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2-7-2001

Original slides by Heather Haydock, modified Nabielek and Sjunnesson March 2002.

HTR

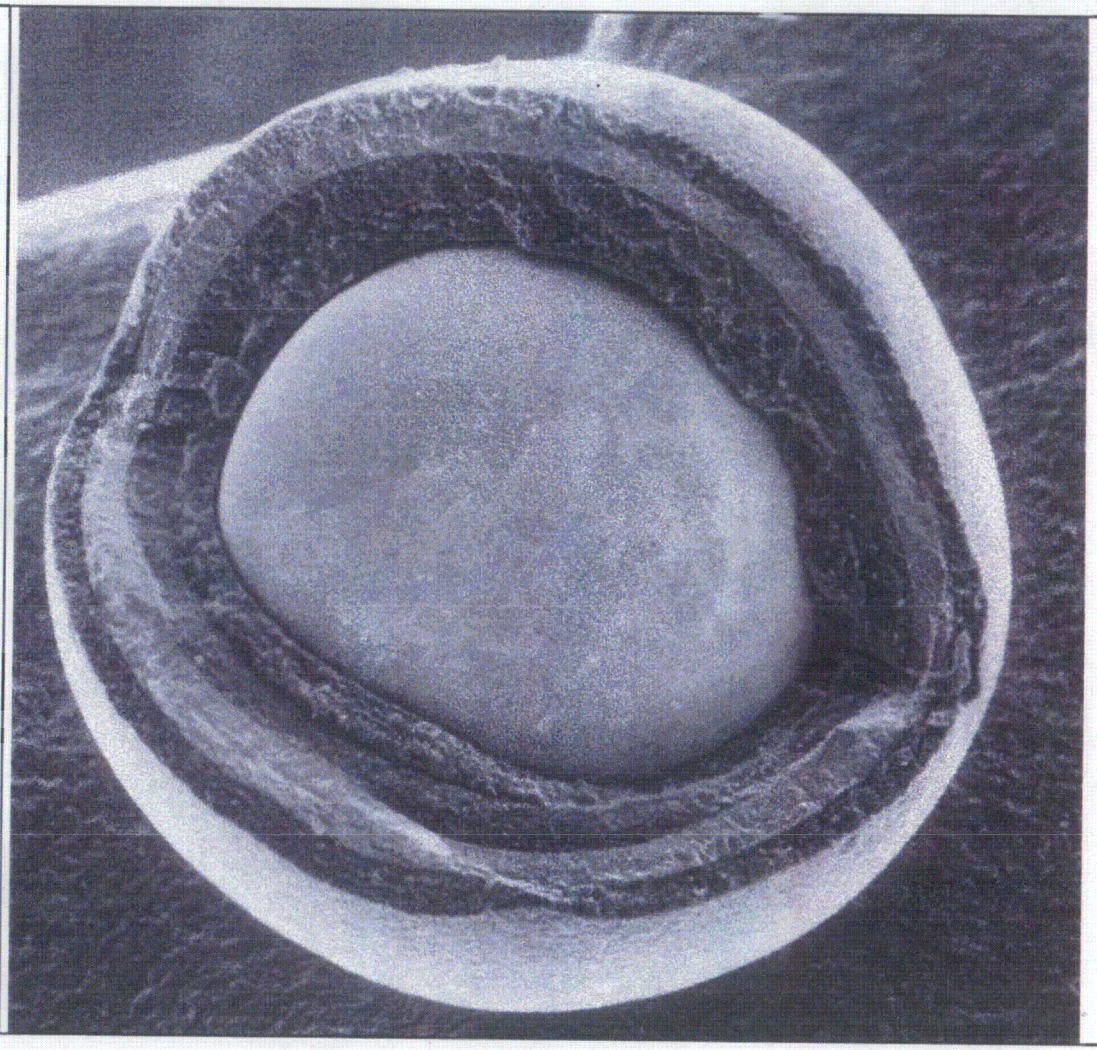
1954 P Fortescue
1956 R Schulten

History of coated particles:

1957 R A U Huddle
1959 W Goeddel
1961 J Oxley, Battelle
fluidised bed coating

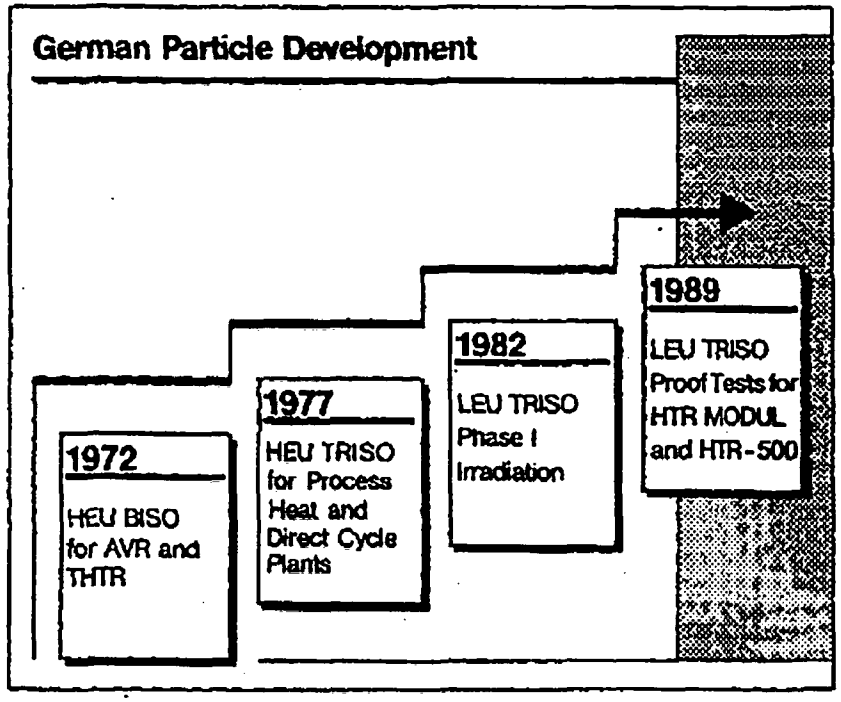
Manufacturing

NUKEM (DE)
UKAEA, BNFL
CERCA (F)
Belgatom
GA, ORNL (USA)
Russia
NFI (J)
China
NECSA (ZA)



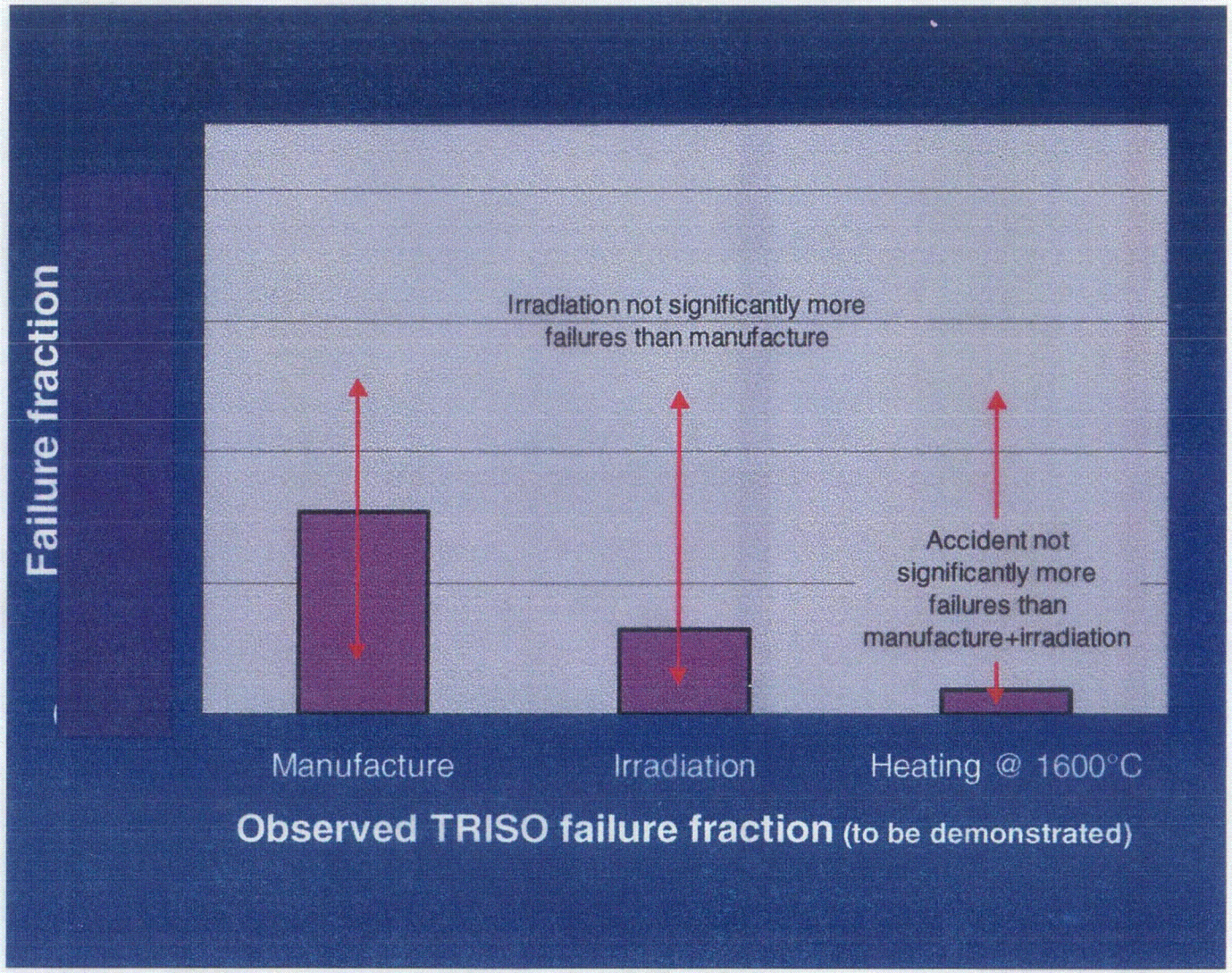
Worldwide History of HTR Fuel Fabrication

Reactor/ Manufacturer	Fuel Description	Total HM (kg)
ROVER/GA LANL	BISO in rods	1
Peach Bottom/ GA	BISO in compacts	3,500
UHTREX/GA LANL	BISO in compacts	200
DRAGON	TRISO, BISO compacts	300
FSV/ GA	TRISO in compacts	33,400
THTR/ NUKEM	BISO in spheres	11,000
AVR/ NUKEM	HEU BISO, TRISO spheres	1,700
AVR/ NUKEM	Modern LEU TRISO spheres	480
US development GA	Modern TRISO UCO	500
HTTR/ NFI	Modern LEU TRISO compacts	900
HTR-10/ INET	Modern LEU TRISO spheres	135

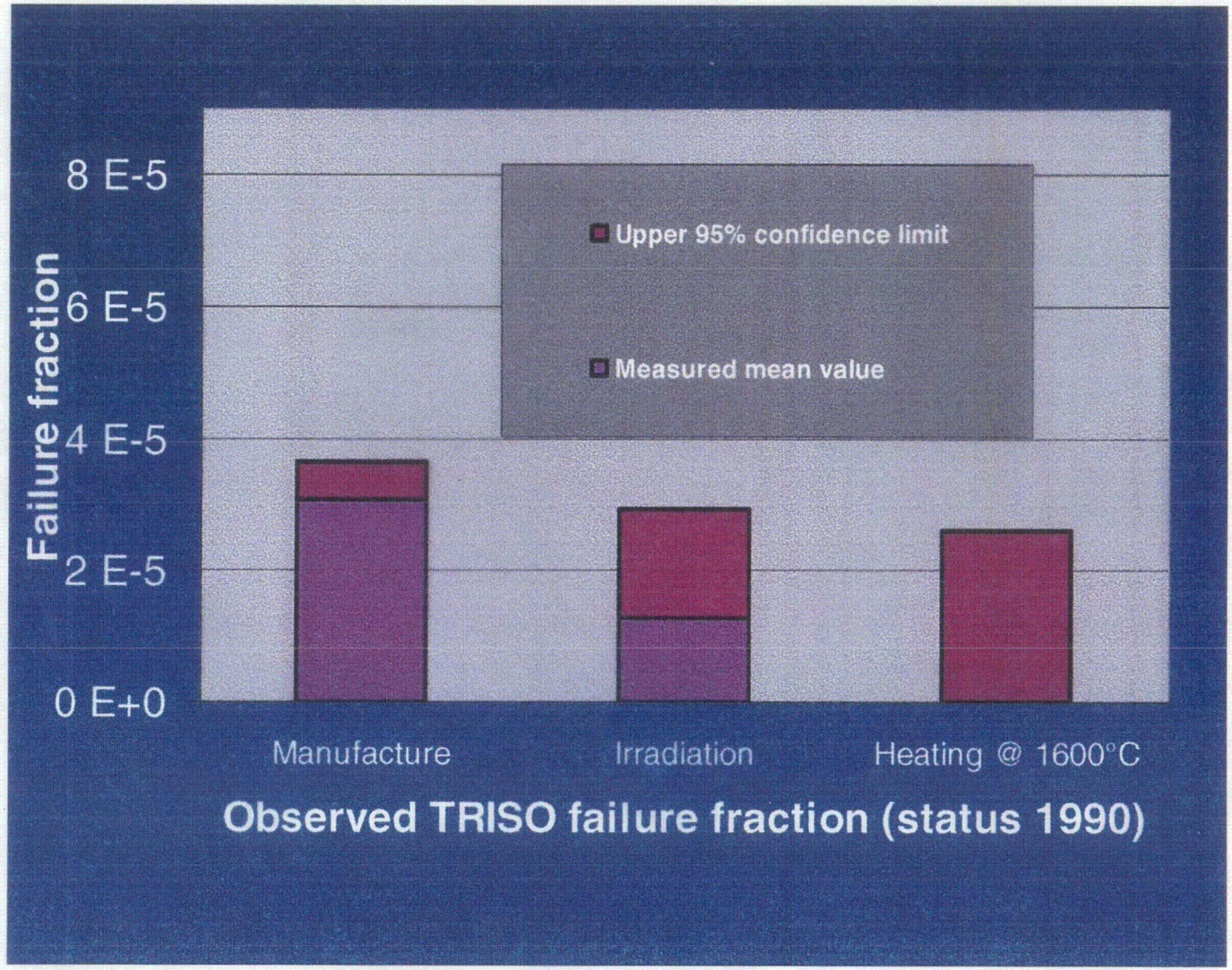


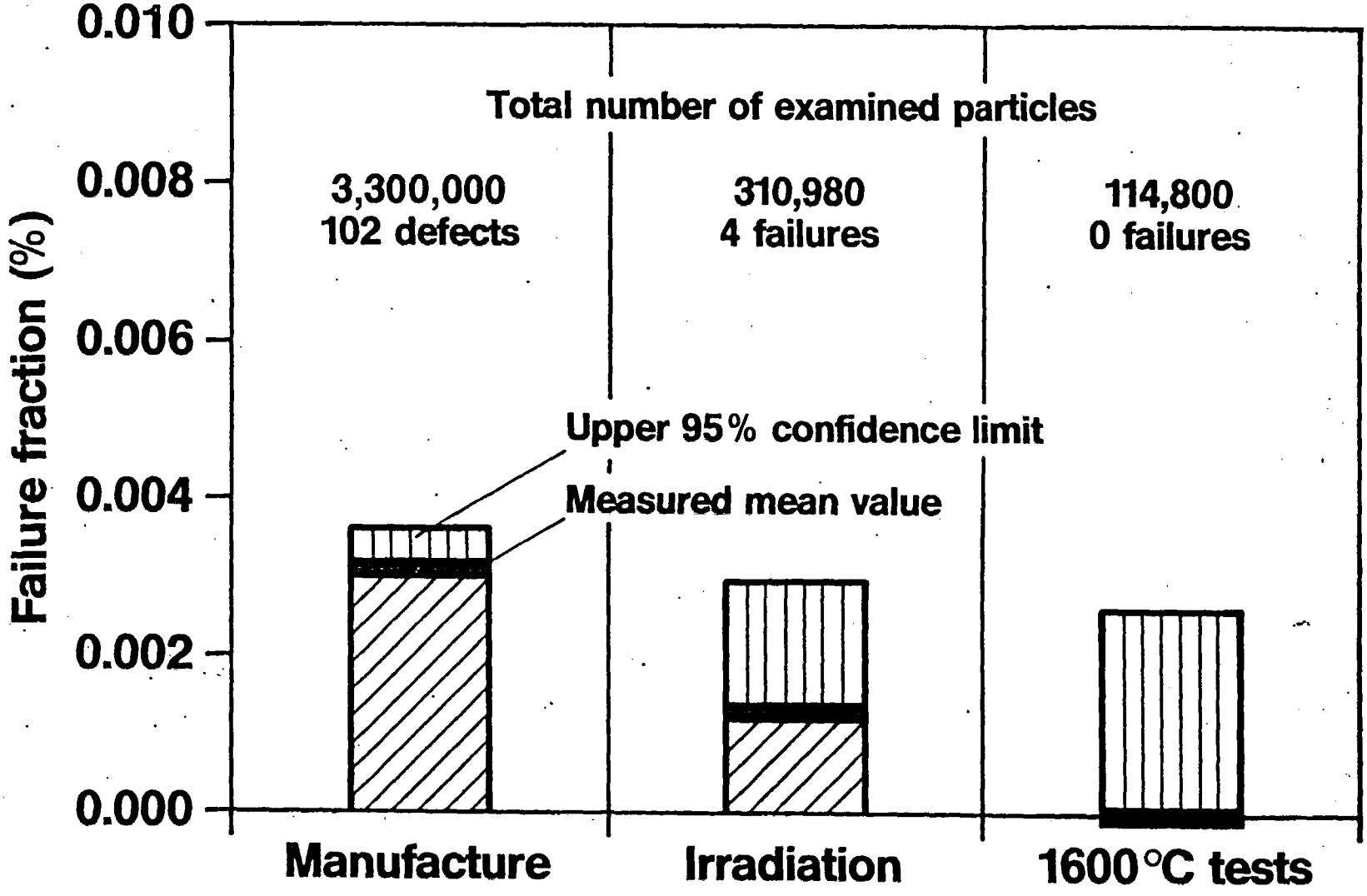
Varying goals in the German fuel development program have also led to a steady increase in the coated particle quality. The high enriched $(Th,U)O_2$ fuels were used in AVR and THTR and – with a TRISO coating – were also qualified for PNP and HHT. Latest development was UO_2 TRISO for the MODUL reactor with demonstration of fission product retention in all normal and off-normal conditions

Target philosophy

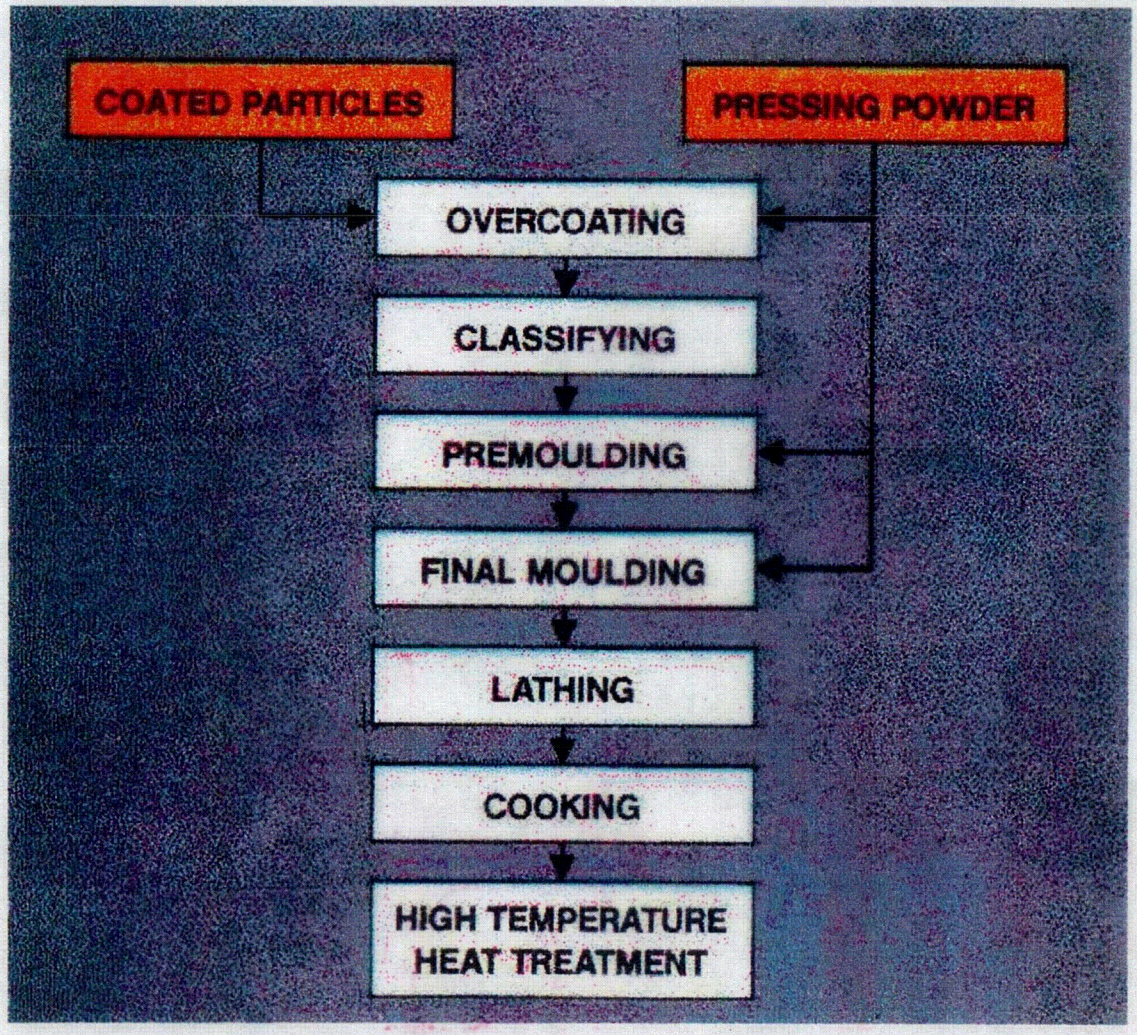


Desirable properties have been achieved and have been demonstrated
(here HEU and LEU TRISO up to 1990)



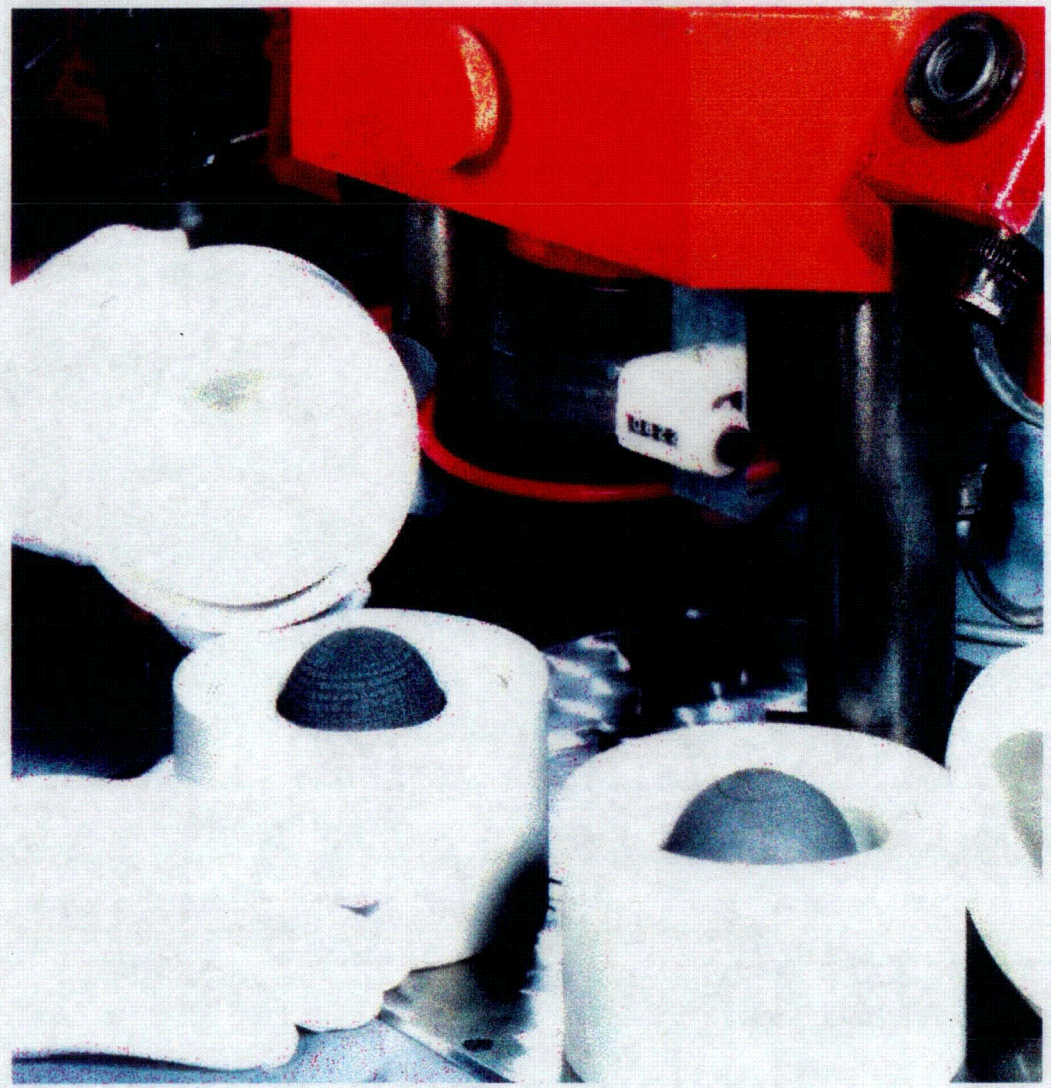

0.010
0.008
0.006
0.004
0.002
0.000
Failure fraction (%)
Total number of examined particles
3,300,000
102 defects
310,980
4 failures
114,800
0 failures
Upper 95% confidence limit
Measured mean value
Manufacture
Irradiation
1600°C tests

