84 D. T. Goodin "US/FRG Accident condition fuel performance models", GA Doc. No. 908293 (DOE-HTGR-85107), Issue A, March 1989.

85 D.T. Goodin "Accident condition fuel performance modelling at GA" in: H. Nabielek (ed.) "US/FRG experts meeting on fuel performance under accident conditions", Proc. Conf. held at ORNL, KFA Internal Report HTA 85-2/90, June 1990.

^{137}Cs release measurement by Schenk and prediction by Goodin for 1600/1800°C heating test of sphere FRJ2-K13/4.



Concern: German Kernel Migration data from HFR-P5 appear to be higher than recommended amoeba line.



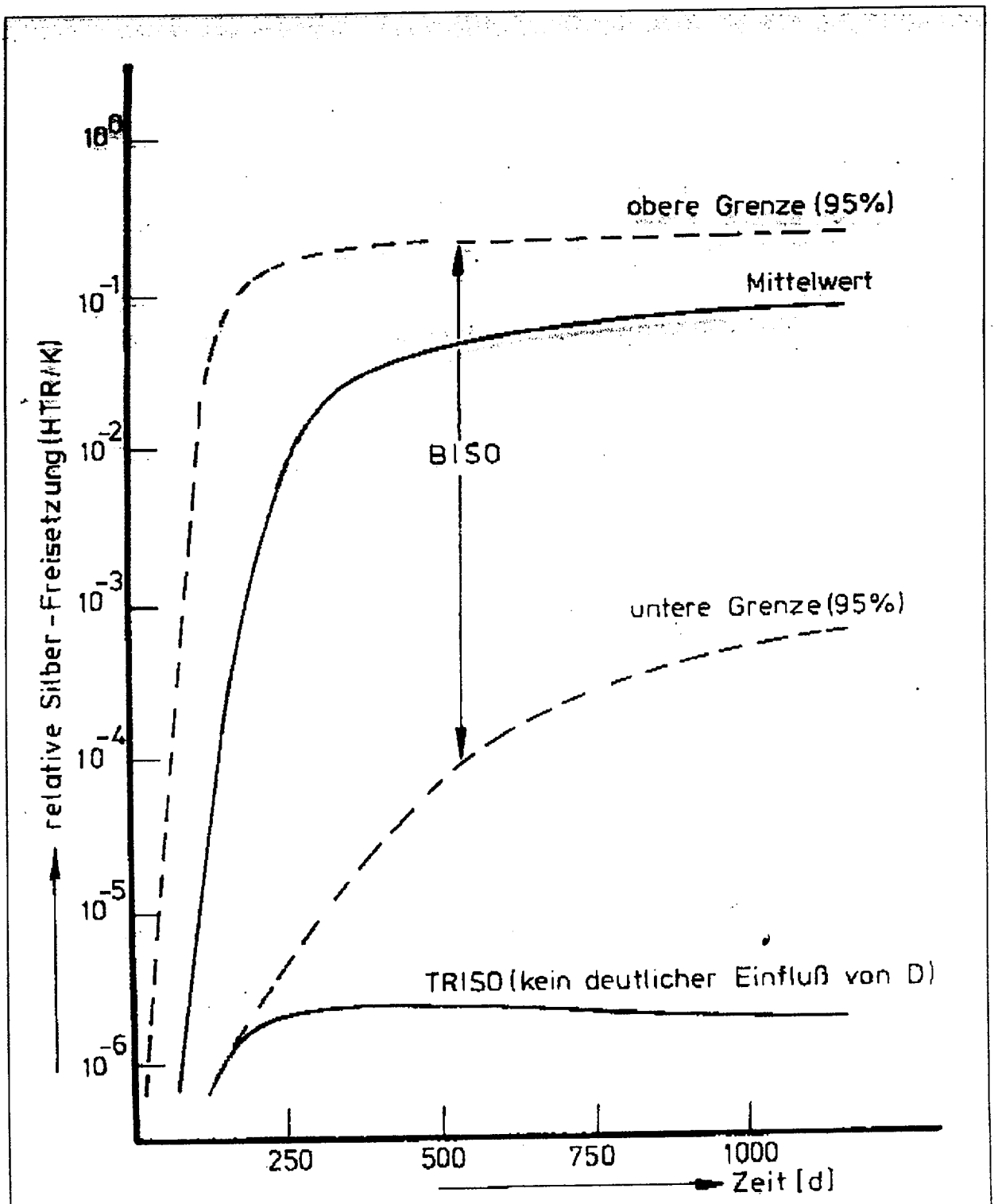


Abb. 6.10: Relative Silber-Freisetzung eines Partikels aus dem Zentrum eines Brennelementes beim Durchlauf durch die zentrale Core-Region (Region 1) eines HTR/K-Reaktors