Fuel Element Manufacturing Process



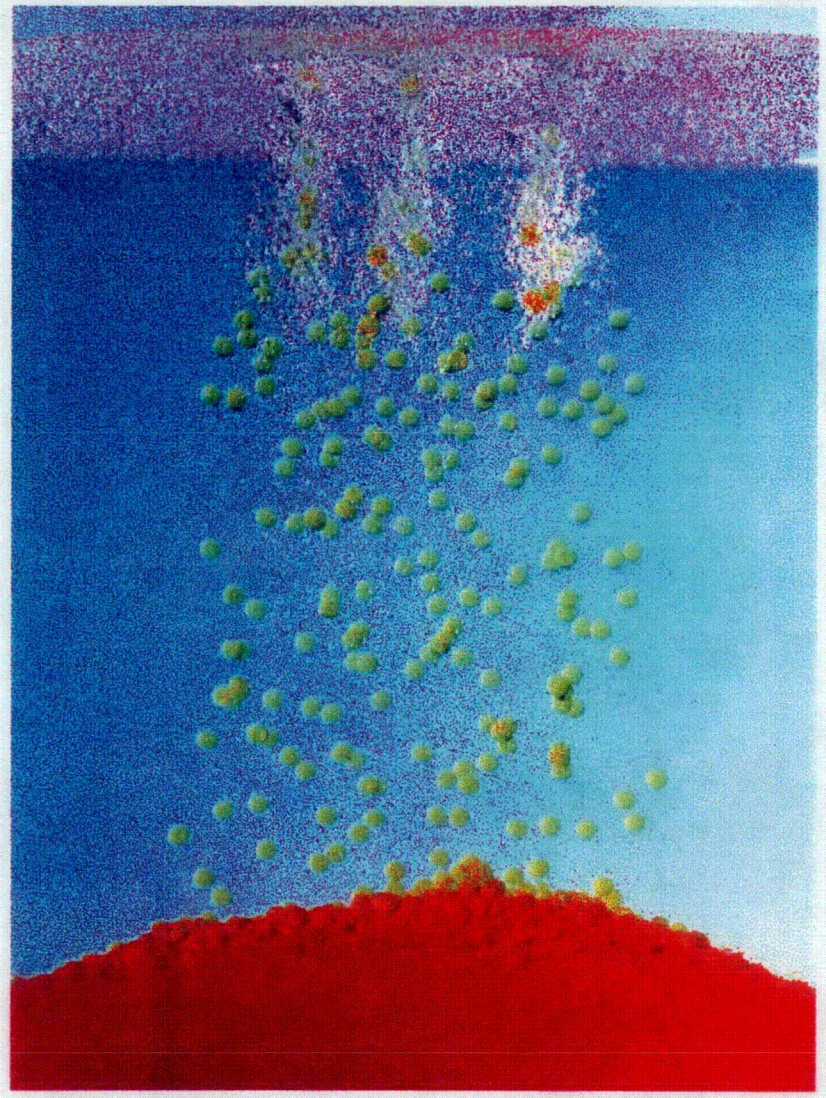
HTR-Fuel Element Fabrication

Fuel Element Manufacturing Process



Rubber Dies for
Fuel Spheres

Kernel Manufacture



Kernels in the Precipitating Agent

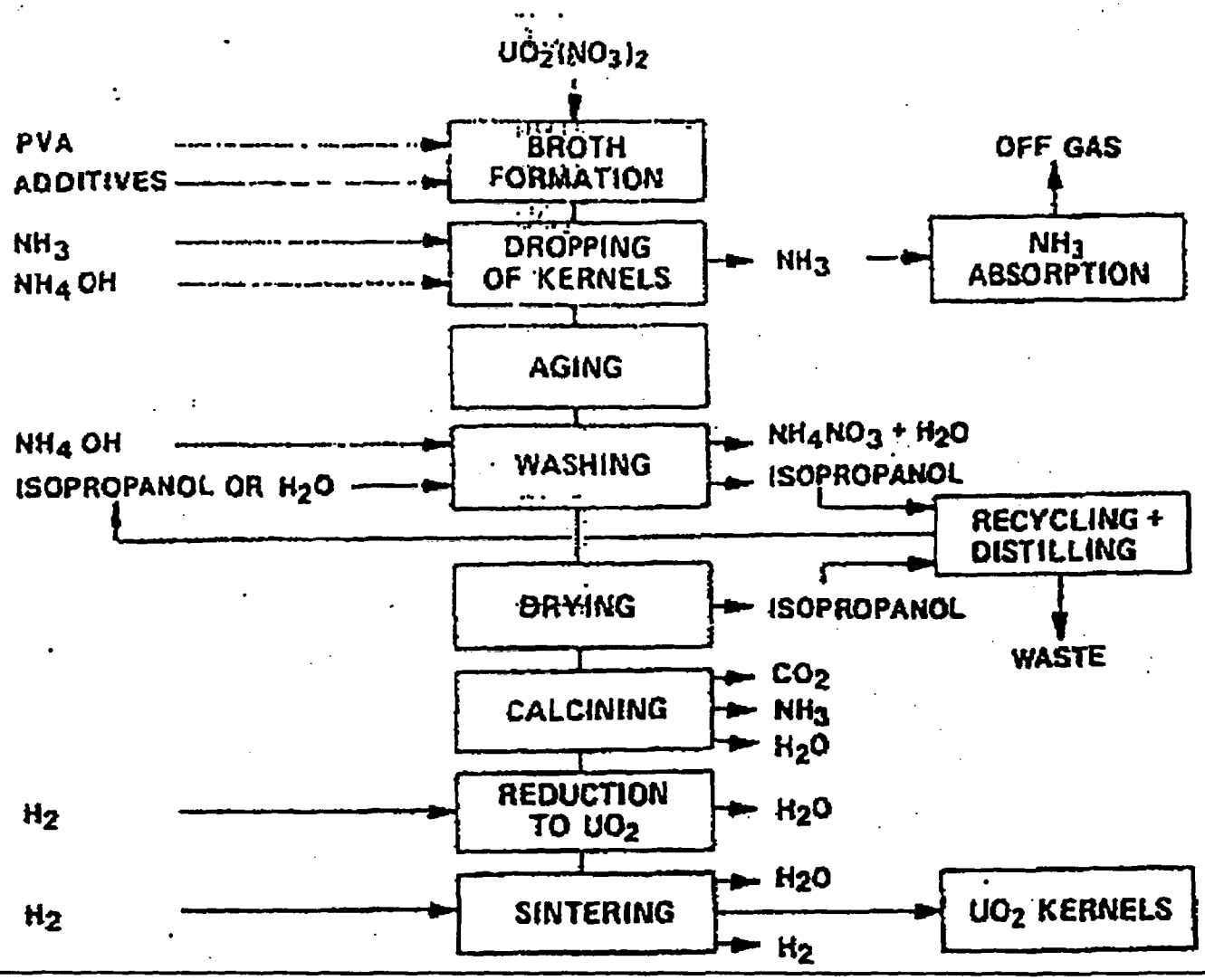
HTR Fuel Specific Characteristics

Kernels:

- diameter
- roundness

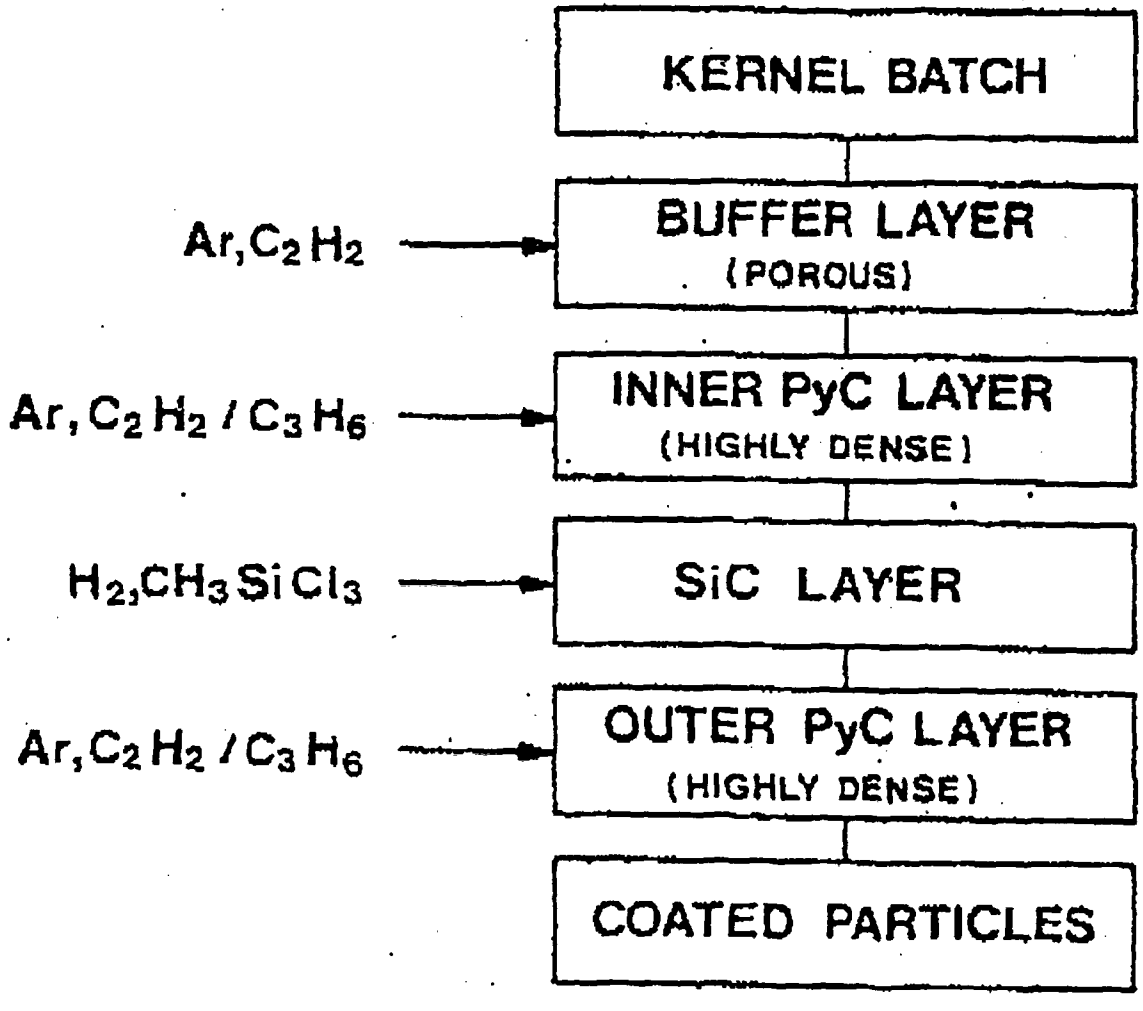
Coated Particles:

- ratio of defect SiC-layers
- diameter
- roundness
- thickness of each layer
- density of each layer
- anisotropy of both dense pyrocarbon layers

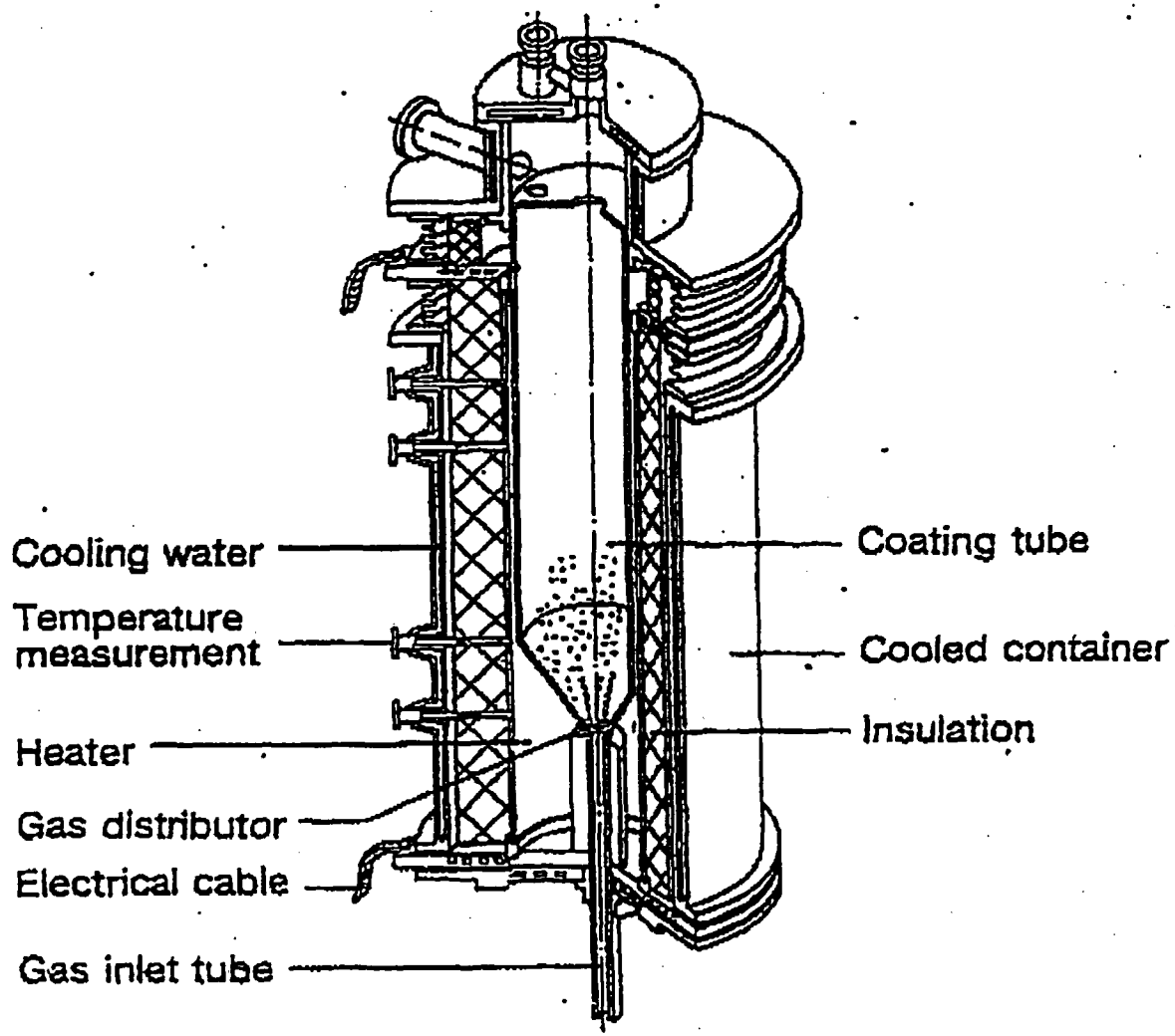


UO_2 kernel formation

Heinz Nabielek, Karl Verfondern



Coating of HTR fuel particles



Fluidized bed coating furnace

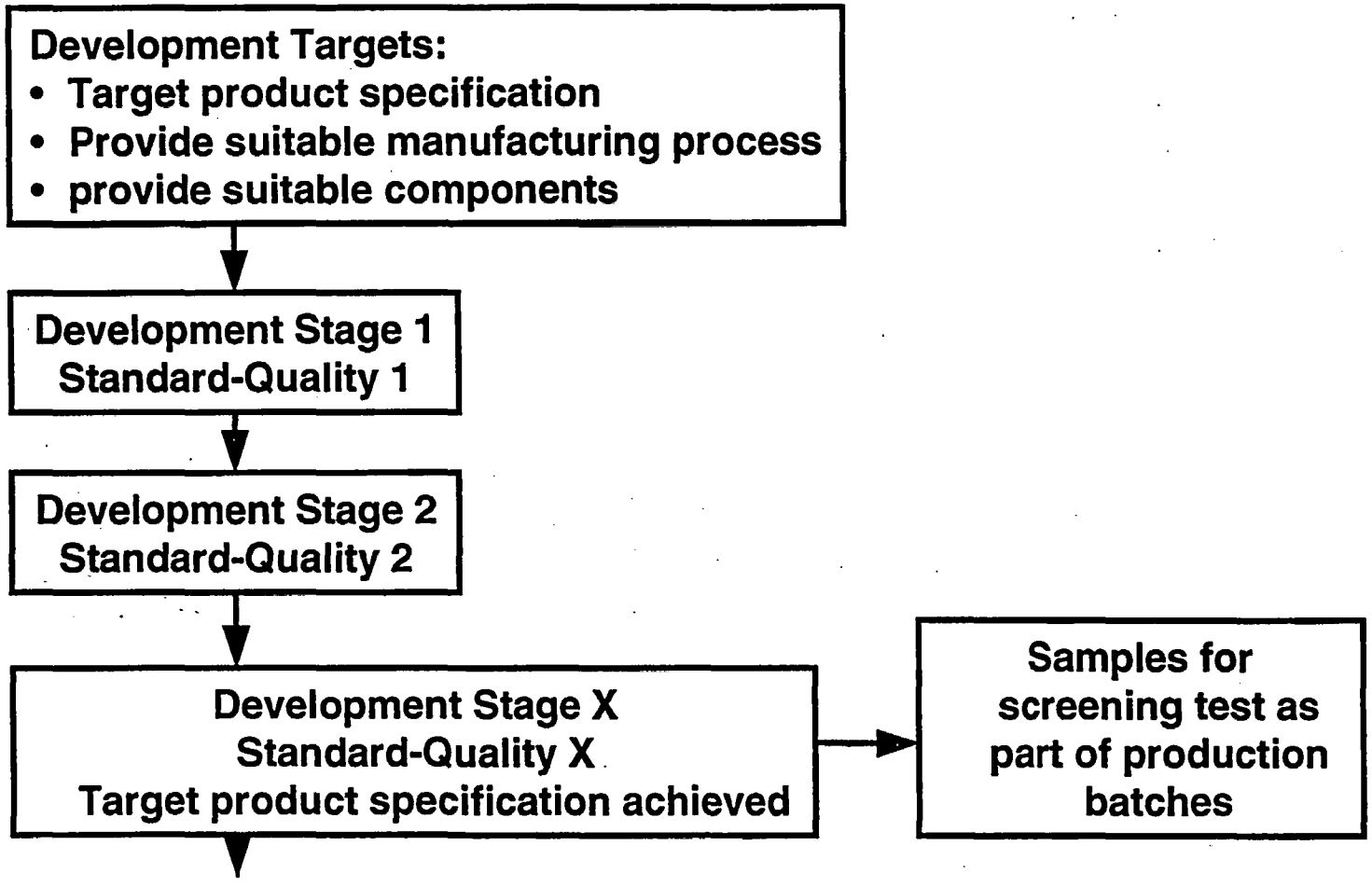
Heinz Nabielek, Karl Verfondern

Special Quality Assurance System and Philosophy

- Target product Specification achieved, demonstrated by a "final" standard-quality
- essential targets for process and components achieved (close to the future production conditions)

→ Sample for reference test as part of the standard quality

Special Quality Assurance System and Philosophy



		Pre - 1985 production			Post - 1985 production	
		AVR 19	LEU Phase 1	AVR 21-1	AVR 21-2	"Prooftest"
Experiment Particle batch		HT 232-298	HFR-K3 FRJ2-K13 EUO 2308	HT 354-383		HFR-K5/K6 EUO 2358-2365
Kernel composition		UO2	UO2	UO2	UO2	UO2
Kernel diameter	µm	500	497	501	502	508
Kernel density	Mg m-3	10.80	10.81	10.85	10.87	10.72
Thickness of coating						
Buffer layer	µm	93	94	92	92	102
Inner PyC layer	µm	38	41	38	40	39
Sic layer	µm	35	36	33	35	36
Outer PyC layer	µm	40	40	41	40	38
Density of coating	Mg m-3					
Buffer layer		1.01	1.00	1.01	1.1	1.02
Inner PyC layer		1.86	~1.9	1.9	1.9	1.92
Sic layer		3.19	3.20	3.20	3.2	3.20
Outer PyC layer		1.89	1.88	1.88	1.9	1.92

Modern UO_2 LEU Triso in Germany

Evaluation of free uranium and defective SiC layers in LEU TRISO fuel elements

Designation of fuel element (FE) population	AVR 19	AVR 21	AVR 21-2	LEU PHASE I	Proof test fuel
Production year	1981	1983	1985	1981	1988
Number of FE lots	14	11	8	–	–
Number of FEs produced	24,600	20,500	14,000	<100	< 200
Type of fuel	LEU UO_2	LEU UO_2	LEU UO_2	LEU UO_2	LEU UO_2
^{235}U enrichment	9.8%	16.7%	16.7%	9.8%	10.6%
Coating batch size	5 kg	3 kg	3 kg	5 kg	5 kg
Number of coating batches	65	54	29	1	8
Number of particle sets	4	2	3	–	1
Number of particles/FE	16,400	9,560	9,560	16,400	14,600
One particle uranium equivalent (in parts per million)	61	105	105	61	68
Evaluation of free uranium from burn-leach measurements					
Mean value (in parts per million)	50.7	43.2	7.8	35.0	13.5
Number of FEs tested in burn-leach	70	55	40	5	10
Number of FEs with 0 particle defects	31	42	38	3	8
Number of FEs with 1 particle defect	26	8	1	1	1
Number of FEs with 2 particle defects	9	2	1	1	1
Number of FEs with 3 particle defects	4	2	0	0	0
Number of FEs with 4 particle defects	0	0	0	0	0
Number of FEs with 5 particle defects	0	0	0	0	0
Number of FEs with 6 particle defects	0	1	0	0	0
Number of FEs with ≥ 7 particle defects	0	0	0	0	0

HTR Fuel Specific Characteristics

Fuel Element:

- heat conductivity of the graphite matrix at 25 °C and 1000 °C
- ratio of defective SiC-layers (burn-leach test)
- corrosion rate
- crushing strengths
- fuel-free zone thickness
- abrasion rate

THTR-Fuel Element Production Experience

Produced:

Kernels:	~ 1000 batches
Coated particles:	~ 4000 batches
Fuel element:	~ 500 lots
	(~ 1.000.000 FE)

Yield:

For each of these products > 95%

Reject:

1 lot of coated particles

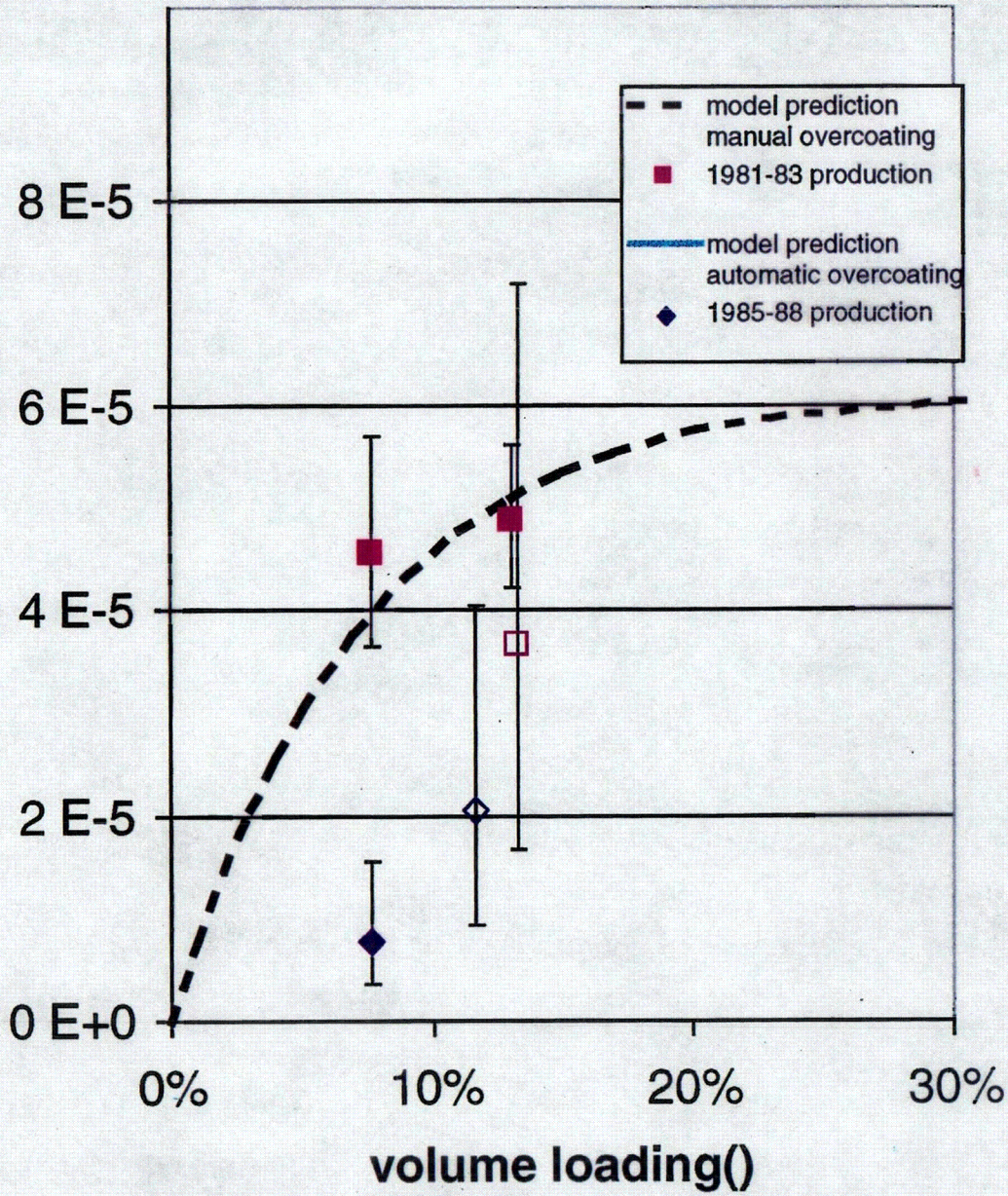
1 lot of fuel elements

Safety:

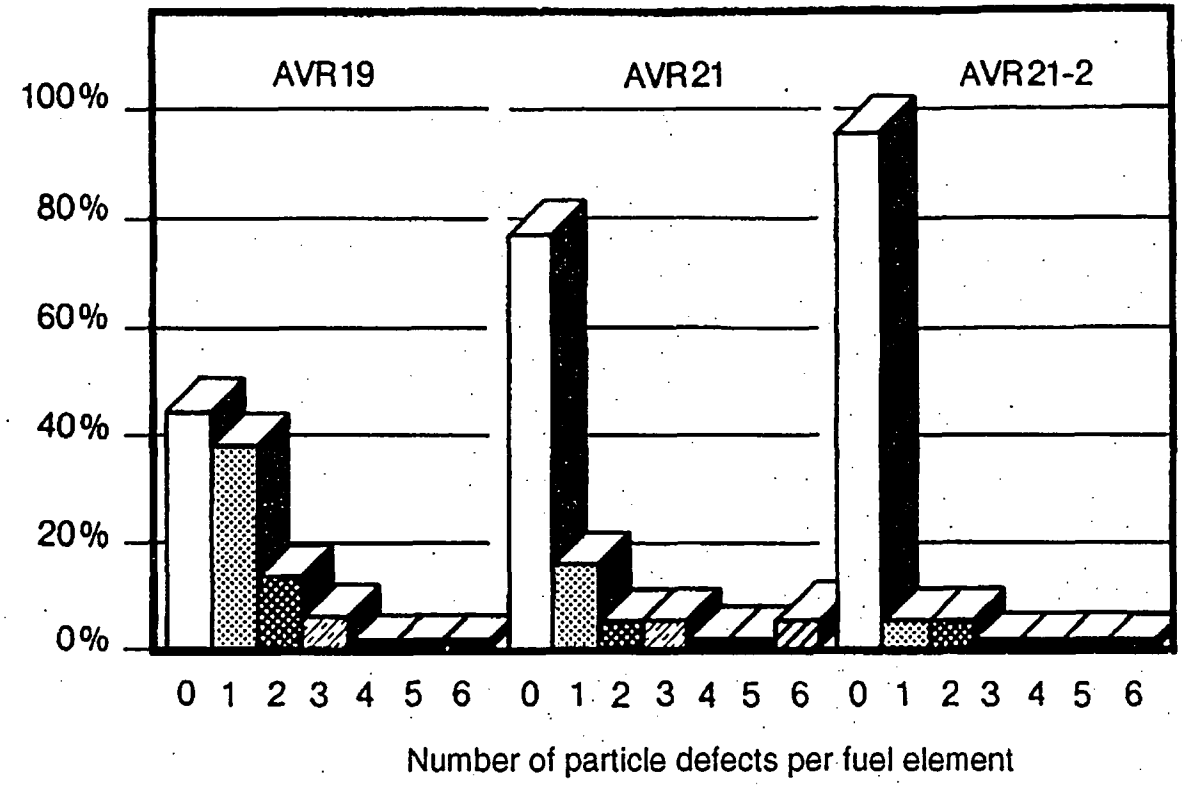
Not a single safety relevant incident

Design Parameter	HEU	LEU
Coated Particles		
Kernel Composition	$(Th,U)\ O_2$	UO_2
Kernel Diameter µm	500	500
Coating Layer Thickness µm	95/40/35/35	95/40/35/35
Coating Layer Sequence	Buffer/PyC/SiC/PyC	Buffer/PyC/SiC/PyC
Fuel Element		
Heavy Metal Loading	11	8-12
U 235 Enrichment	93 %	7-13 %
No. Particles per Element	19,000	10,000-20,000
Volume Loading of Particles	13 %	10-15 %
Operating Requirements		
Mean Operating Time d	1100-1500	700
Max. Burnup MWd/t_{HM}	120,000	90,000
Max. Fluence [E>0.1 MeV]	$4.5 \times 10^{25}\ m^{-2}$	$3.3 \times 10^{25}\ m^{-2}$
Max. Fuel Temperature °C	1020	1030
Max Power/Element kW	2.7	4.1

Free U fraction from TRISO defects

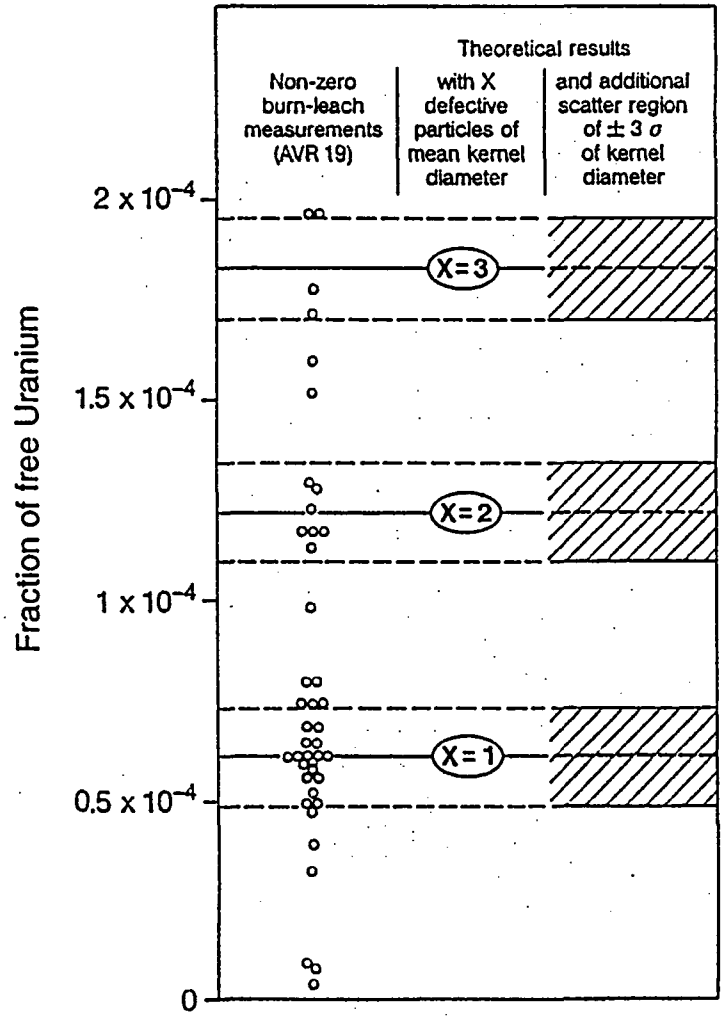


- Fraction of LEU UO_2 Triso defects during cold isostatic sphere pressing as a function of particle volume loading.
- Data are obtained by burn-leach and correspond to "SiC failure fraction" in US and Japanese terminology.
- All results on NUKEM fuel elements are below $6x10^{-5}$.



Coated particle defects during sphere manufacture are rare random events. The frequency of spheres with zero defects has steadily improved during the years 1981-1985.
Nucl. Eng. Des. 121/2 (1990)

Diagram shows non-zero free uranium measurements in the seventy burn-leach tests from NUKEM quality control of the AVR 19 (GLE 3) production of 24,600 spherical fuel elements for AVR. This is a destructive test on 5 FEs per lot from the 14 lots in this production.



Measured free uranium corresponds to the contents of an integer number of coated particles; here zero, one, two or three out of 16,400 particles in a sphere.

The burn-leach with spherical fuel elements consists of the following steps:

-burn graphite and oPyC at 800°C

-leach with HNO_3

-determine U in solution

HTR fuel: main criteria quantified ...

Manufacture

burn-leach on particles $< 1x10^{-6}$ *

burn-leach on fuel body $< 6x10^{-5}$

Irradiation

in-pile R/B $<< 1x10^{-6}$ *

PIE shows $F(^{137}Cs) < 2x10^{-5}$

... and ... $F(^{110m}Ag) < 2x10^{-3}$

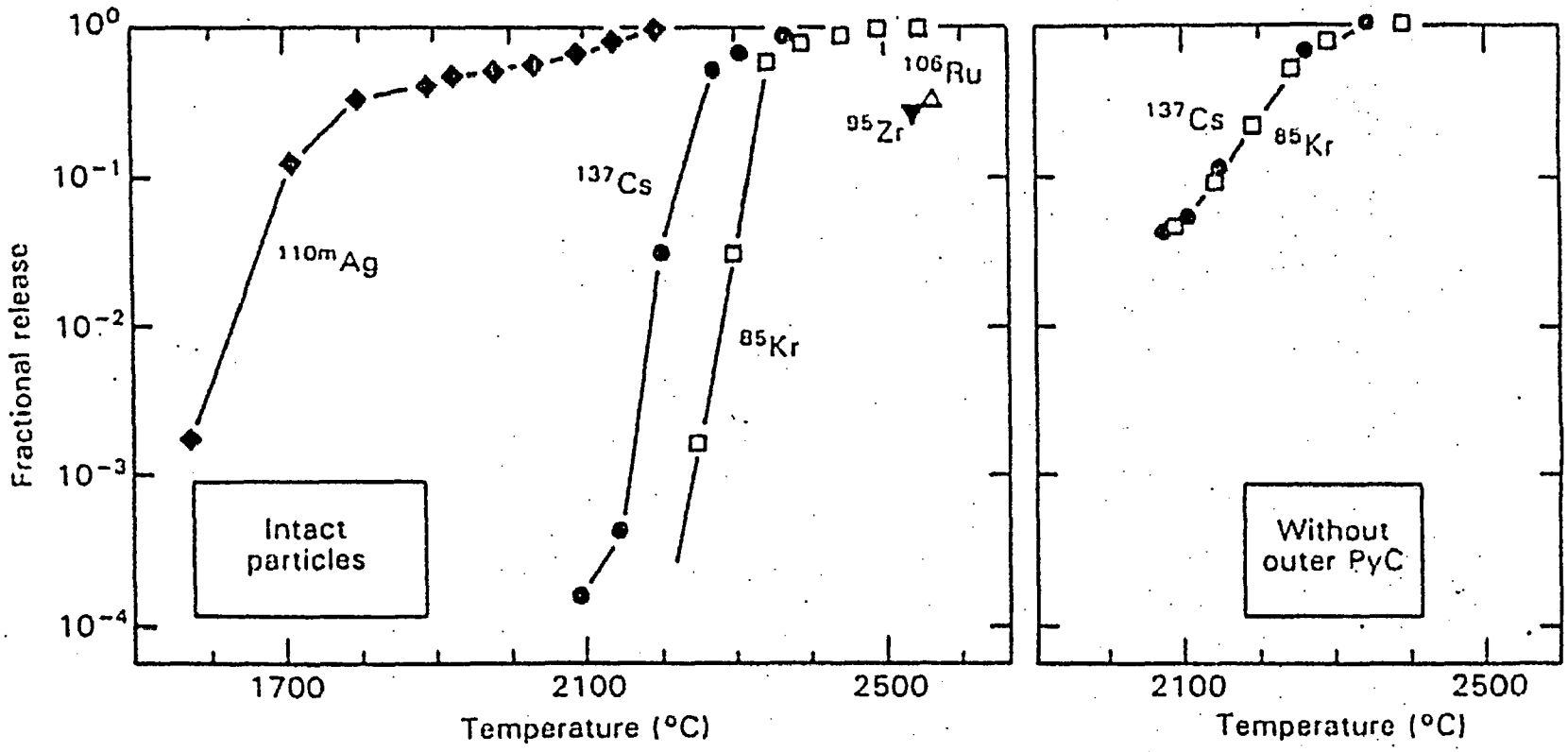
Heating

shows $F(^{85}Kr) < 2x10^{-6}$ *

and $F(^{137}Cs) < 2x10^{-4}$

* Necessary, but not sufficient

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.

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**

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

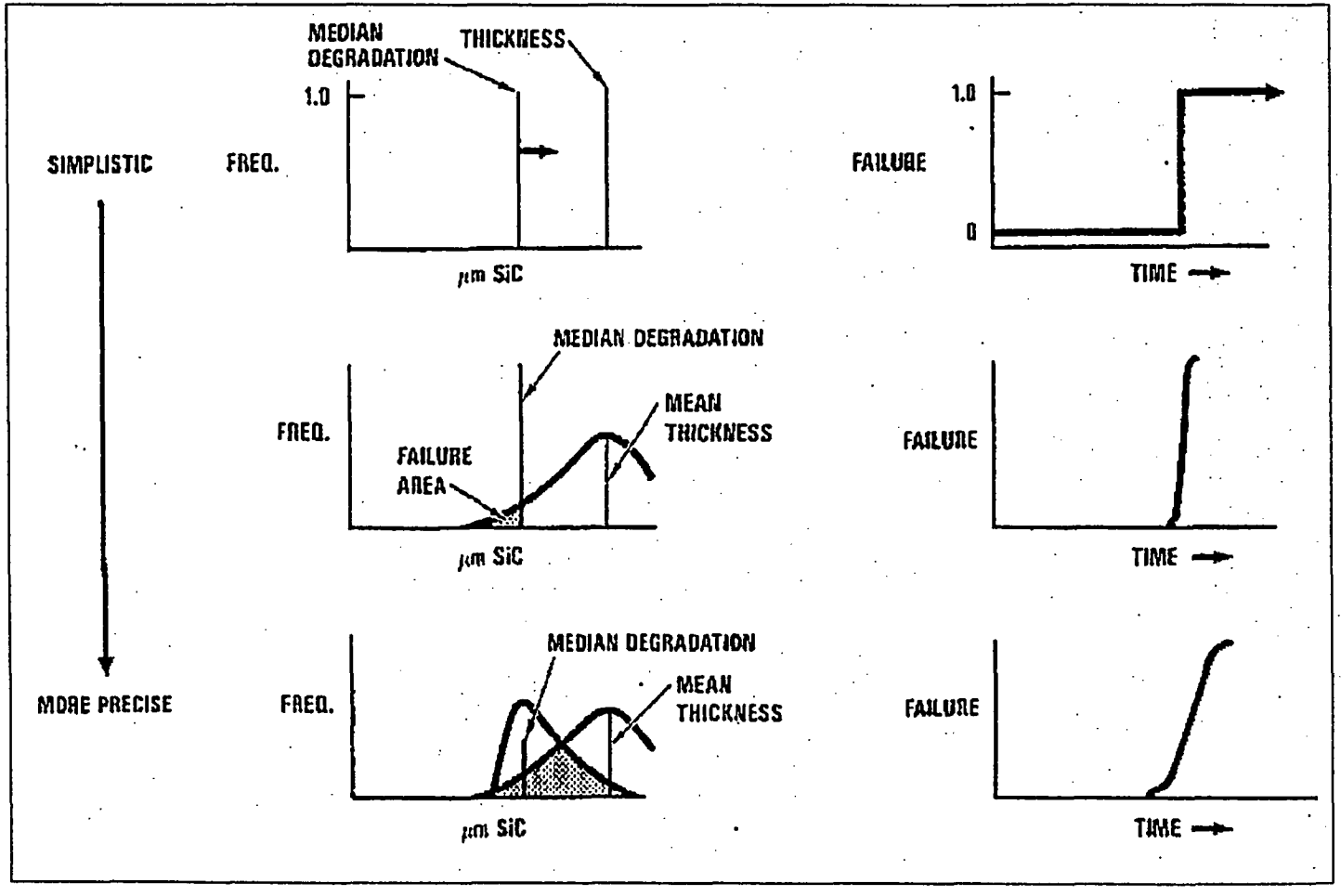
Φ... SiC failure fraction = Cs release fraction
t ... heating time
m... Weibull modulus

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

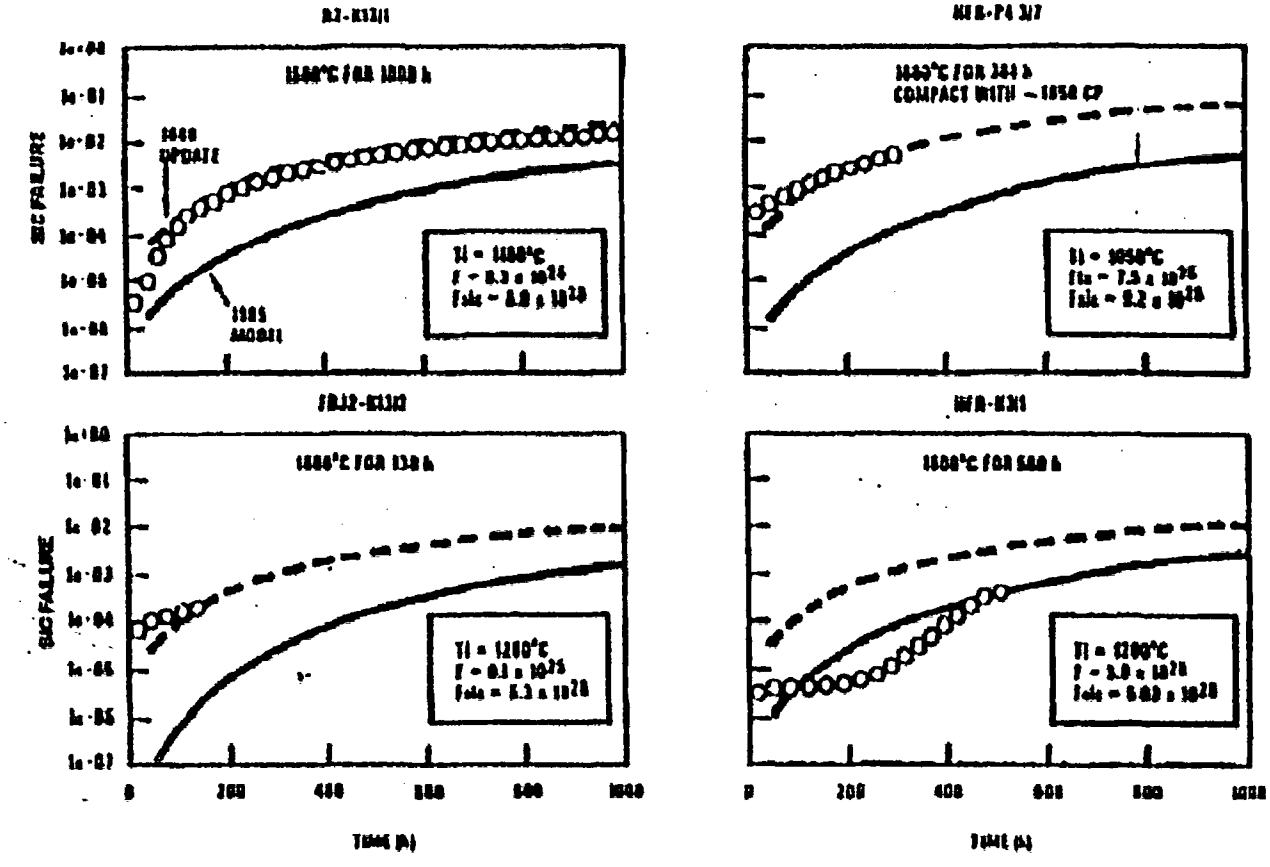
$$F_{Kr}(t) = \int_0^t \frac{d\Phi(t')}{dt'} F_{PyC}(t-t')\, dt'$$

F_{Kr} ... Kr release fraction from particles
F_{PyC}... Kr diffusion fraction through outer PyC

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.



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



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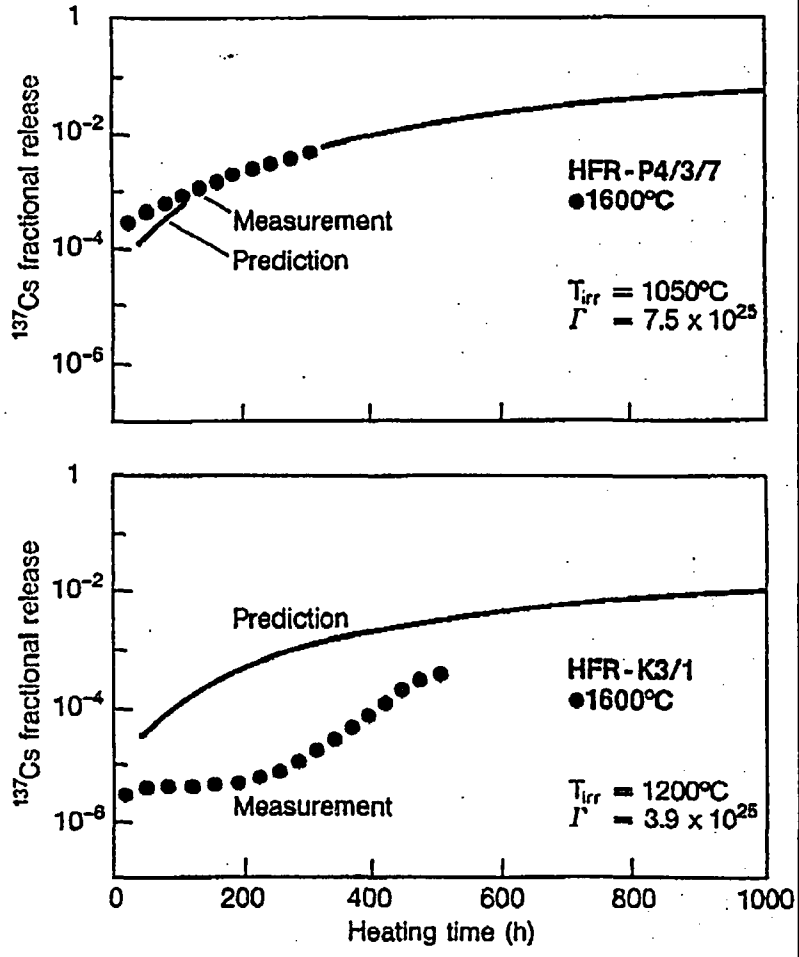
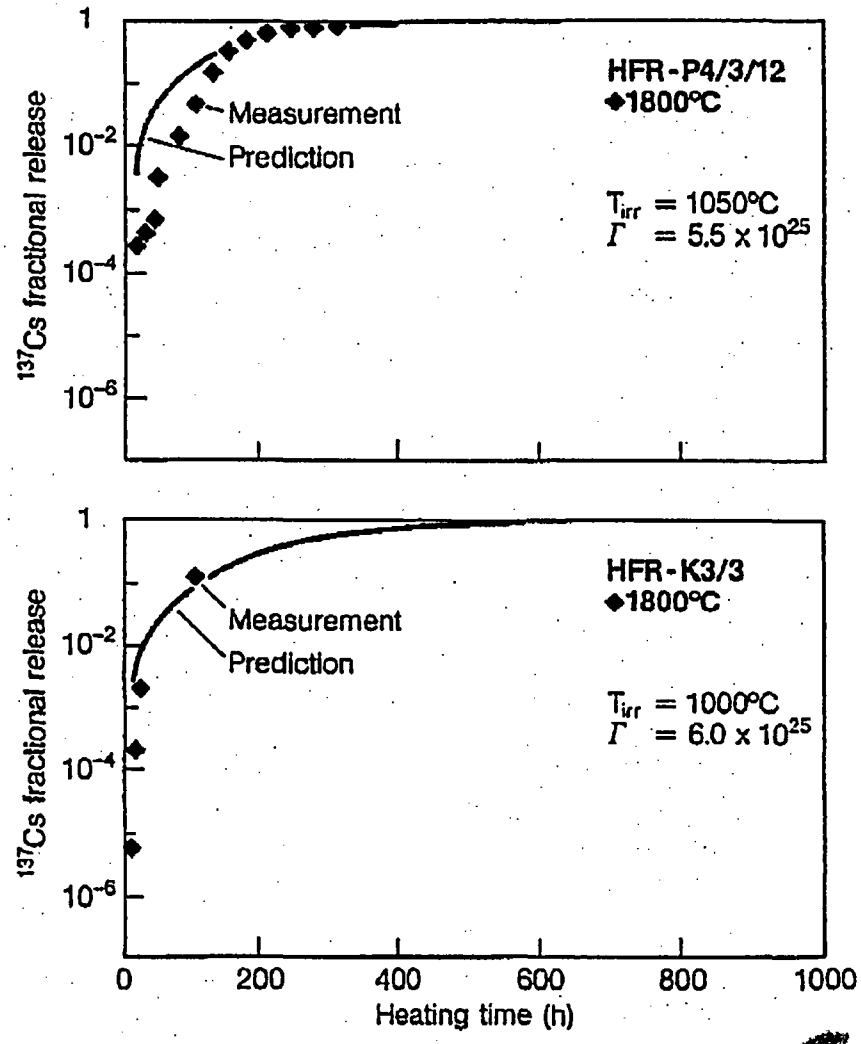
Goodin-Nabielek Approach with SiC Degradation 9