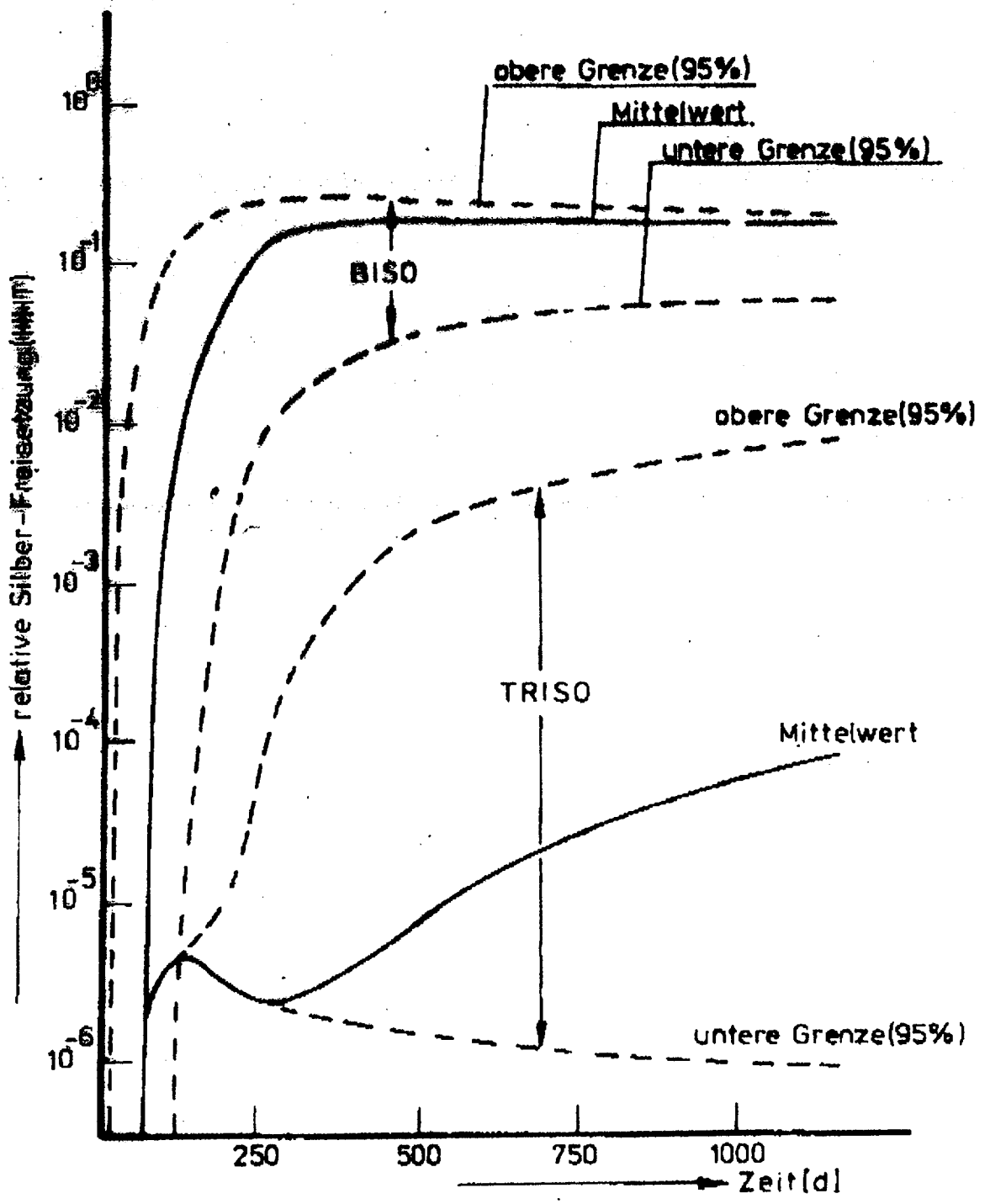
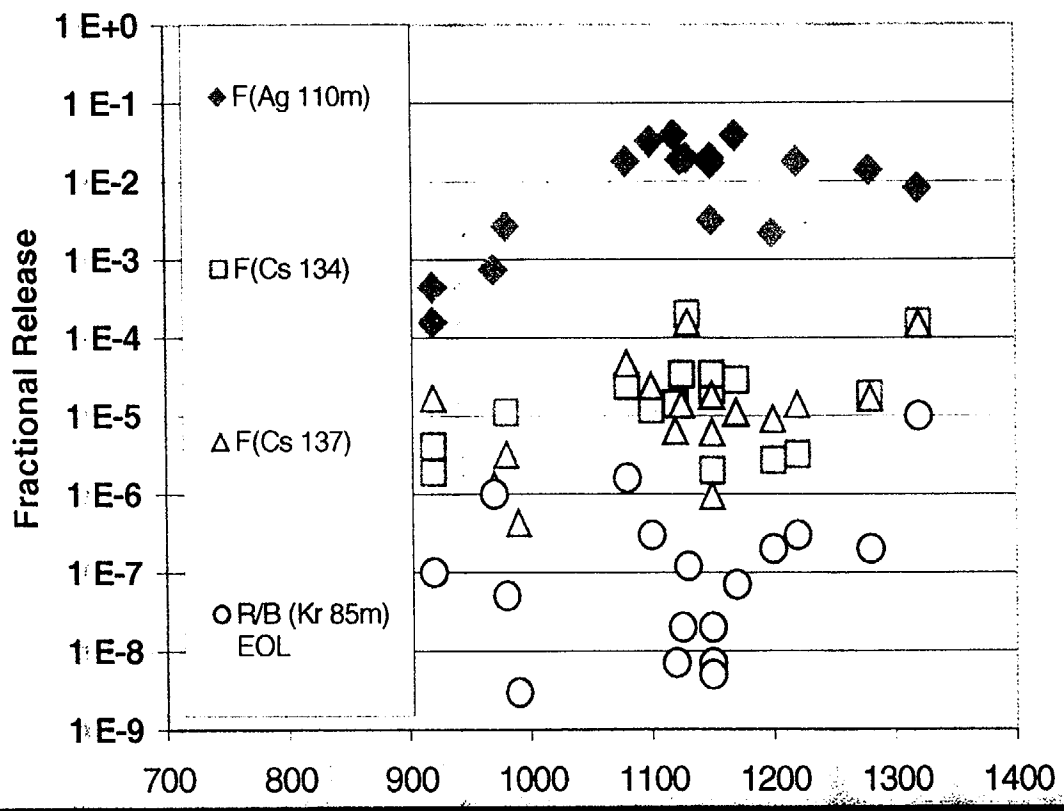


Abb. 6.11: Relative Silber-Freisetzung eines Partikels aus dem Zentrum eines Brennelementes beim Durchlauf durch die zentrale Core-Region (Region 1) eines HHT-Reaktors

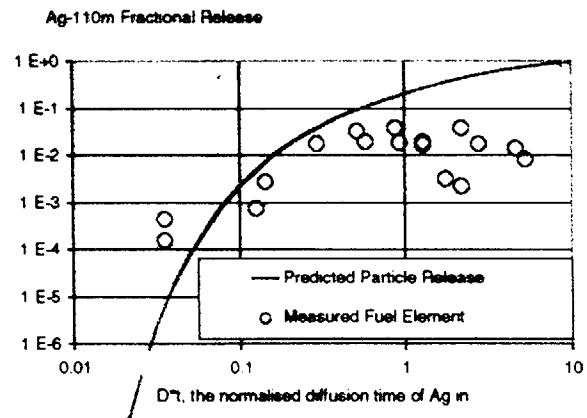
Diagram shows ^{110m}Ag and ^{137}Cs release fractions as a function of temperature in 230-530 d irradiation tests as measured in post irradiation examination work. Also shown is end-of-life R/B(^{85m}Kr).



Gas turbines are planned for all modern HTRs—therefore, we need to:

- Re-analyse existing data (i.e. temperatures and Ag releases)
- Perform dedicated irradiation tests
- Consider design/ material improvements
- If all else fails, redesign the HTR