Fig. 90: ^{137}Cs release measurement by Schenk and prediction by Goodin for 1600°C heating tests (compact HFR-P4/3/7 and sphere HFR-K3/1)



^{137}Cs release measurement by Schenk and prediction by Goodin for 1800°C heating tests (compact HFR-P4/3/12 and sphere HFR-K3/3)



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Goodin-Nabielek Approach with SiC Degradation 10

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

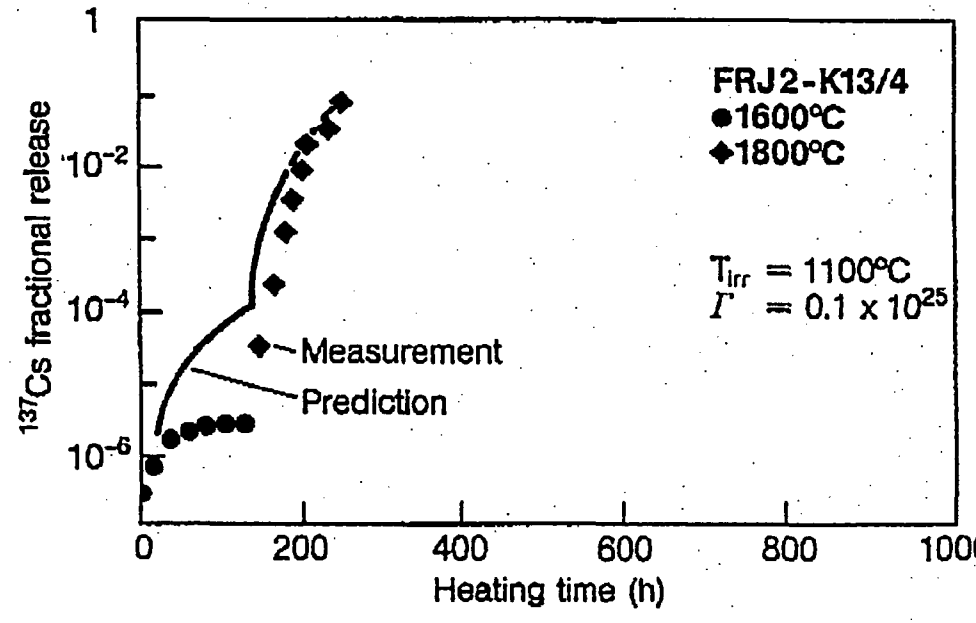
$$k_o(s^{-1}) = 5.03 \cdot 10^{-4} \times \left(\frac{\text{fission density}}{1 \cdot 10^{26}}\right)^{2.09} \times \left(\frac{\text{fast fluence}}{1 \cdot 10^{25}\,m^{-2}}\right)^{.041} \times \left(\frac{\text{irr. temp.}}{K}\right)^{4.14} \qquad (18)$$

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

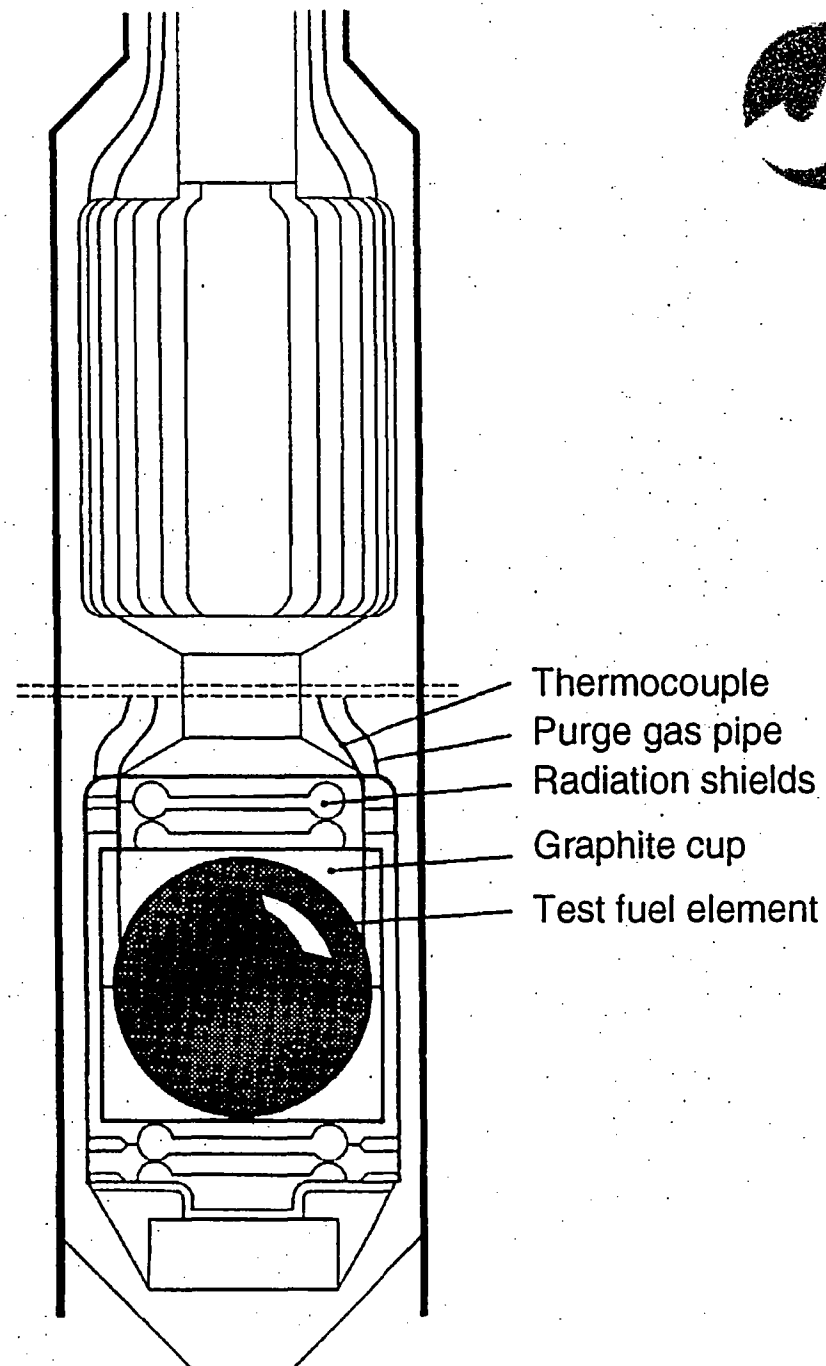
84 D. T. Goodin "US/FRG Accident condition fuel performance models", GA Doc. No. 908293 (DOE-HTGR-85107), Issue A, March 1989.

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-IB-2/90, June 1990.

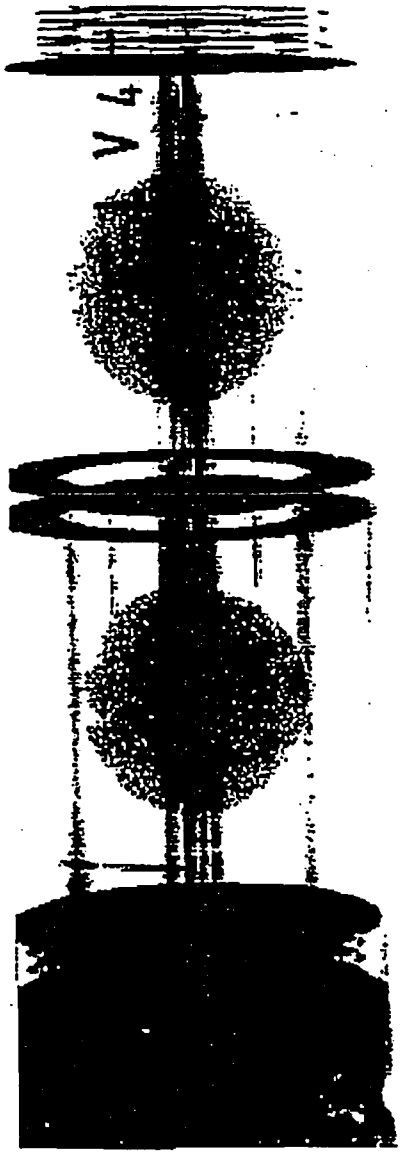
^{137}Cs release measurement by Schenk and prediction by Goodin for 1600/1800°C heating test of sphere FRJ2-K13/4.



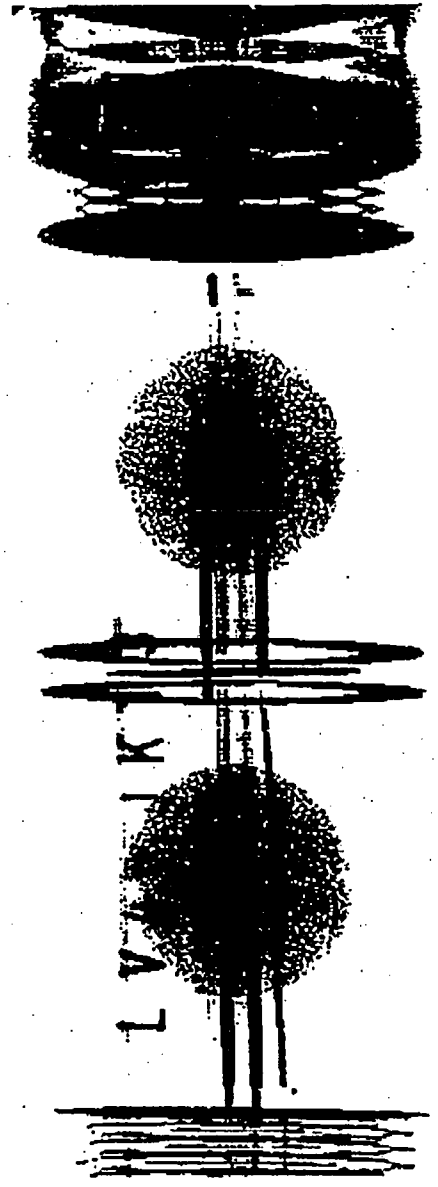
Every fuel element in an irradiation experiment is contained in an isolated cell. Temperatures are closely controlled by adjusting the He/Ne ratio in the purge gas which also allows the on-line determination of the fission gas release rate.



Capsule 1

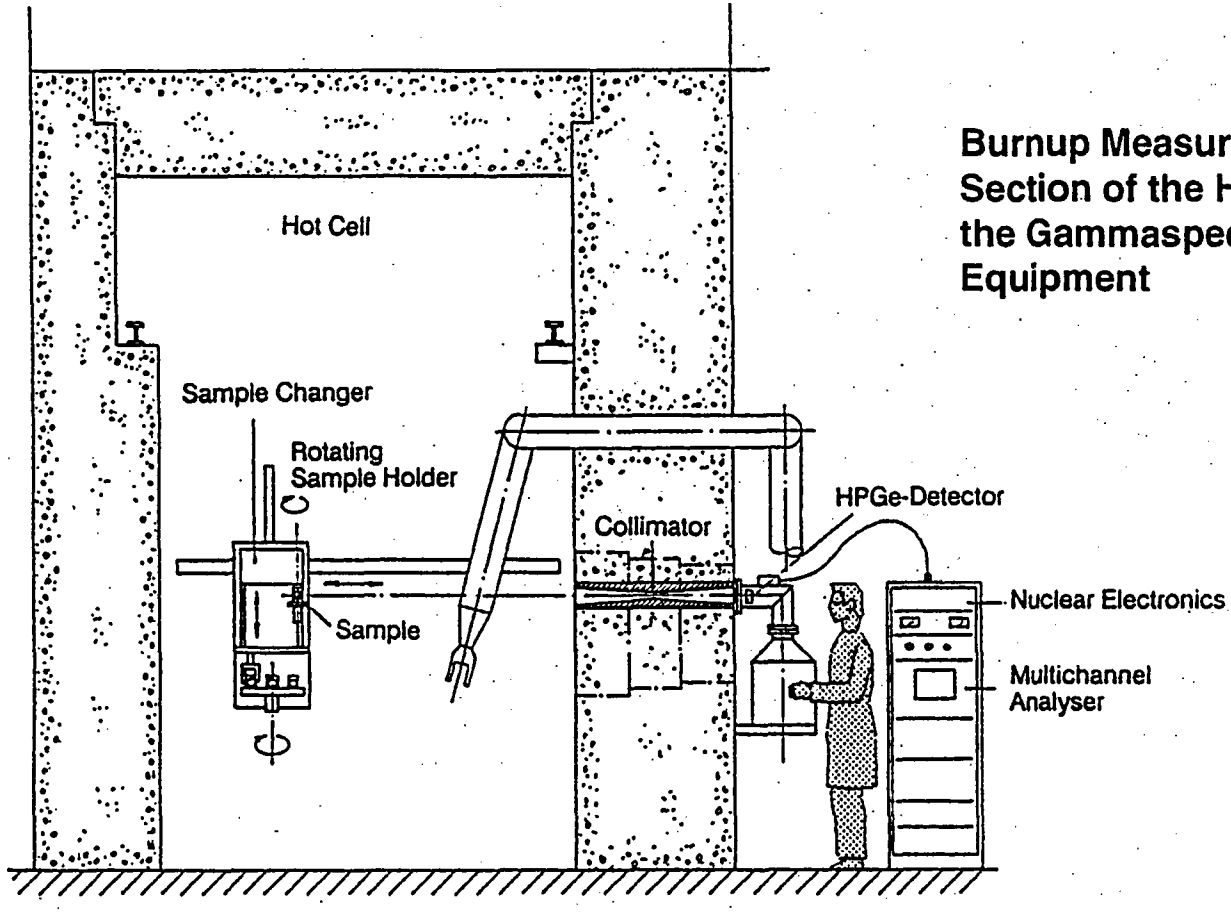


Capsule 2



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

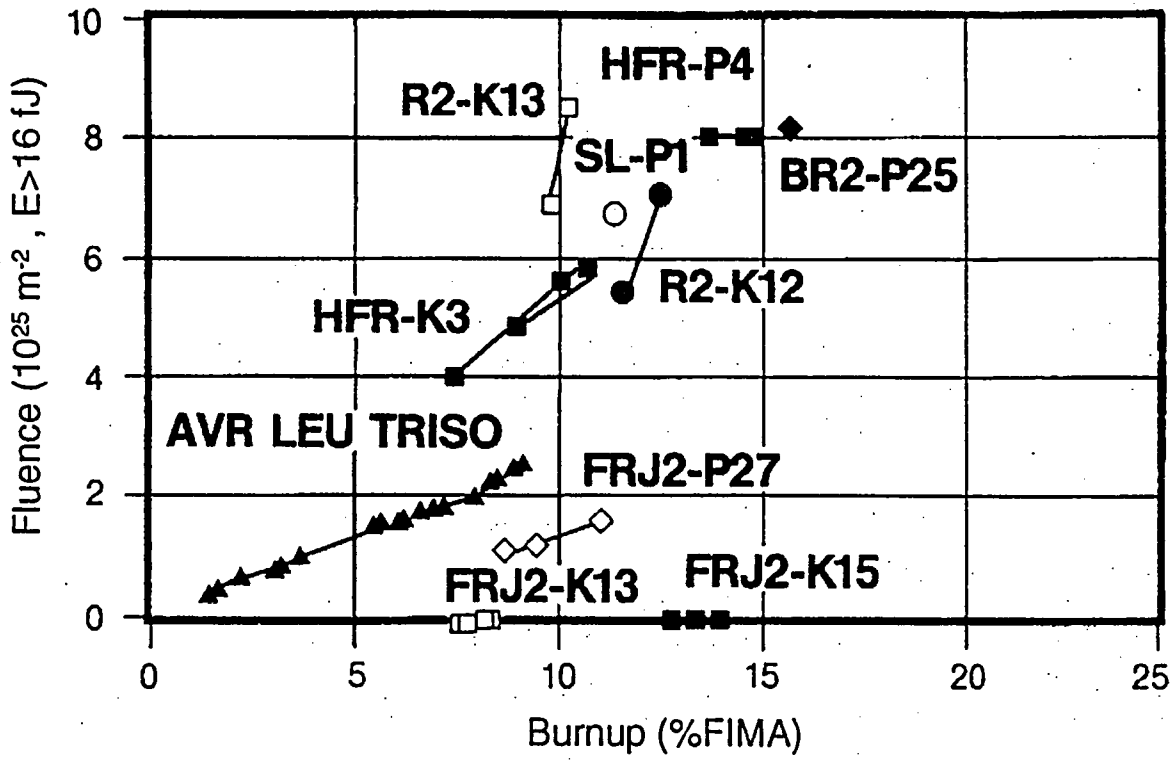
Processing of fuel elements in the Hot Cells, e.g. during gamma spectrometry for burnup determination



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

German Irradiation Experiments with HTR Fuel

RDD program	Old LEUs 1972-1976	HEU Program for PNP and HHT 1977-1981			LEU Program 1982-1993
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_xO_y Triso+ ThO_2 Triso	UO_2 TRISO
Test Goal					
Particle Performance	HFR-M5 DR-S6	BR2-P24	BR2-P25	BR2-P23	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-P25	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

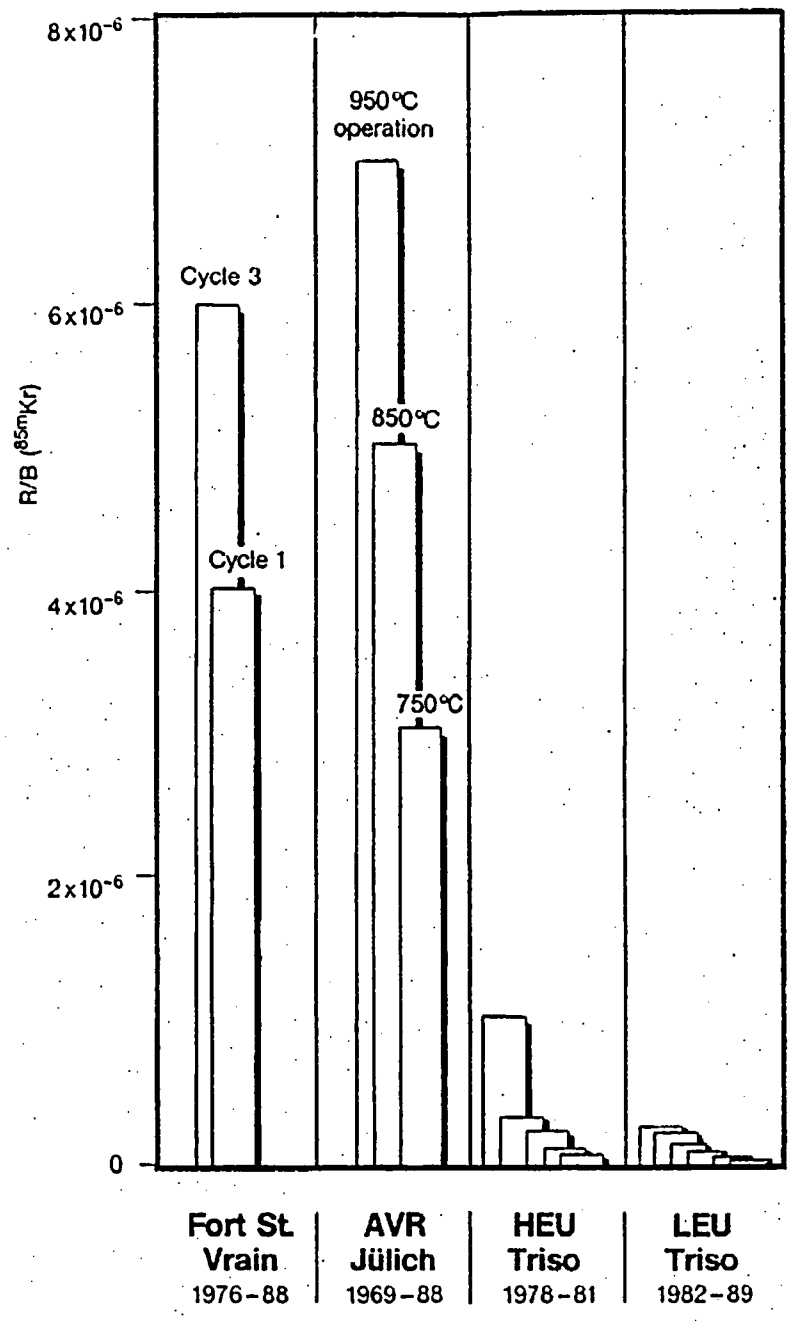


The main parameters in fuel irradiation testing are heavy metal burnup and accumulated fluence of fast neutrons. Irradiation temperature, perhaps the most important parameter of all, is usually around 1000° in the tests shown here.