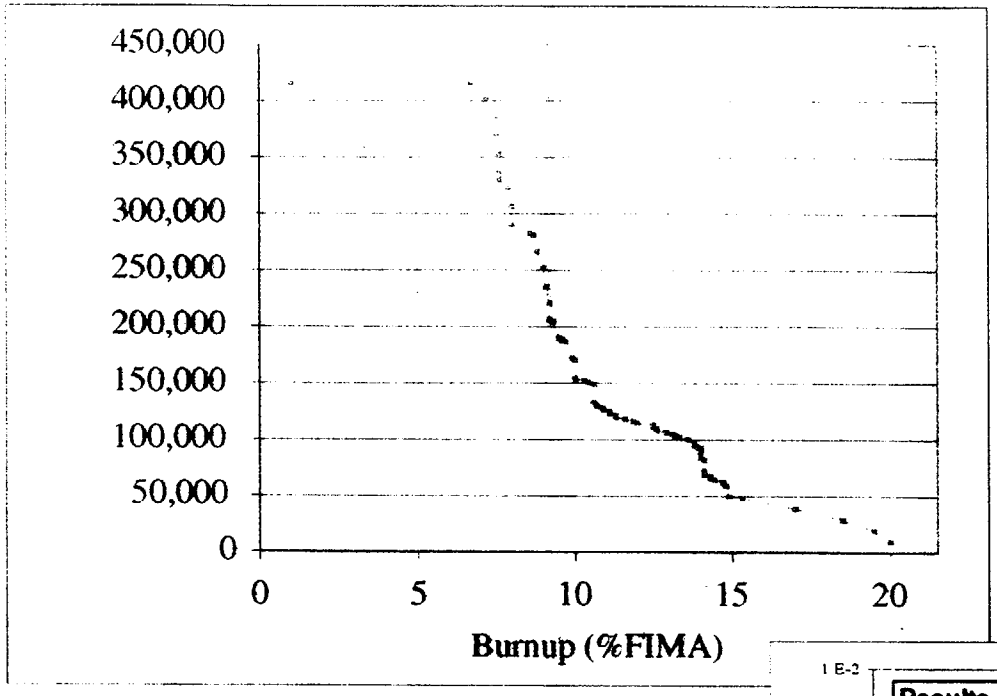
A gross comparison to a silver release prediction — with the diffusion coefficient of silver in SiC as recommended by IAEA TECDOC 978 "Fuel performance and fission product behaviour in gas cooled reactors" and using a primitive breakthrough formula — is shown below:

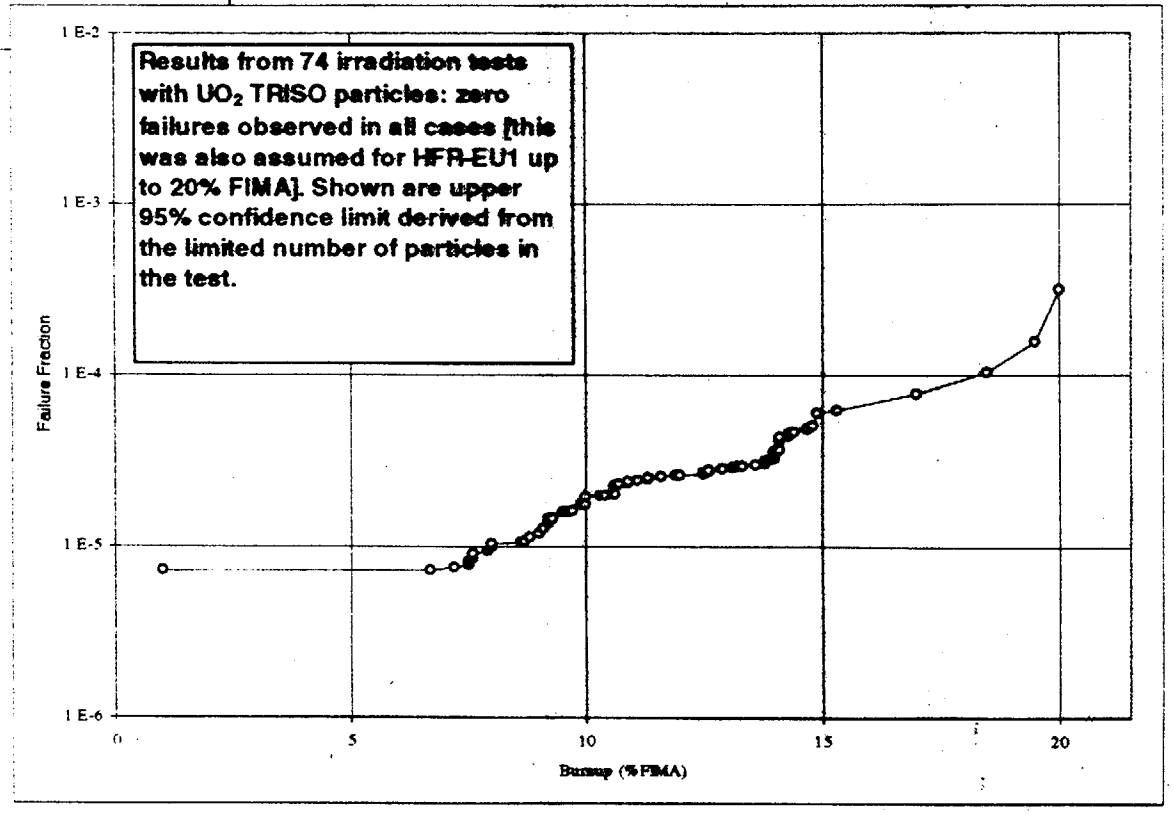


Of course, we need a much more sophisticated evaluation of these experiments taking account of distributions in space and time.

This is particularly necessary in the case of silver where first the stable ^{109}Ag is generated, is starting to be released by diffusion and is later activated to ^{110m}Ag.

Statistical Limits of Irradiation Testing:





AVR reloads in the years 1966 to 1987

Reload number	Insertion date	Fuel element type	Number of fuel elements	Coated fuel particle Kernel	Coating	Enrichment
0	July 66	UCC	30155	$(Th,U)C_2$	HTI BISO	93%
1	Oct. 68	T	7510	$(Th,U)C_2$	HTI BISO	93%
3	Apr. 69	GK	17770	$(Th,U)C_2$	HTI BISO	93%
4	July 70	GK	6210	$(Th,U)C_2$	HTI BISO	93%
5-1	Nov. 70	GK	25970	$(Th,U)C_2$	HTI BISO	93%
5-2	Dec. 71	GO1	20825	$(Th,U)O_2$	HTI BISO	92%
7	Jan. 73	GO1	7840	$(Th,U)O_2$	HTI BISO	93%
6-1	Oct. 73	GO1	11000	$(Th,U)O_2$	HTI BISO	92%
6-2	Dec. 73	GLE1	2446	UO_2 UO_2	LTI BISO LTI BISO	15% 0.7%
8-1	May 74	GFB1	1440	UO_2 ThO_2	LTI BISO LTI BISO	93% -
8-2	May 74	GFB2	1610	UO_2 ThO_2	LTI TRISO LTI BISO	93% -
9	Sep. 74	THTR-1	5145	$(Th,U)O_2$	HTI BISO	93%
10	Dec. 74	THTR-2	10000	$(Th,U)O_2$	HTI BISO	93%
11	Dec. 74	THTR-2	5000	$(Th,U)O_2$	HTI BISO	93%
12	Mar. 76	GO1	11325	$(Th,U)O_2$	HTI BISO	93%
14	Nov. 76	GO1	9930	$(Th,U)O_2$	HTI BISO	93%
13-1	Dec. 77	GFB3	6077	UC_2 ThO_2	LTI TRISO LTI BISO	90% -
13-3	Dec. 77	GFB5	5354	UCO ThO_2	LTI TRISO LTI TRISO	92% -
13-2	July 80	GFB4	5861	UC_2 ThO_2 with gettering additives	LTI TRISO LTI BISO	90% -
15	Feb. 81	GO2	6067	$(Th,U)O_2$	LTI TRISO	93%
18	July 81	GO3	11547	$(Th,U)O_2$	HTI BISO with low heavy metal contamination	93%
19	July 82	GLE3	24615	UO_2	LTI TRISO	10%
21	Feb. 84	GLE4	20250	UO_2	LTI TRISO	17%
20	Oct. 85	GO2	11854	$(Th,U)O_2$	LTI TRISO	93%
22	Sep. 86	THTR	15228	$(Th,U)O_2$	HTI BISO	93%
21-2	Oct. 87	GLE4	8740	UO_2	LTI TRISO	17%
		SUM	289789			