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

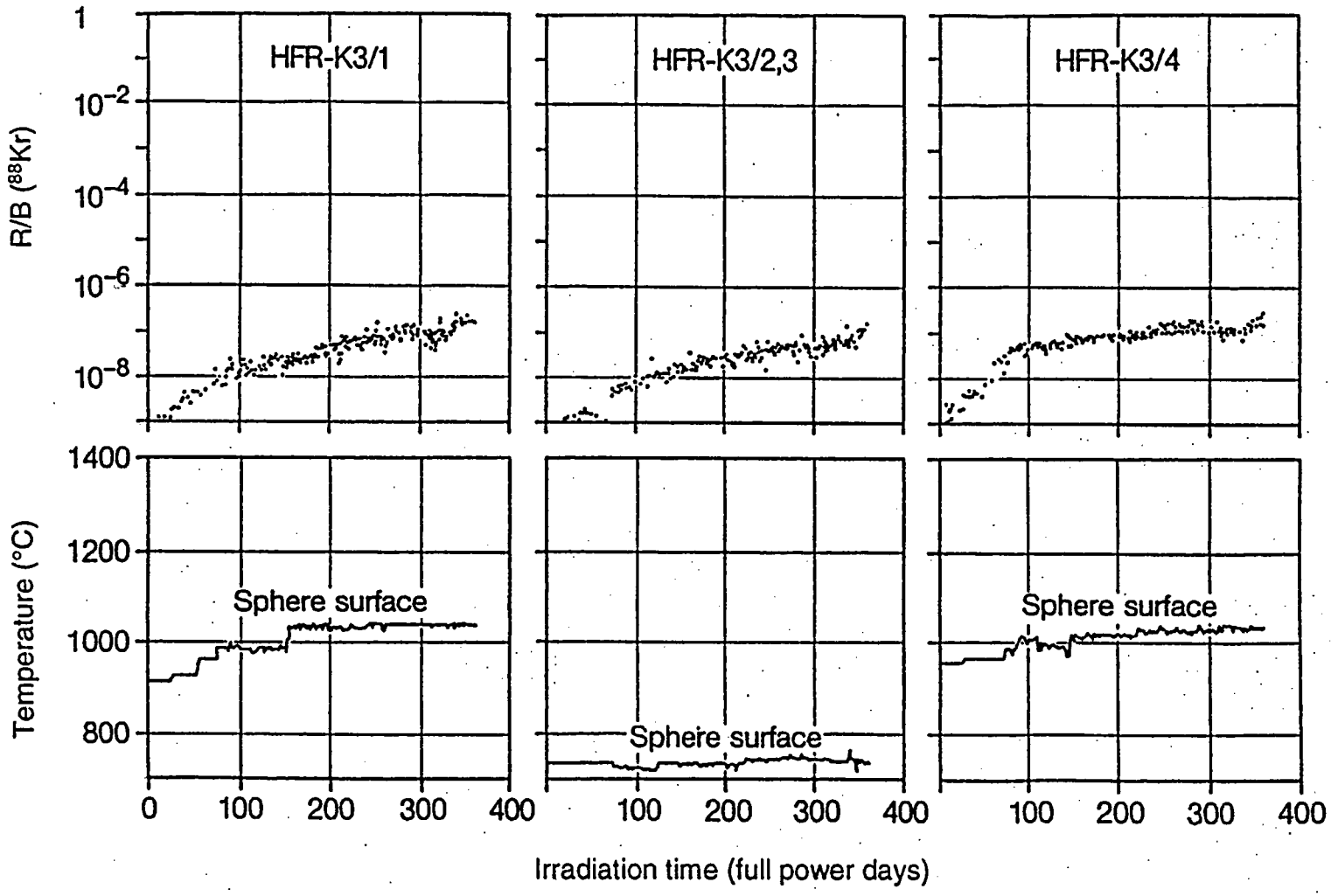
for fission products into the primary circuit of an HTR are:

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

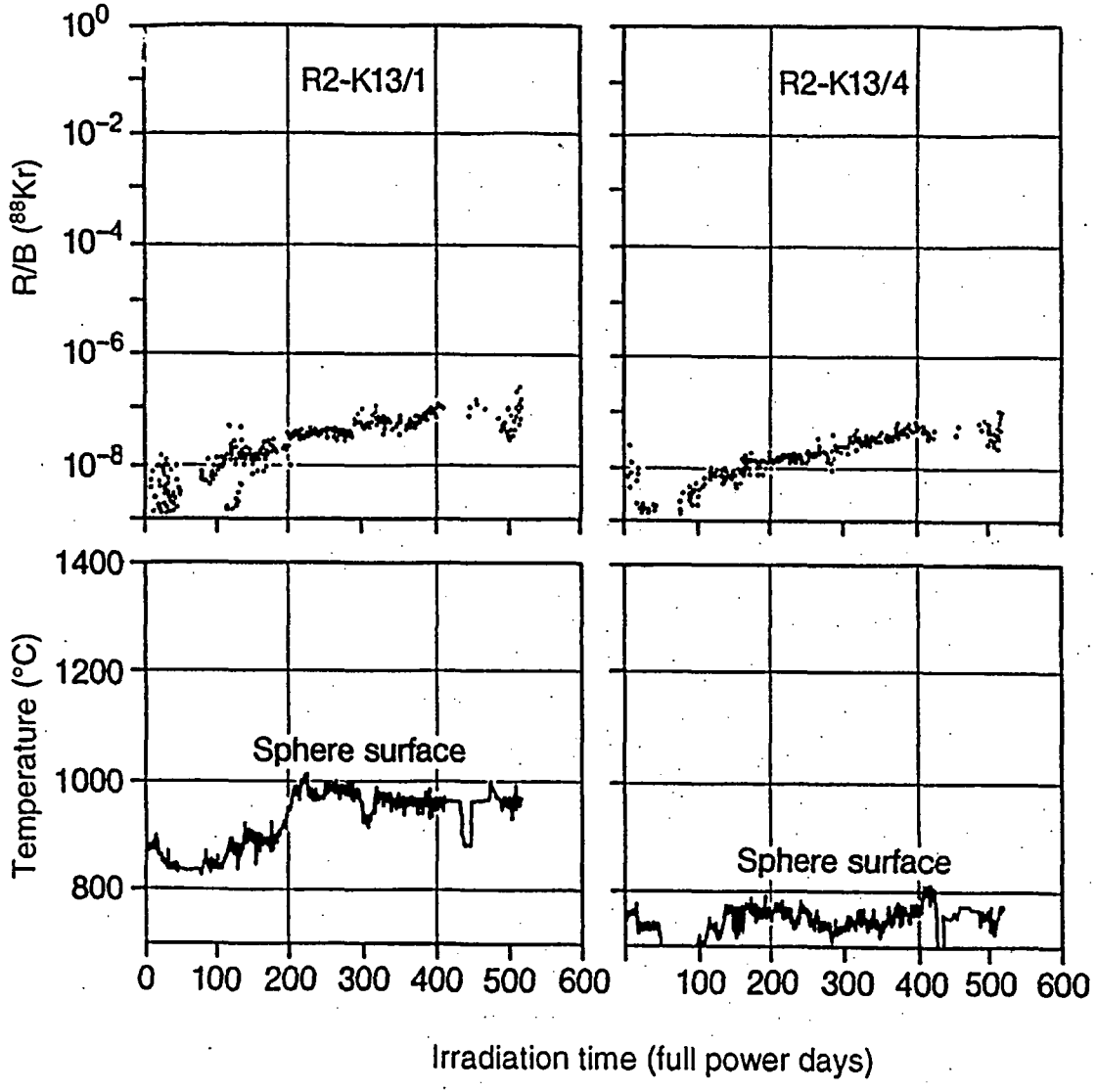
Sequence of release is

^{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

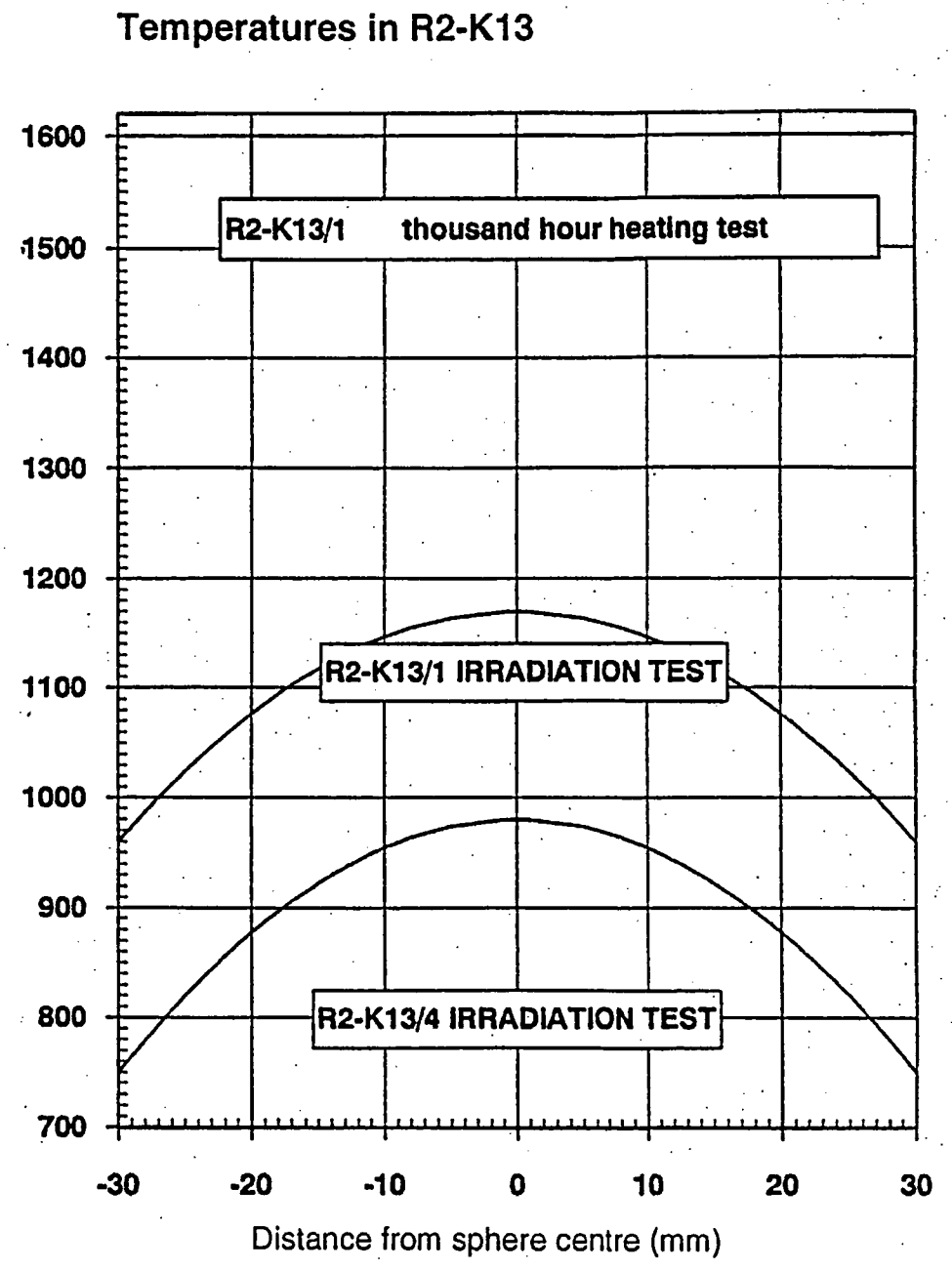


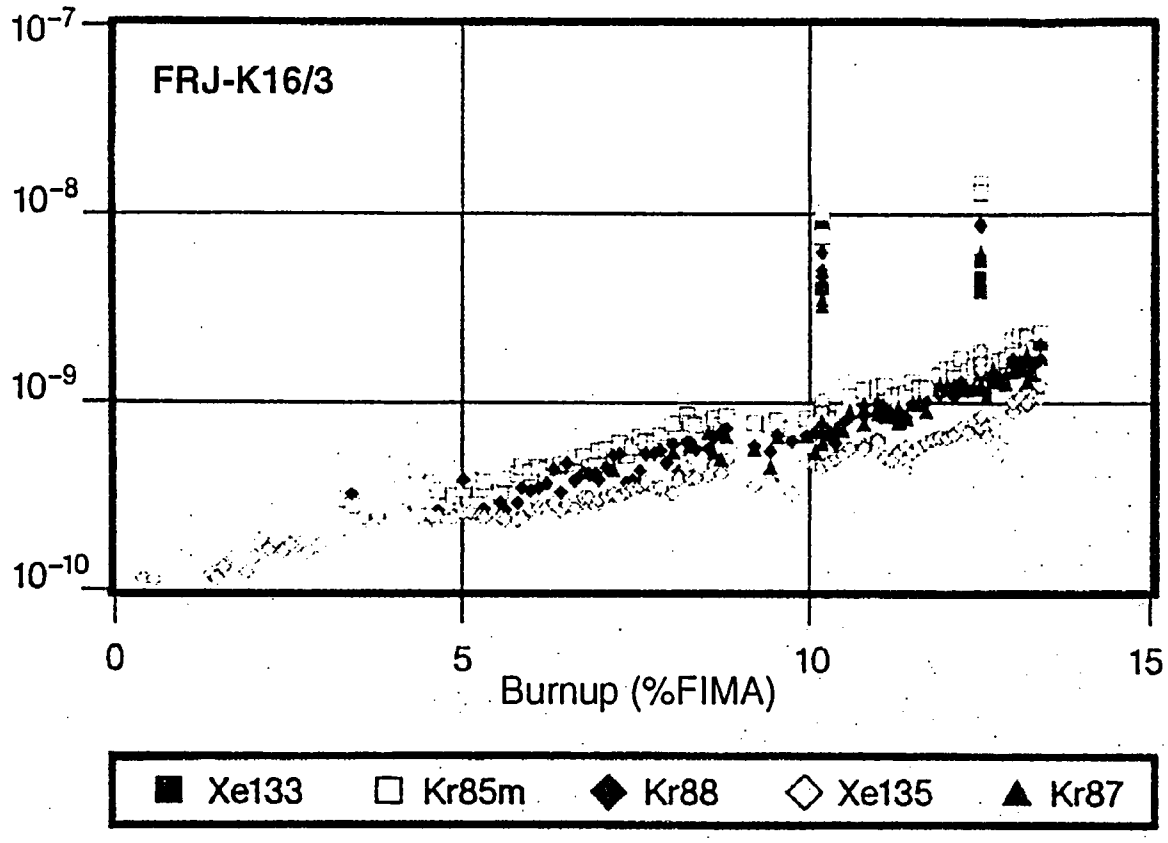
Gas release and temperature during irradiation of experiment HFR-K3



Gas release and surface temperatures in experiment R2-K13

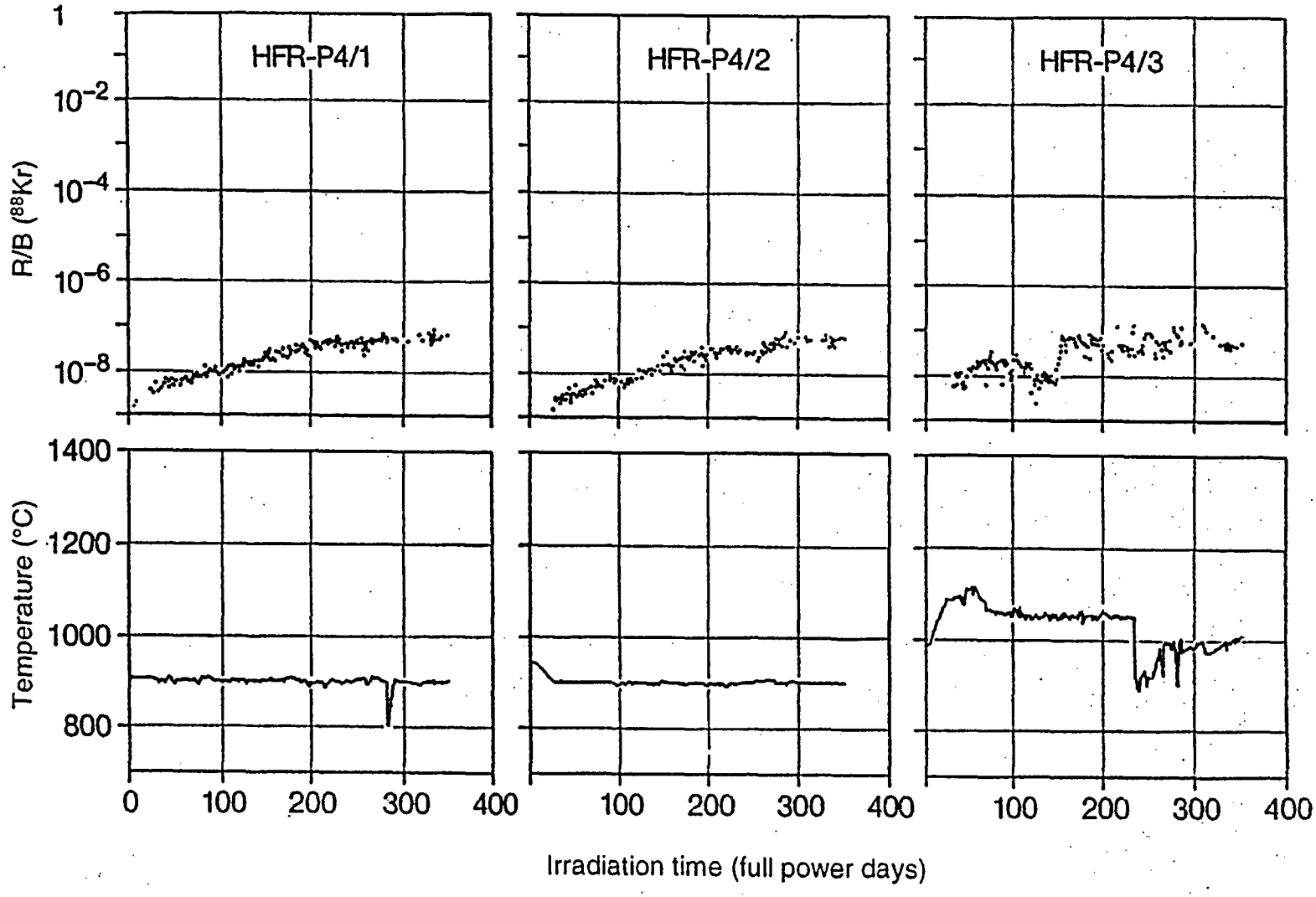
Radial distribution of irradiation temperatures (time average) of R2-K13 test and during 1600 °C heating test



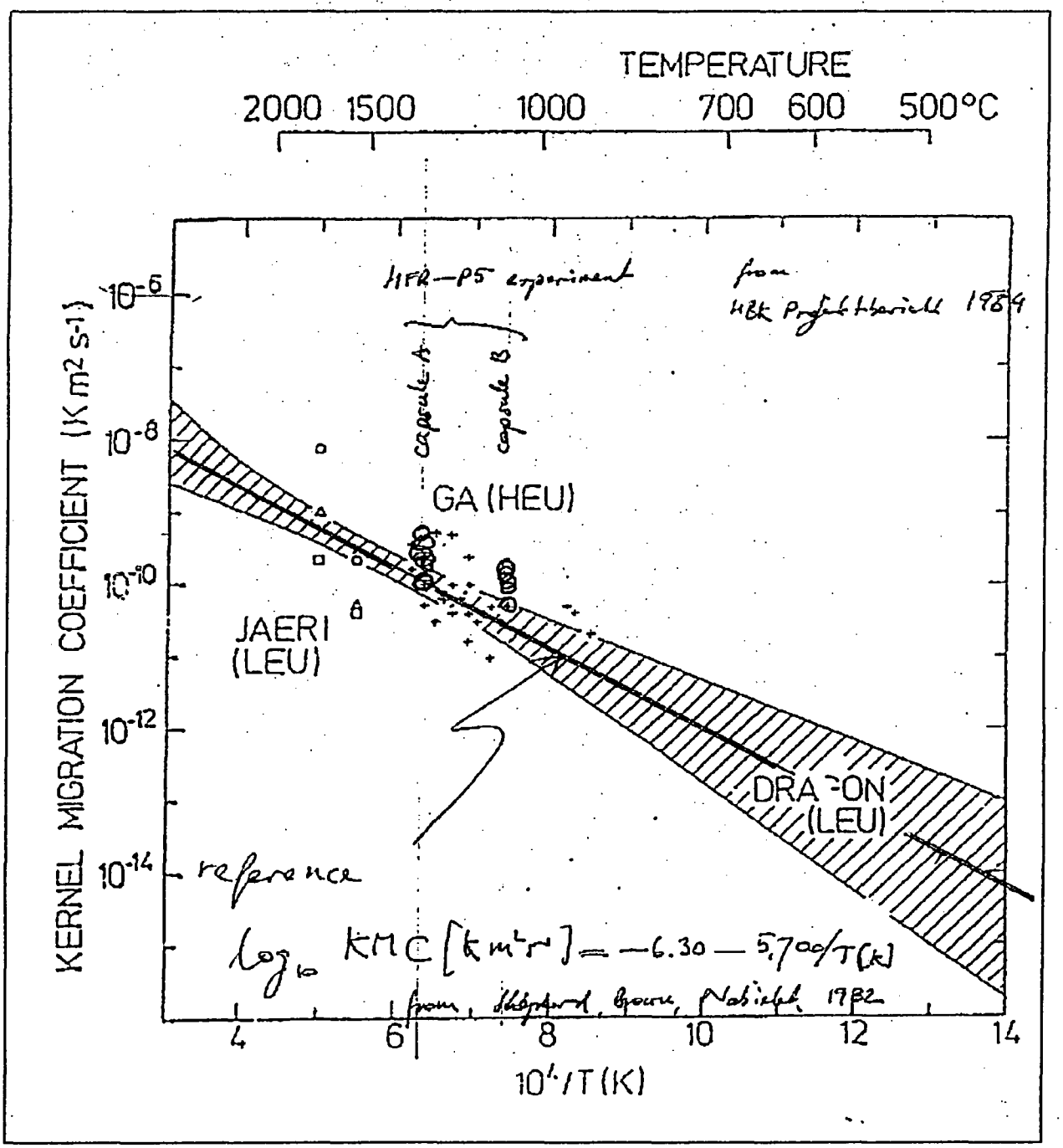


The release rate of short-lived fission gases, R/B, as a function of burnup in an on-going irradiation experiment in the Jülich DIDO reactor (FRJ2-K15, status from June 1989). The spikes in R/B have been measured during +200°C temperature transients. The slow increase in R/B is an artifact of the birthrate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

Irradiation to near 15 % FIMA



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



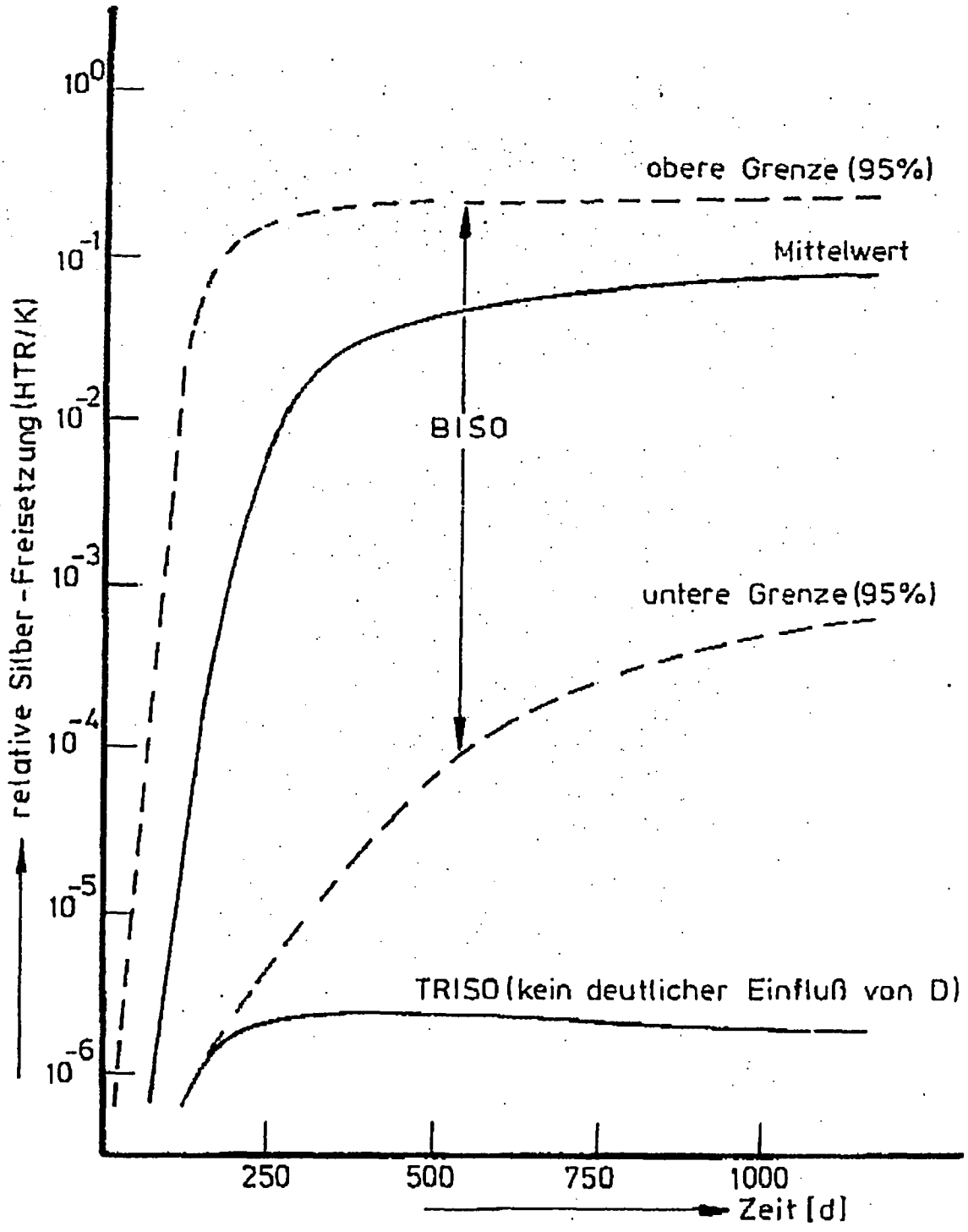


Abb. 6.10: Relative Silber-Freisetzung eines Partikels aus dem Zentrum eines Brennelementes beim Durchlauf durch die zentrale Core-Region (Region 1) eines HTR/K-Reaktors

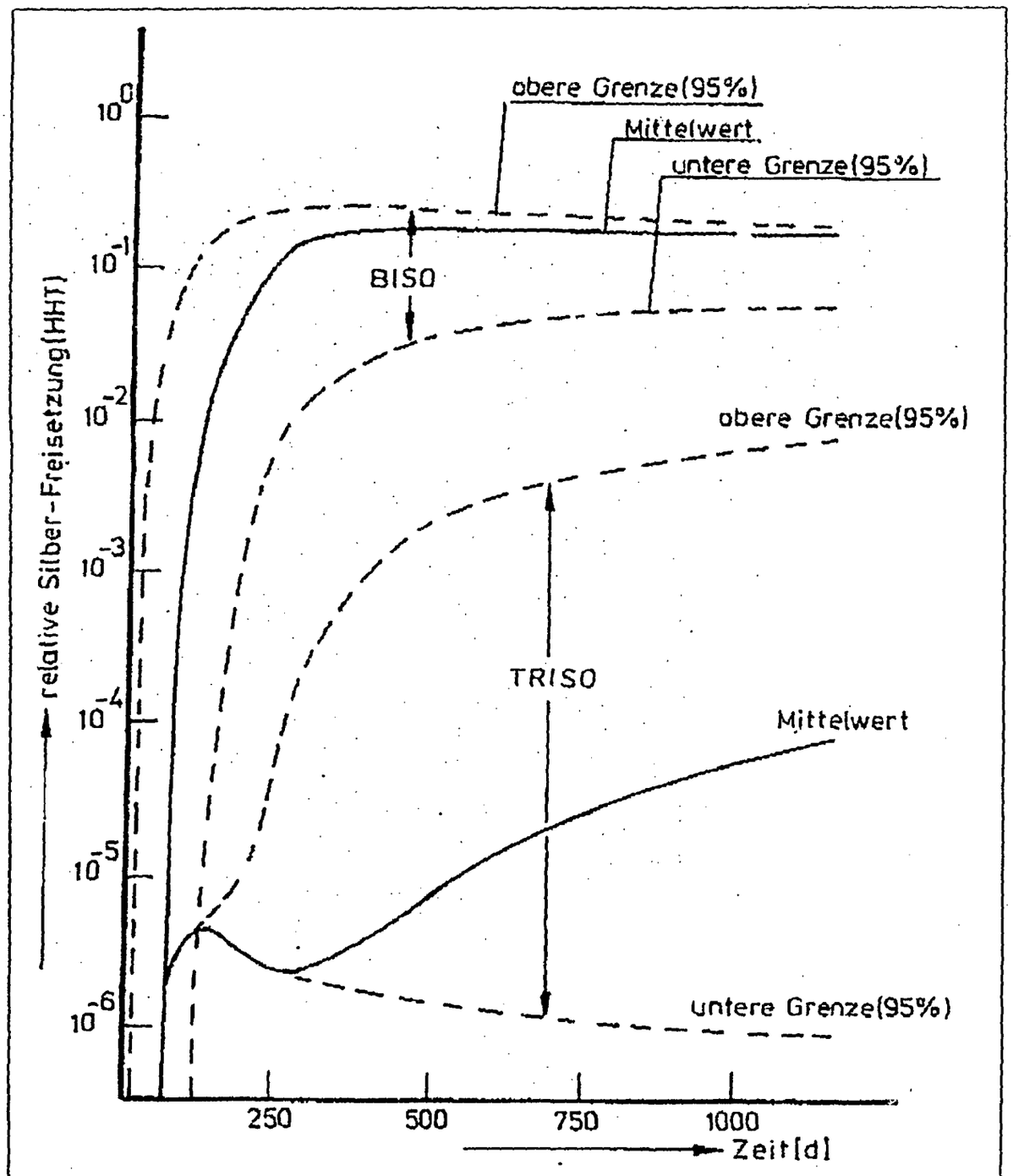


Abb. 6.11: Relative Silber-Freisetzung eines Partikels aus dem Zentrum eines Brennelementes beim Durchlauf durch die zentrale Core-Region (Region 1) eines HHT-Reaktors

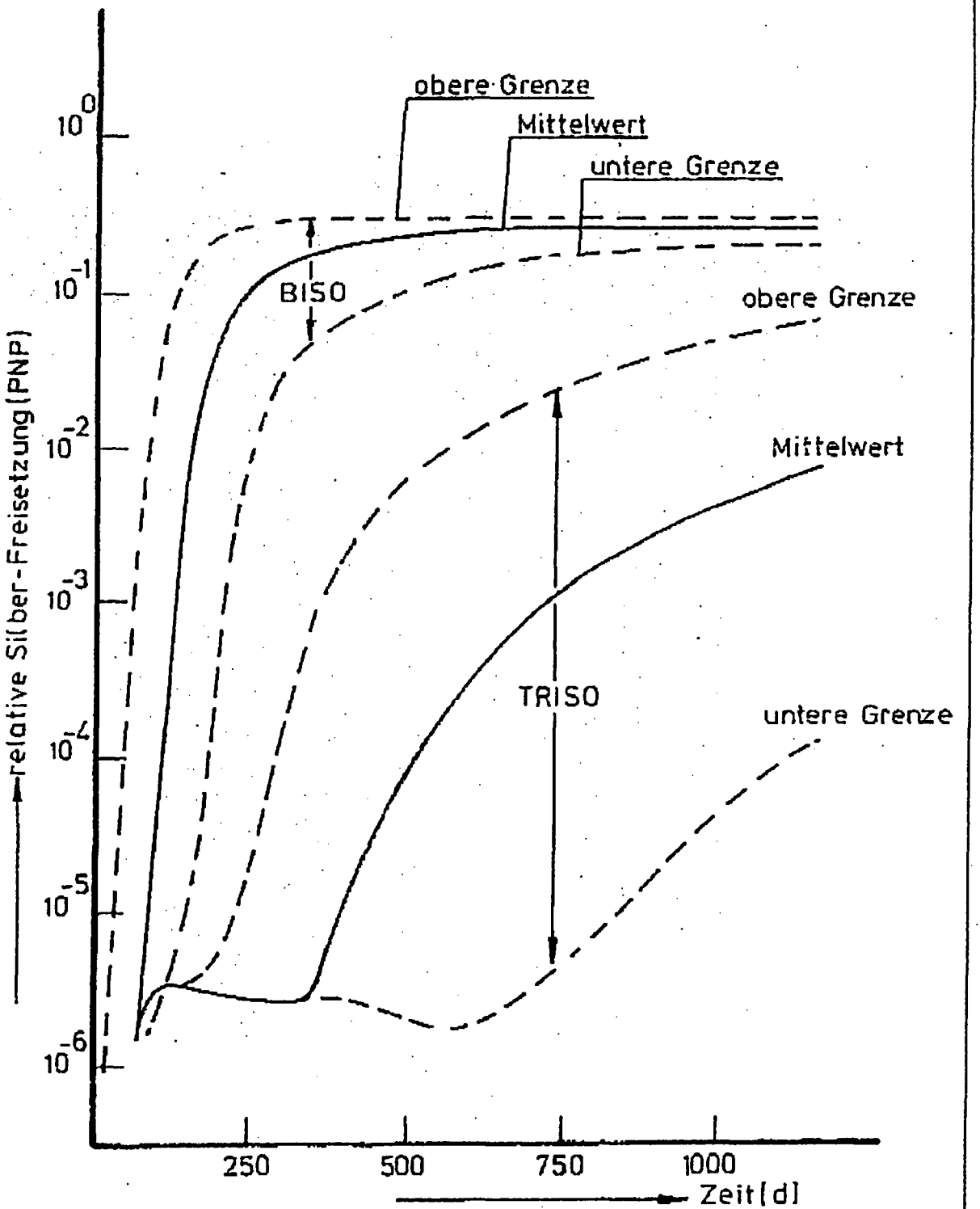
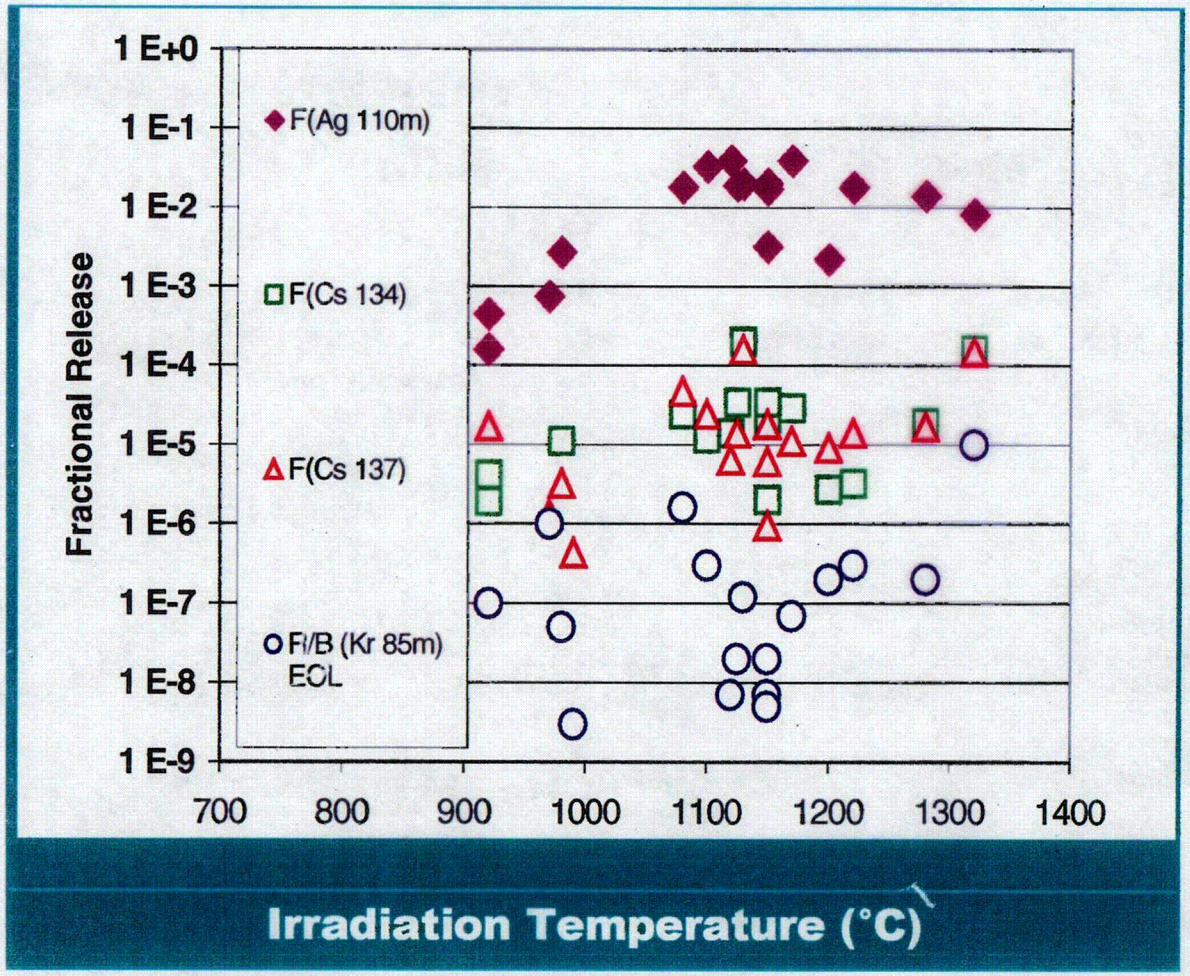


Abb. 6.12: Relative Silber-Freisetzung eines Partikels aus dem Zentrum eines Brennelementes beim Durchlauf durch die zentrale Core-Region (Region 1) eines PNP-Reaktors

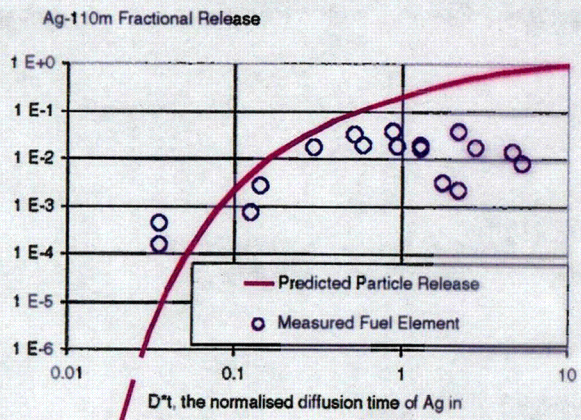
Diagram shows ^{110m}Ag and ^{137}Cs release fractions as a function of temperature in 230-530 d irradiation tests as measured in post irradiation examination work. Also shown is end-of-life R/B(^{85m}Kr).



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

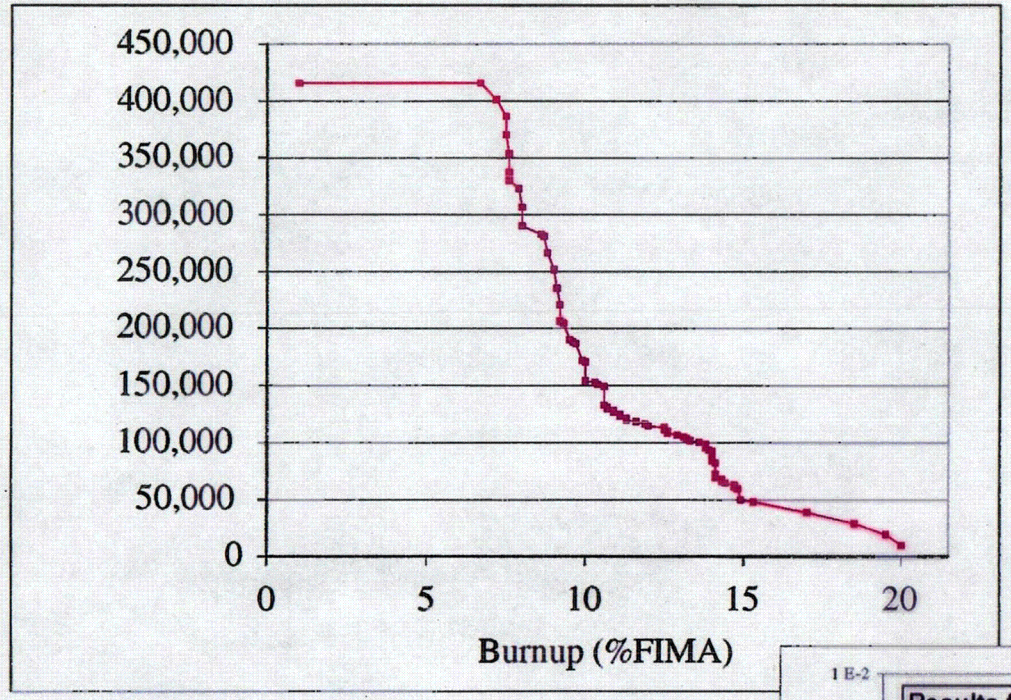
- Re-analyse existing data (i.e. temperatures and Ag releases)
- Perform dedicated irradiation tests
- Consider design/ material improve-ments
- If all else fails, redesign the HTR

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:

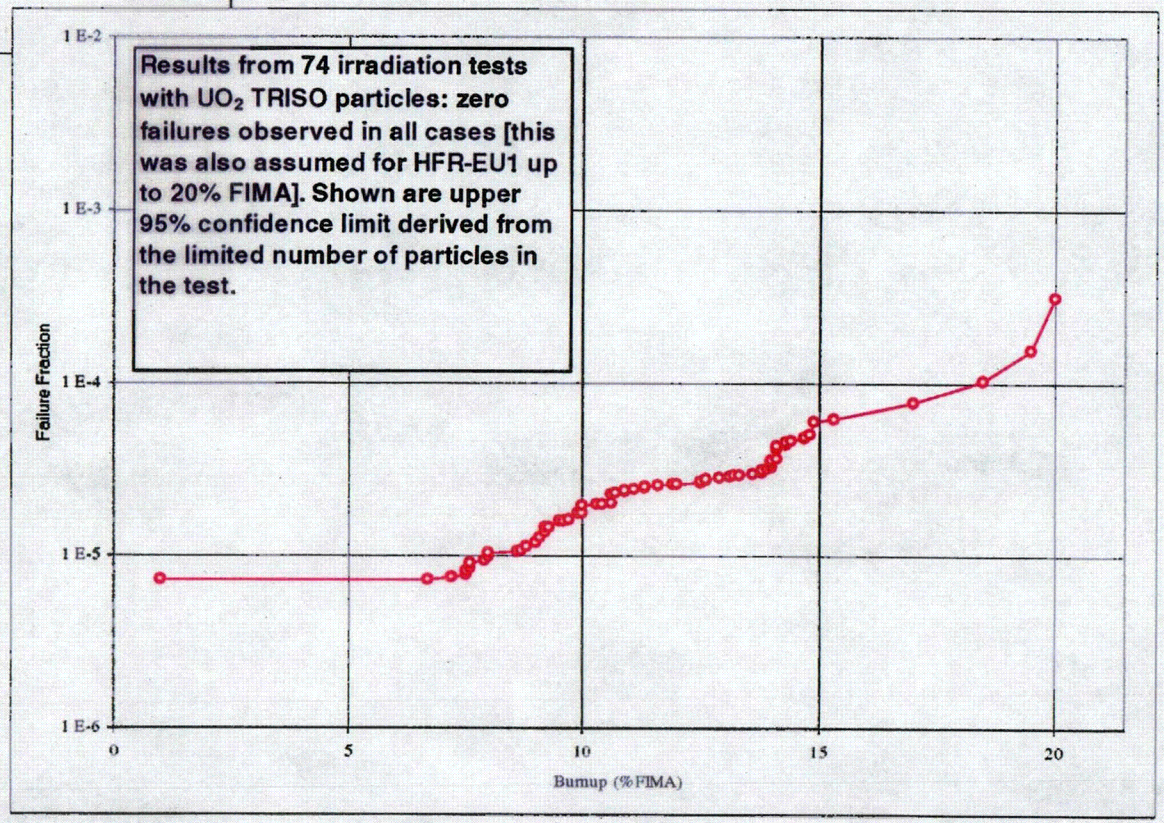


Of course, we need a much more sophisticated evaluation of these experiments taking account of distributions in space and time.

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:

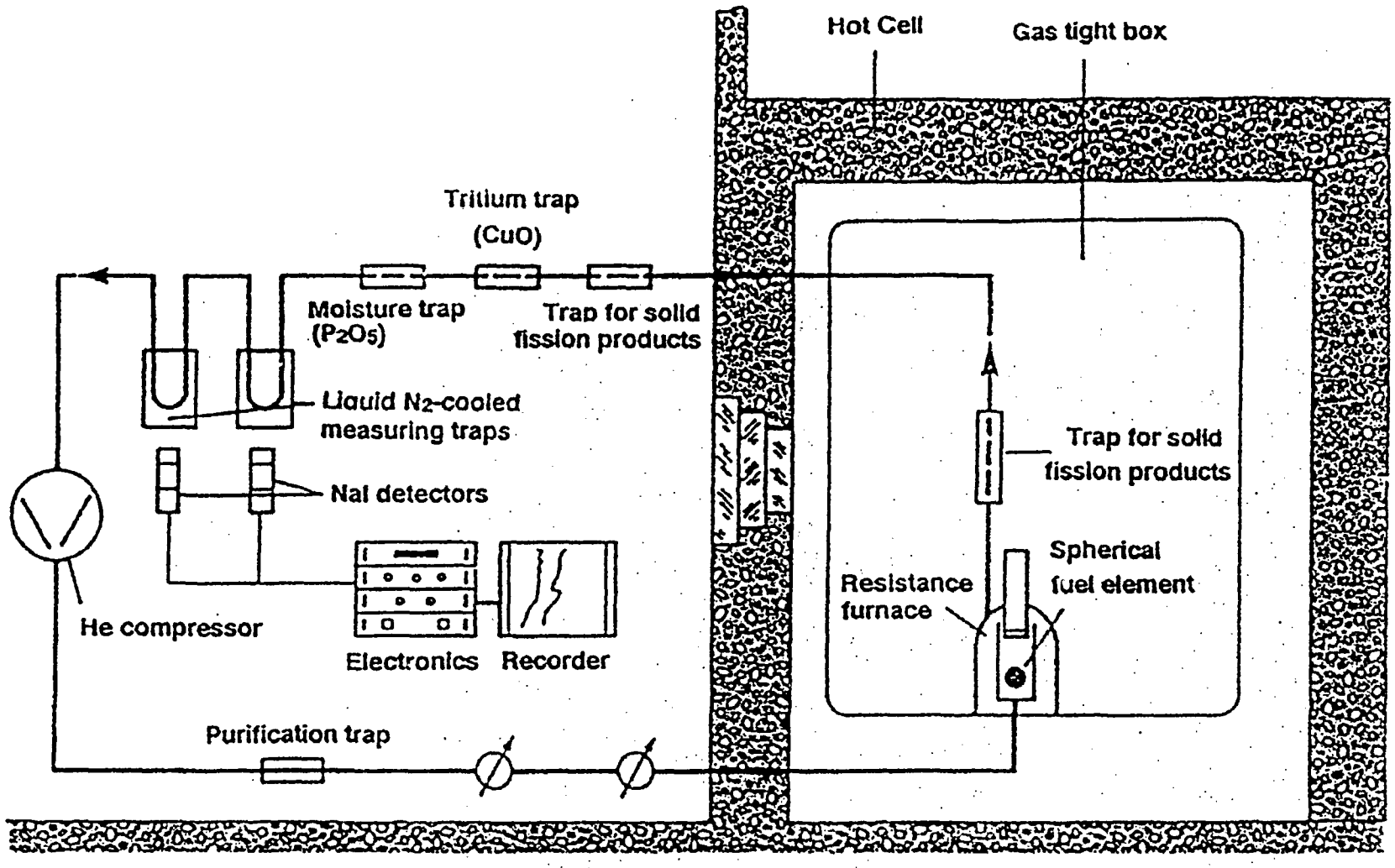


AVR reloads in the years 1966 to 1987

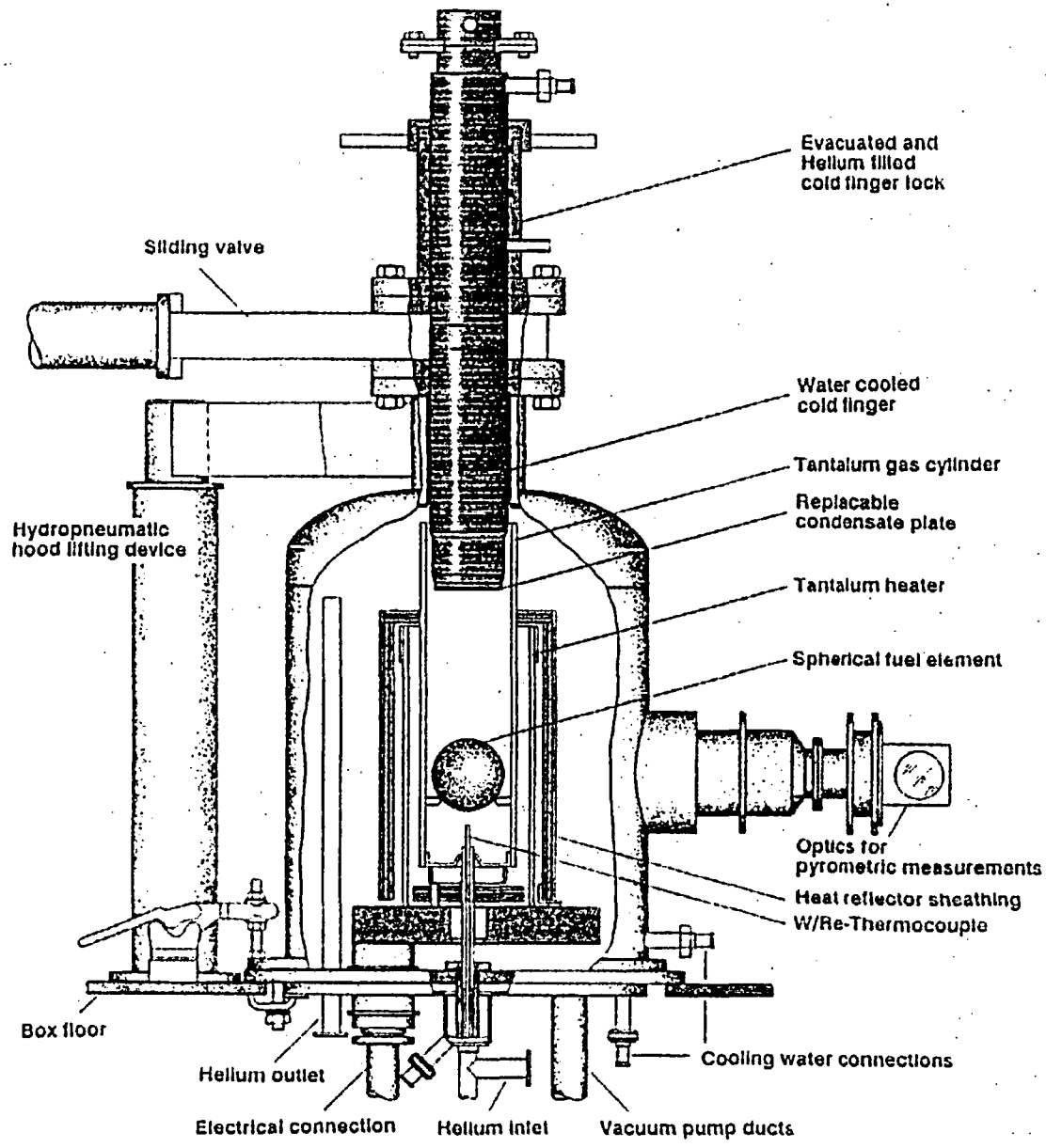
Reload number	Insertion date	Fuel element type	Number of fuel elements	Coated fuel particle Kernel	Coating	Enrichment
0	July 66	UCC	30155	$(Th,U)C_2$	HTI BISO	93%
1	Oct. 68	T	7510	$(Th,U)C_2$	HTI BISO	93%
3	Apr. 69	GK	17770	$(Th,U)C_2$	HTI BISO	93%
4	July 70	GK	6210	$(Th,U)C_2$	HTI BISO	93%
5-1	Nov. 70	GK	25970	$(Th,U)C_2$	HTI BISO	93%
5-2	Dec. 71	GO1	20625	$(Th,U)O_2$	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	LTI BISO	15%
				UO_2	LTI BISO	0.7%
8-1	May 74	GFB1	1440	UO_2	LTI BISO	93%
				ThO_2	LTI BISO	-
8-2	May 74	GFB2	1610	UO_2	LTI TRISO	93%
				ThO_2	LTI BISO	-
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	LTI TRISO	90%
				ThO_2	LTI BISO	-
13-3	Dec. 77	GFB5	5354	UCO	LTI TRISO	92%
				ThO_2	LTI TRISO	-
13-2	July 80	GFB4	5861	UC_2	LTI TRISO	90%
				ThO_2	LTI BISO	-
15	Feb. 81	GO2	6067	$(Th,U)O_2$	LTI TRISO	93%
18	July 81	GO3	11547	$(Th,U)O_2$	HTI BISO	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			