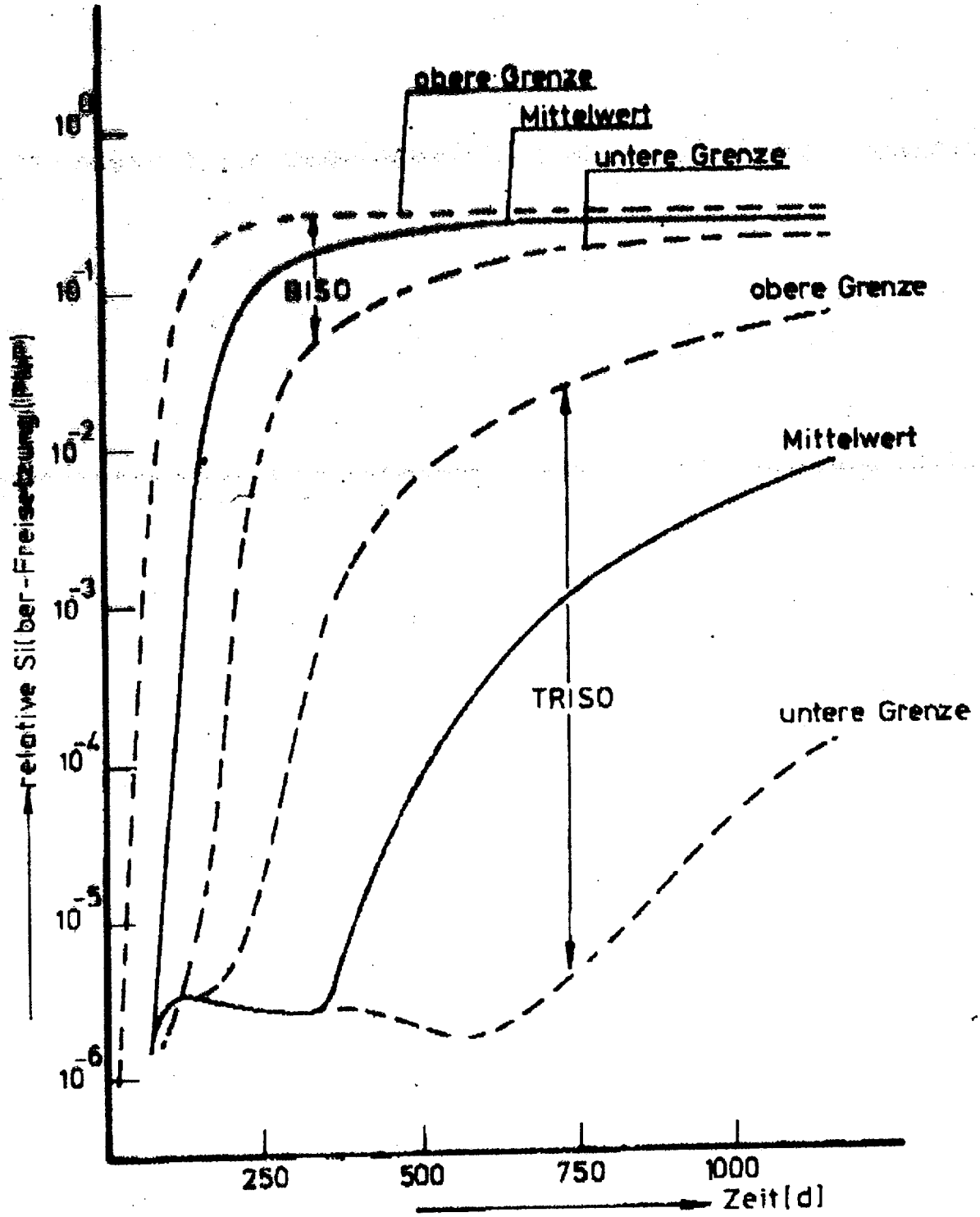


Abb. 6.12: Relative Silber-Freisetzung eines Partikels aus dem Zentrum eines Brennelementes beim Durchlauf durch die zentrale Core-Region (Region 1) eines PNP-Reaktors

Suggested HTR fuel work, to be discussed:

(i) ^{110m}Ag: Re-evaluate release data during normal operations for better source term data base in direct cycle applications.

(ii) Determine influence of burnup > 10% FIMA on irradiation performance, in particular for potential reduction of 1600°C capability.

(iii) Analyse accident condition performance > 1600°C for an improved coated particle model.

Confidence base for HTR

- HTR is alive again—more so than ever before
- Good international basis
  - Fuel manufacture in Britain, US, Germany, Russia, Japan, China
  - Complete irradiation qualification in Germany; many test results from USA, UK, Japan, Russia, China
  - Extensive results from core heatup simulations testing in Germany, USA, Russia and Japan

Next Steps

- Irradiation and core heatup testing in European HTR program [high burnup and >1600°C]
- Experiment planning and evaluation in cooperation with PBMR in South Africa
- German support in South African fuel manufacture
- US-German proposals…
  - international irradiation test, PIE, analysis
  - model development and applications