Heatup Testing

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

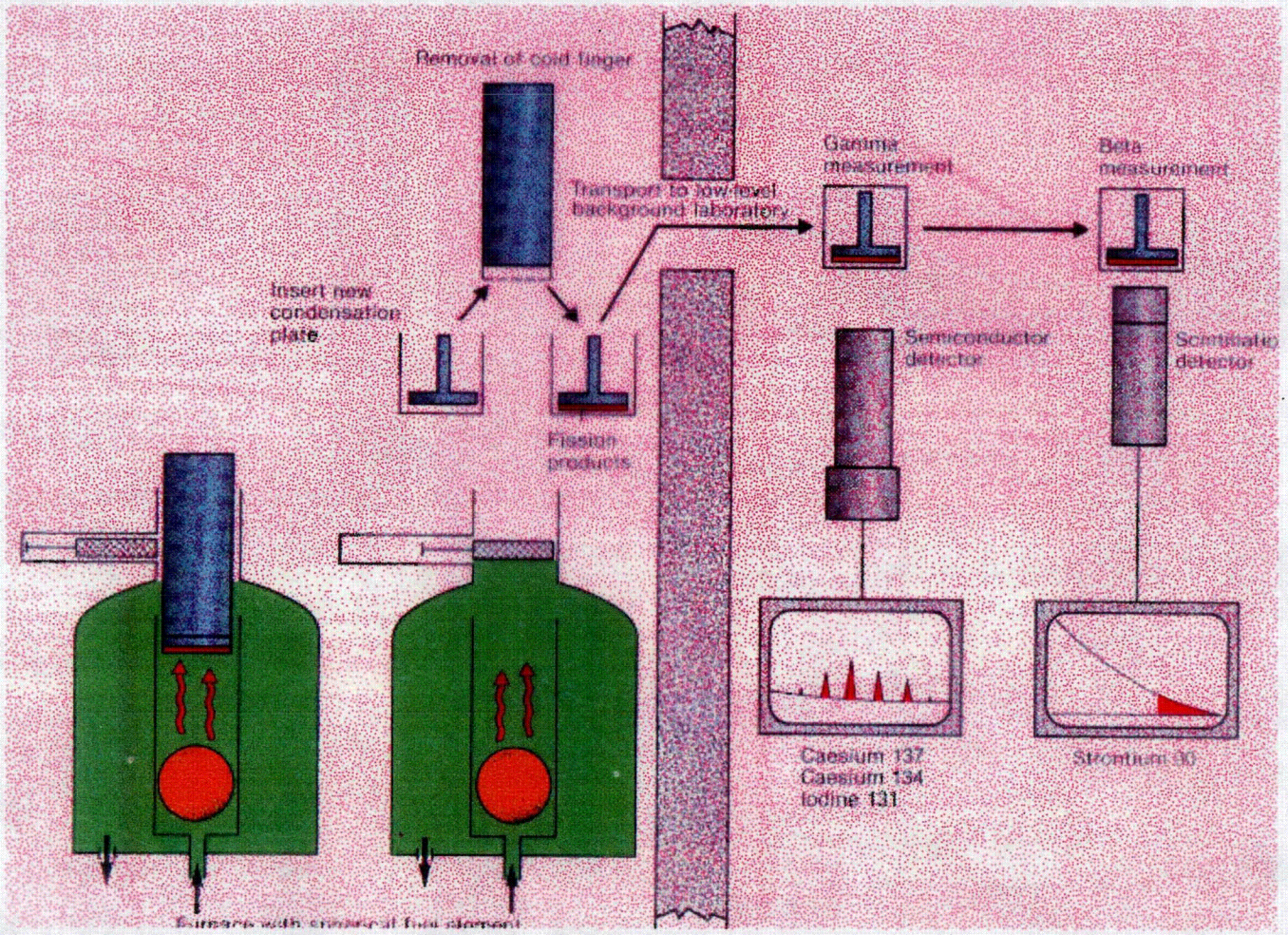


Sweep gas circuit of cold finger apparatus (KÜFA).



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

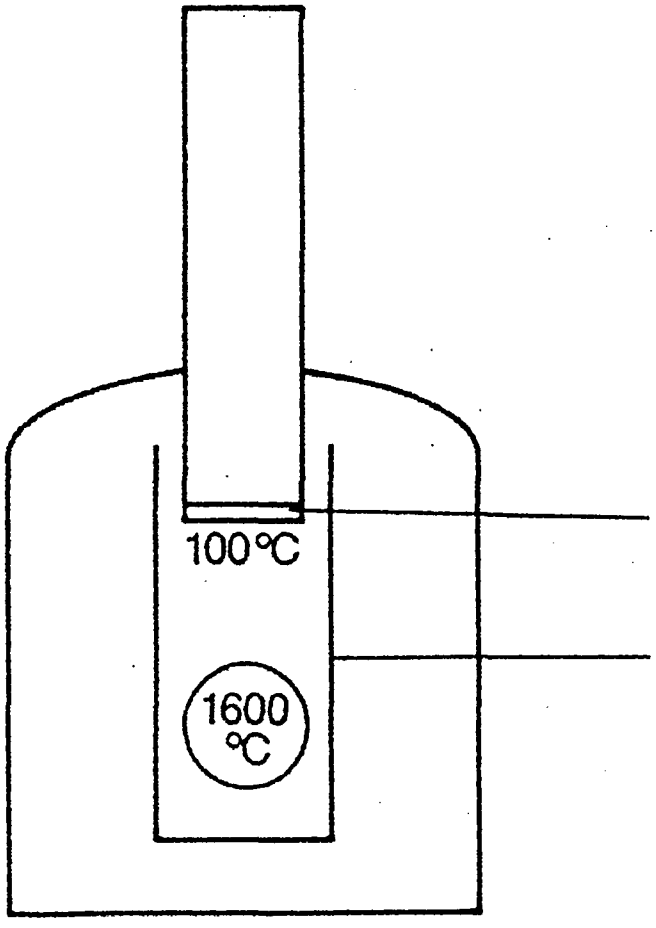
Schema of KÜFA solid fission product measurements



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

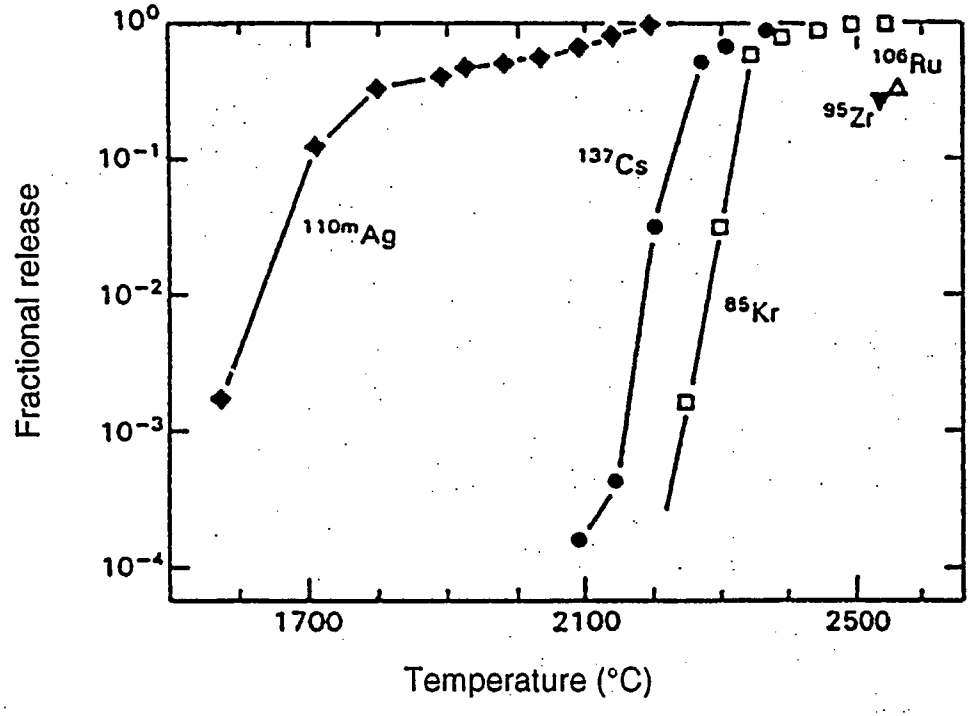
Important fission products		
ELEMENT	ISOTOPE	HALF LIFE
Solid fission products		
Cesium	^{137}Cs	30 years
	^{134}Cs	2 years
Strontium	^{90}Sr	29 years
Iodine	^{131}I	8 days
Fission gases		
Krypton	^{85}Kr	11 years
Xenon	^{133}Xe	5 days

Plate-out fractions of KÜFA furnace cold finger plate

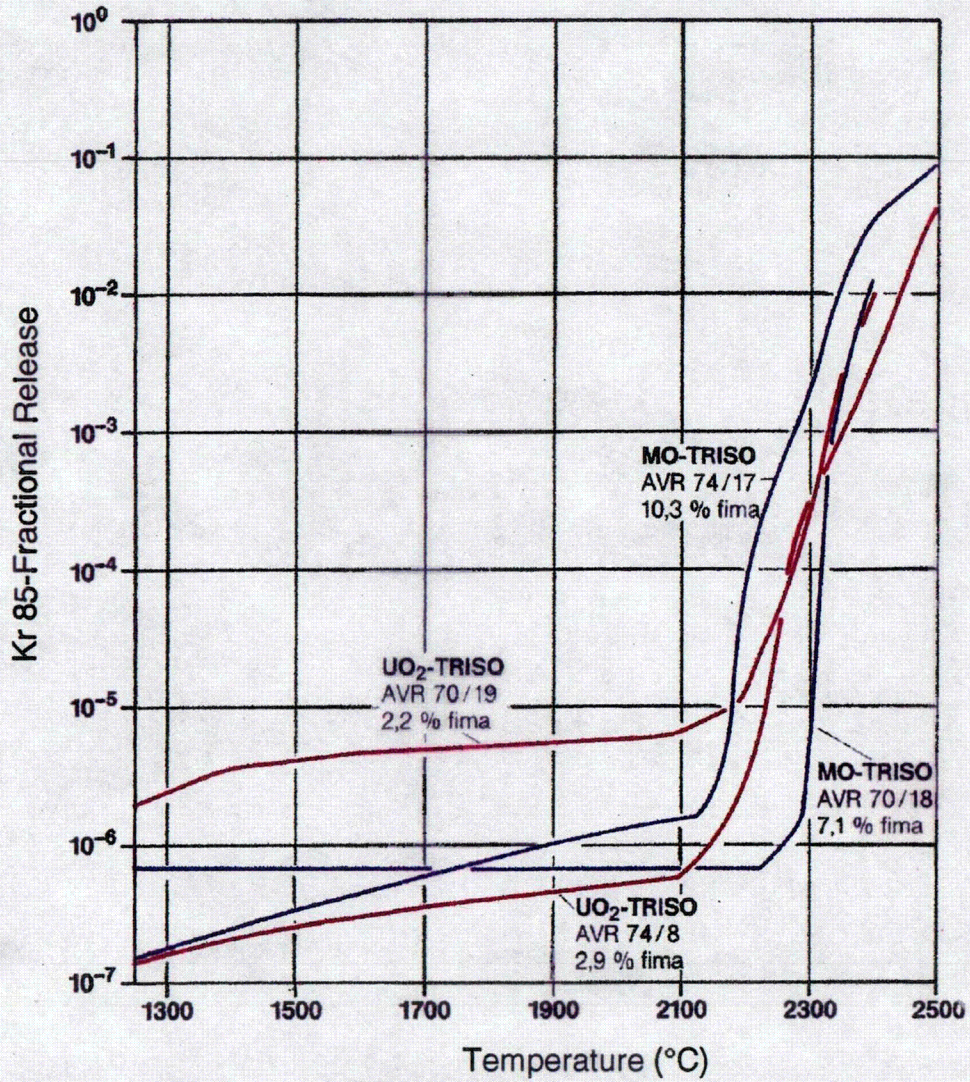


	Released Fraction	
	Cs, I, Ag	Sr
plate	0.7	0.2
Ta-tube	0.02	0.8
Furnace	0.3	-

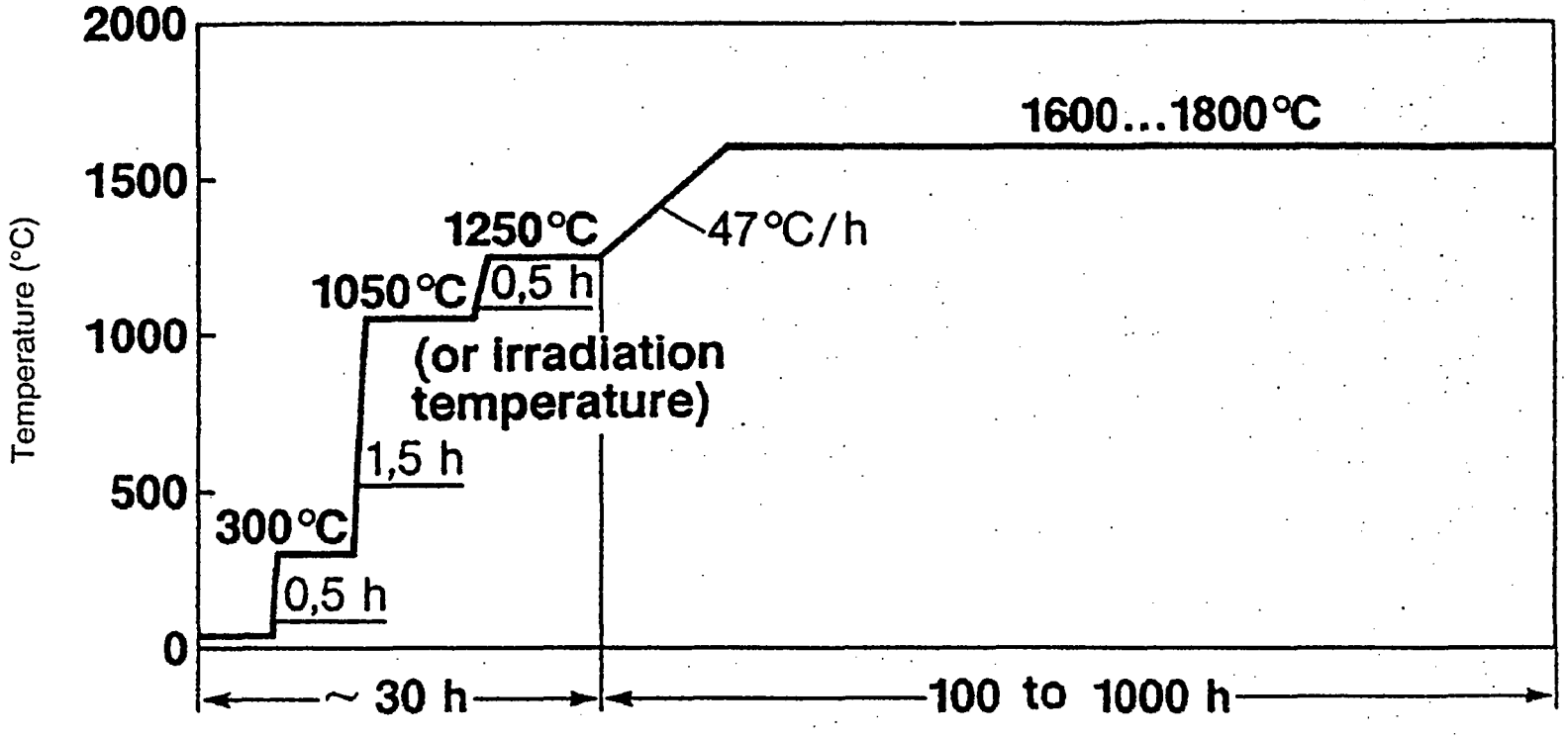
Sequence of fission product release is
^{110m}Ag, ^{137}Cs, ^{134}Cs, ^{85}Kr, ^{90}Sr, ^{106}Ru, ^{95}Zr



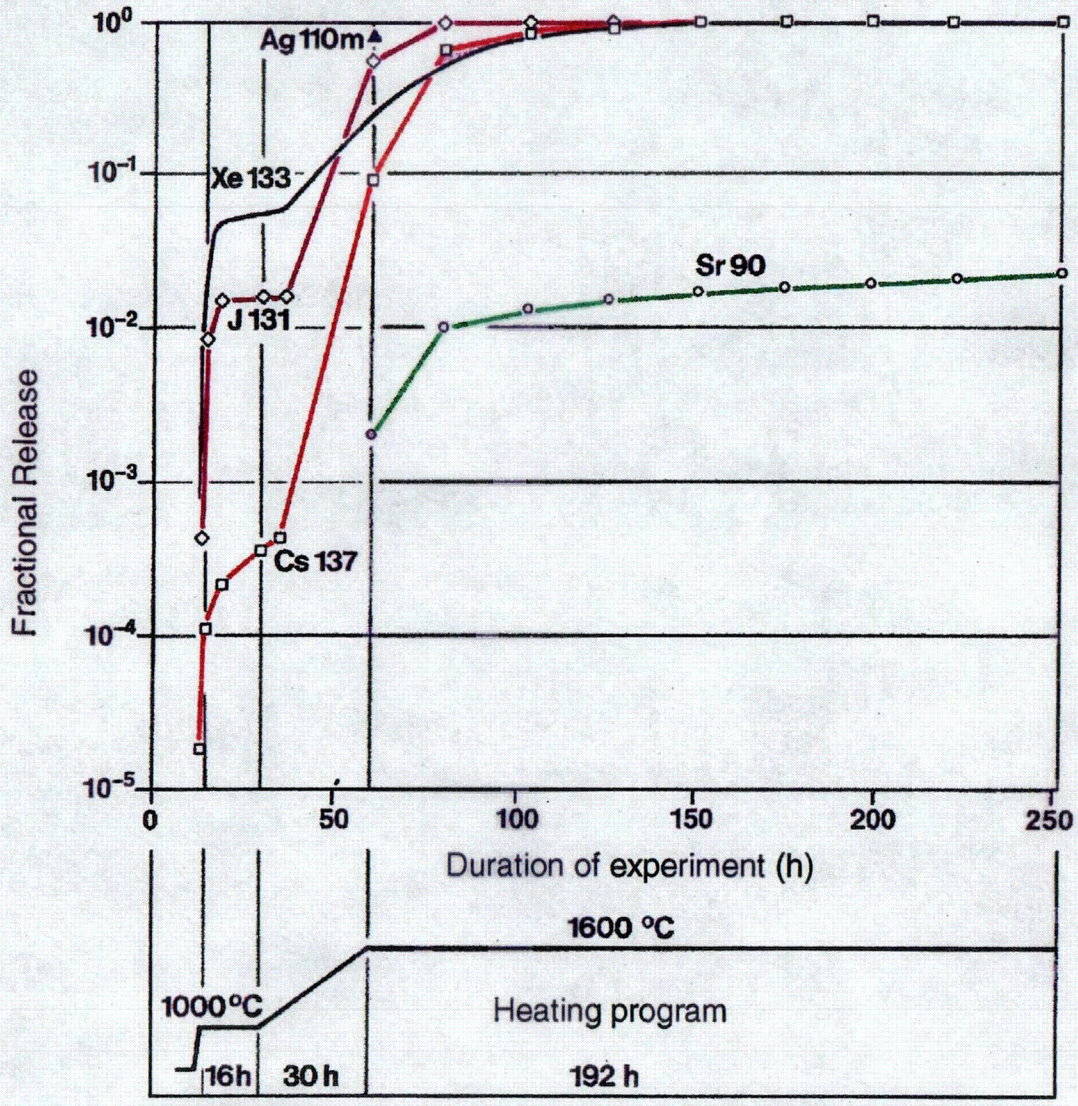
Here the fractional release during temperature ramp to 2600°C [GA data, from Fig 4-39, page 192 of IAEA-TECDOC-978]



Gas release during temperature ramp to 2500°C.

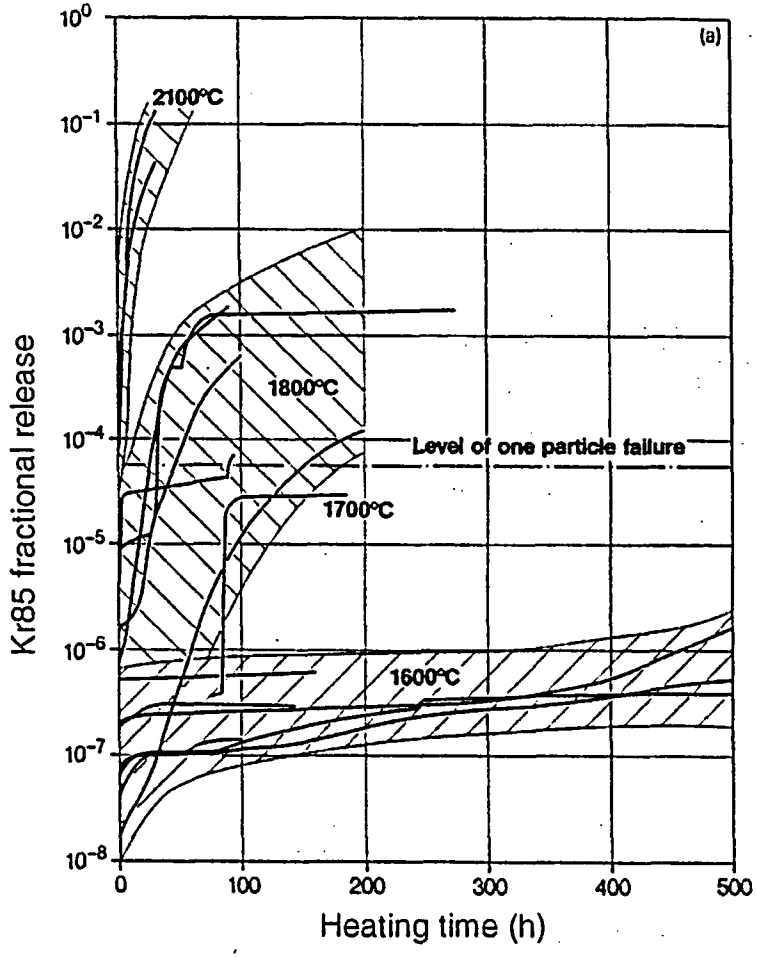


Temperature / time correlation in standard isothermal tests.

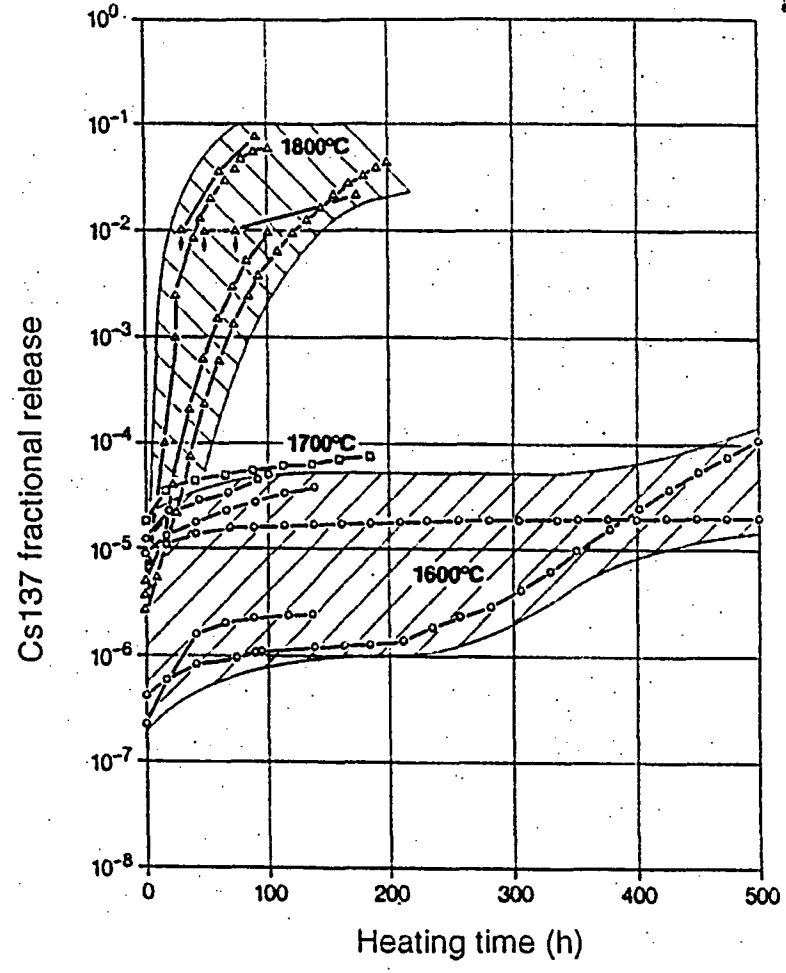


Fission product release from UO_2 kernels (FRJ2-P28/C6) during heating test at 1600 °C.

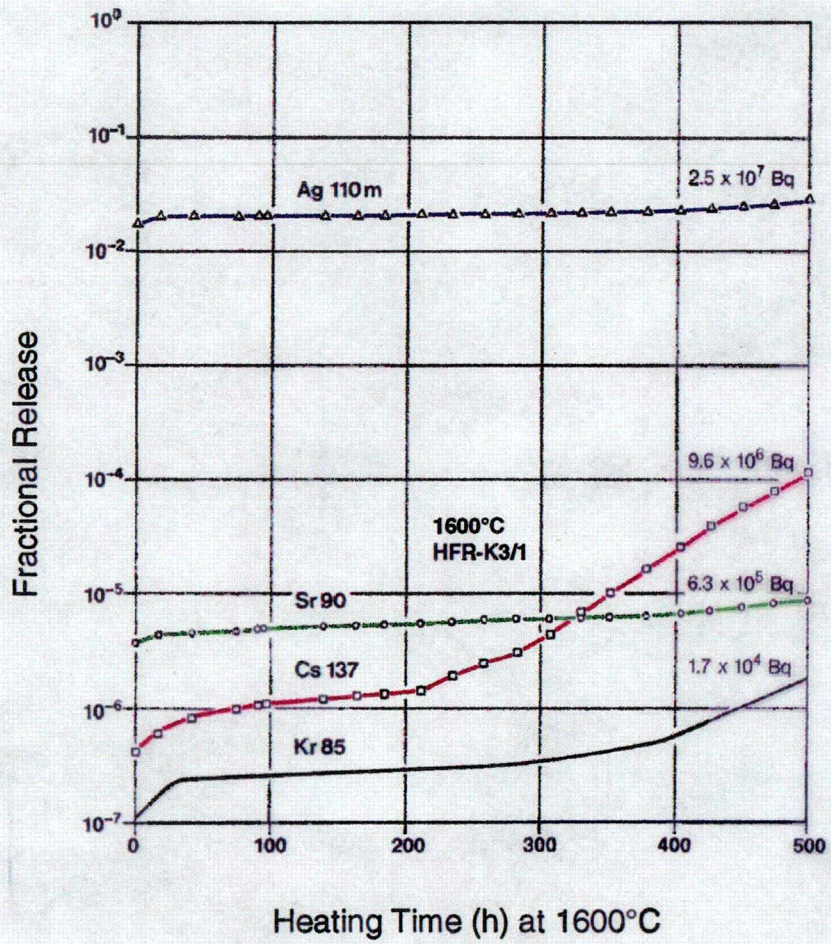
Heating tests at 1600-2100 °C



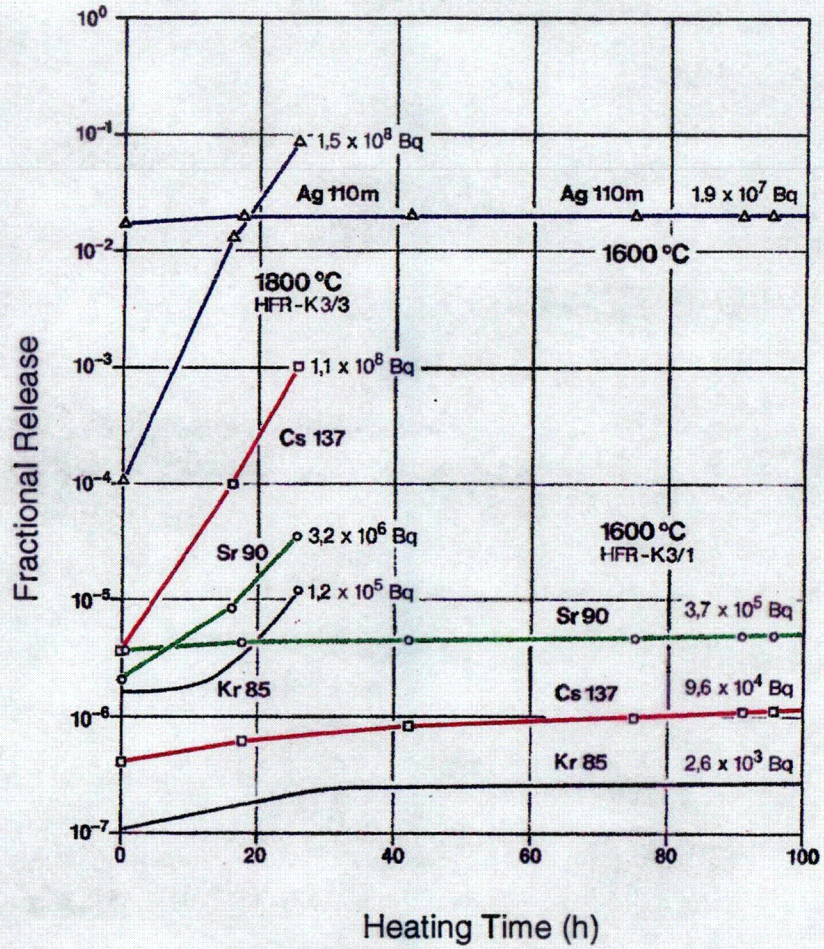
Krypton release during tests with irradiated spherical fuel elements at 1600 to 2100 °C.



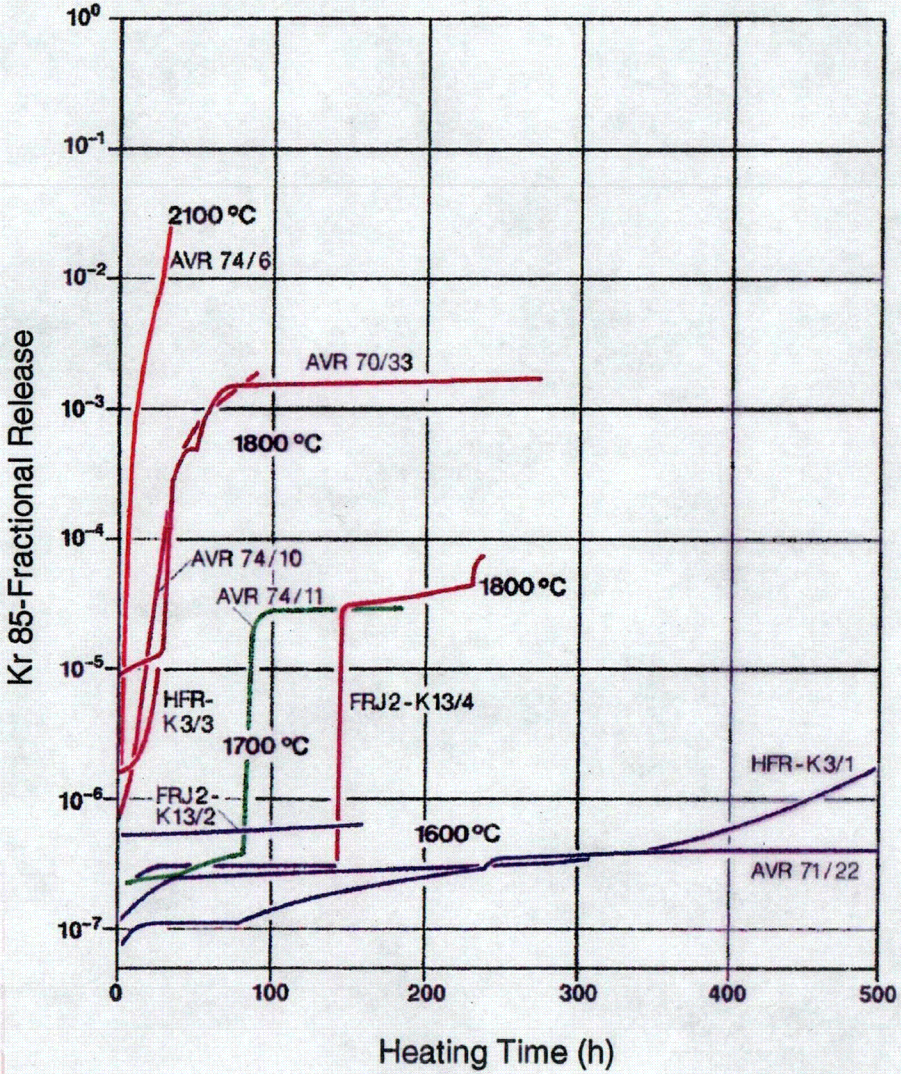
Caesium release from heated spheres as a function of heating times up to 500 hours.



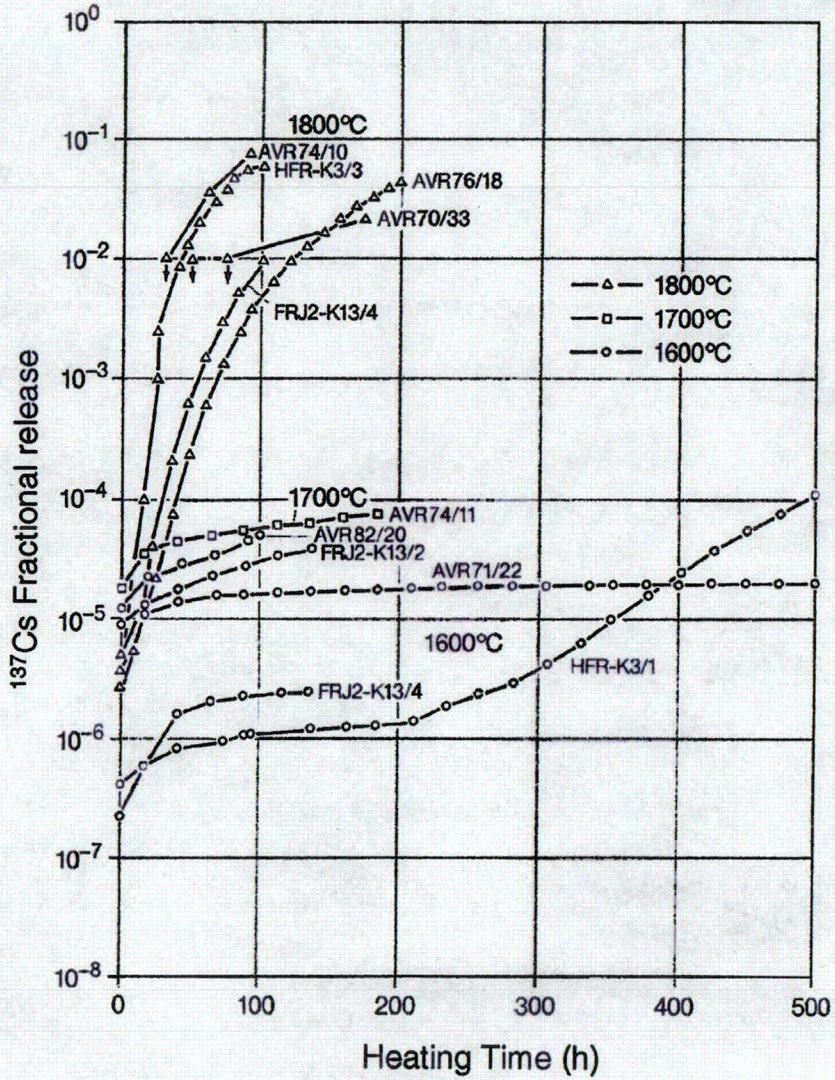
Fission product release from fuel element HFR-K3/1.



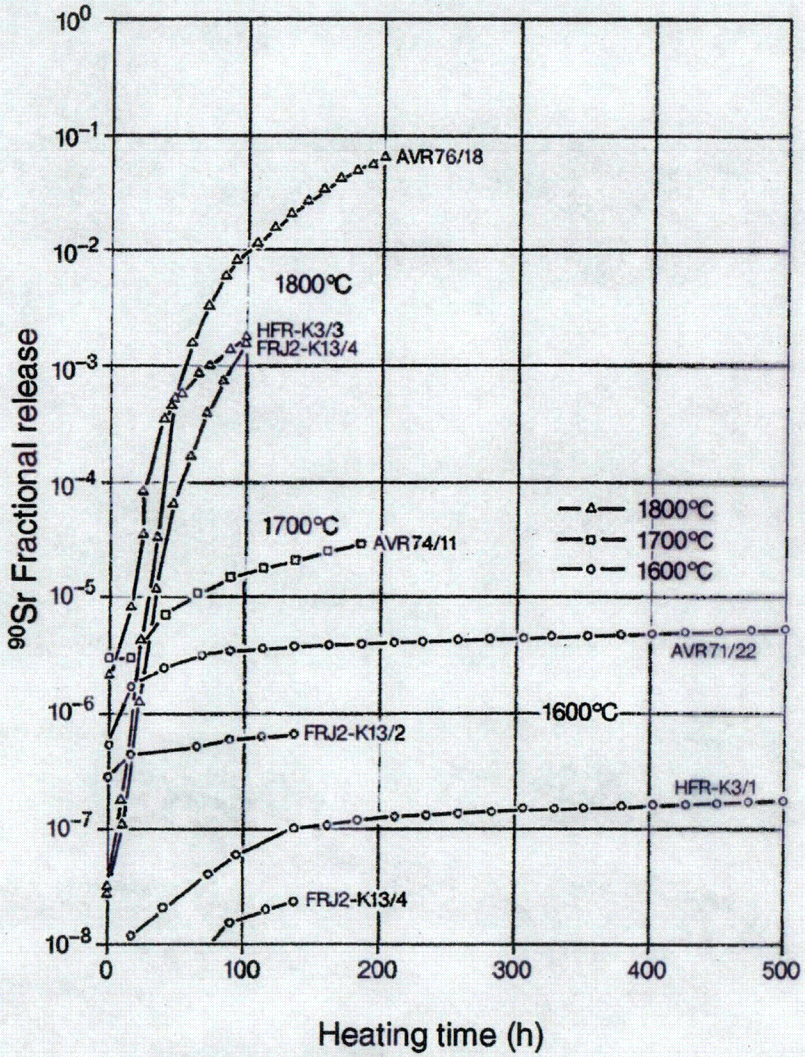
Fission product release at 1600°C and 1800°C.



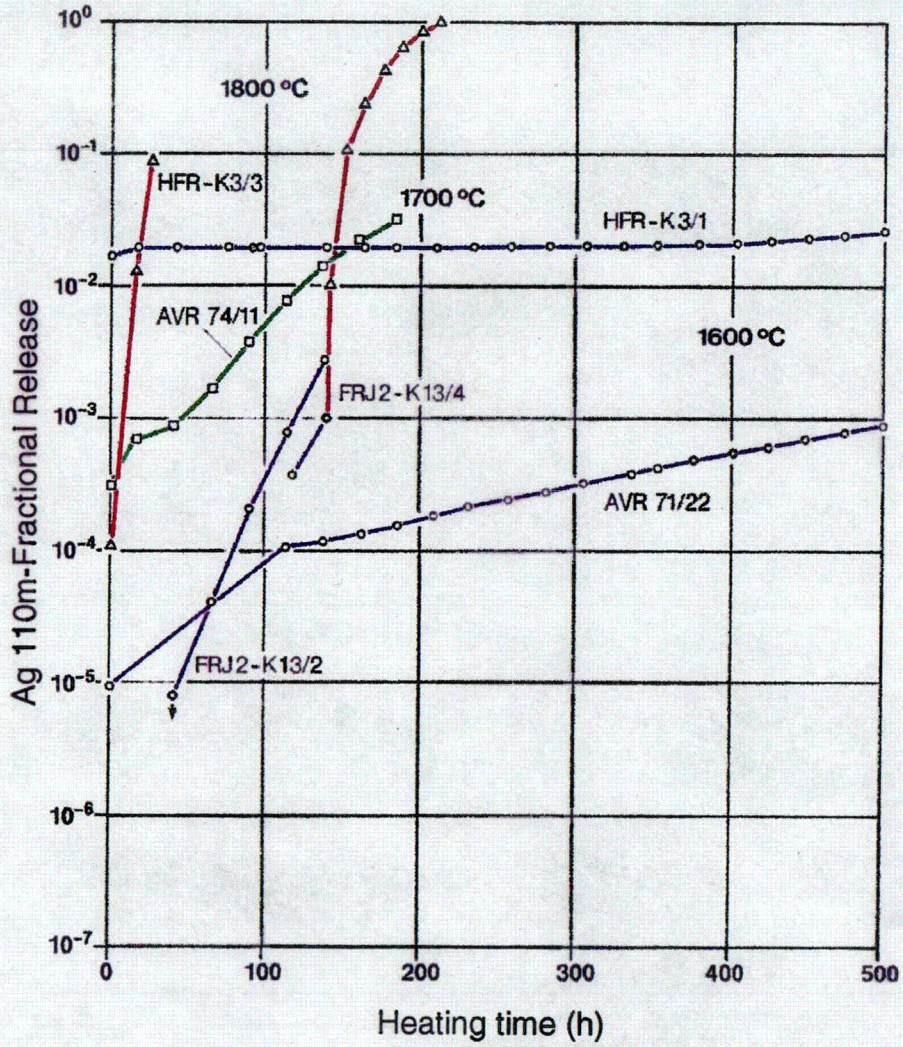
Krypton release in isothermal heating tests.



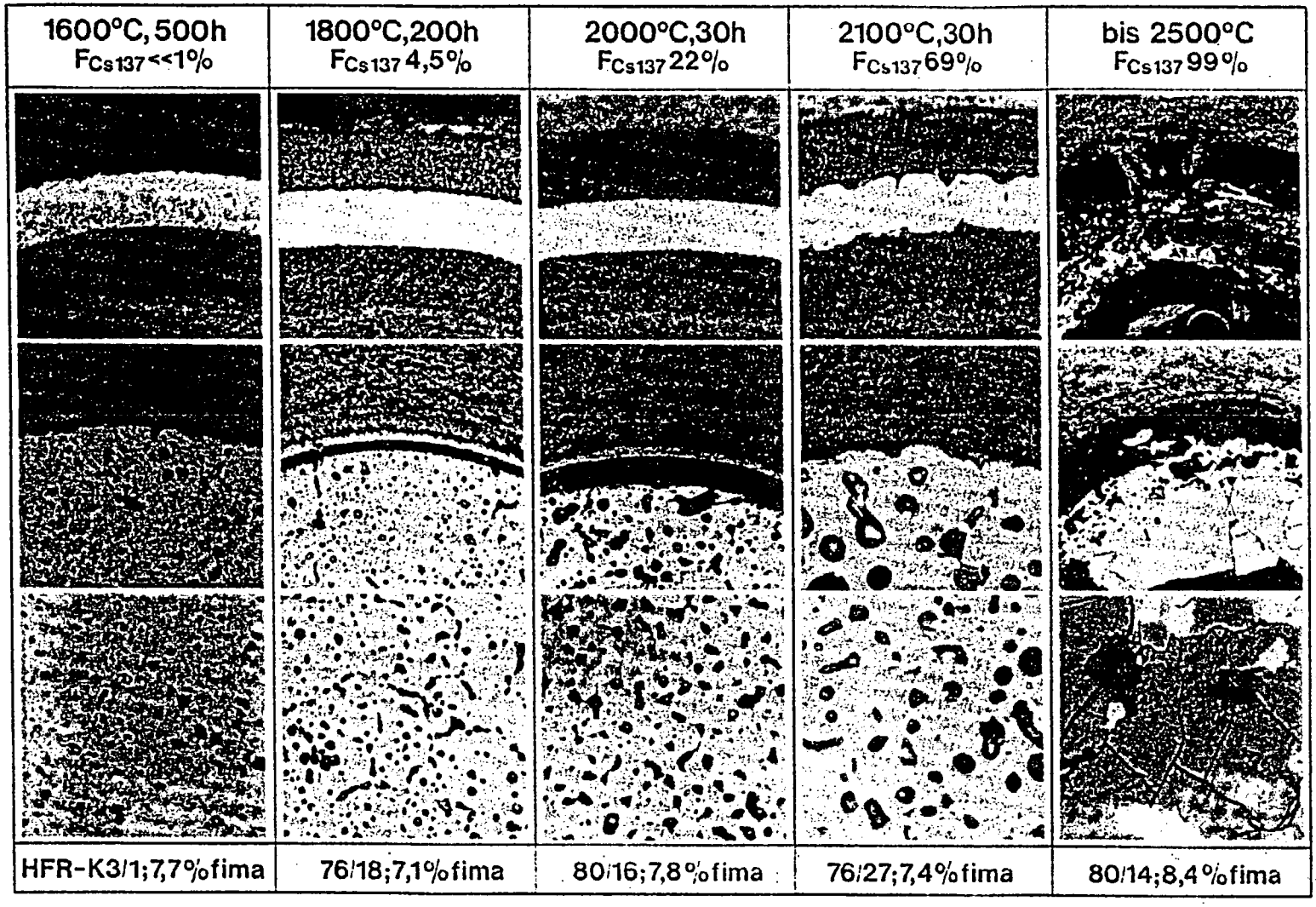
Cesium release in isothermal heating tests.



Strontium release in isothermal heating tests.

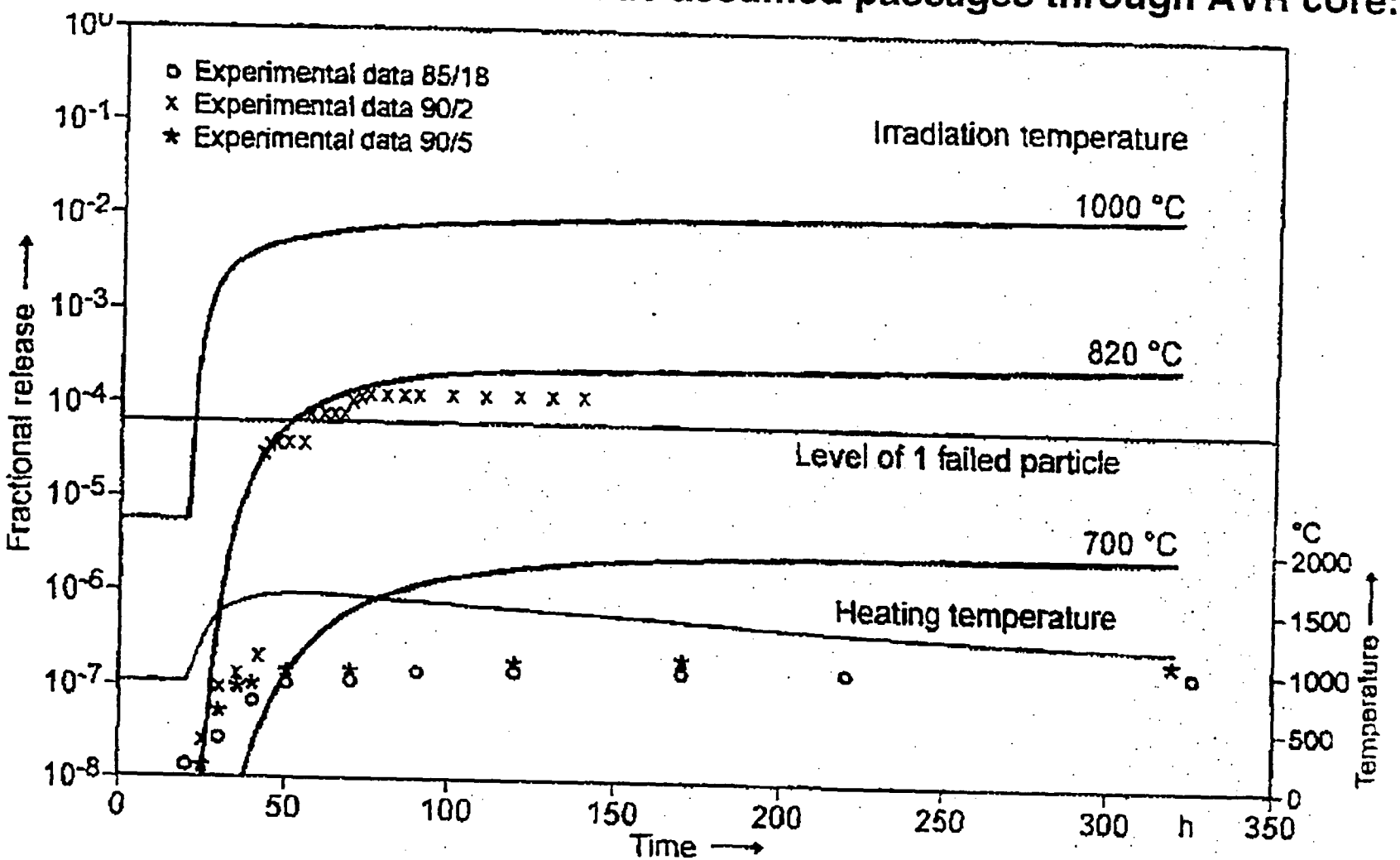


Ag 110m release from fuel elements with UO_2 TRISO particles.

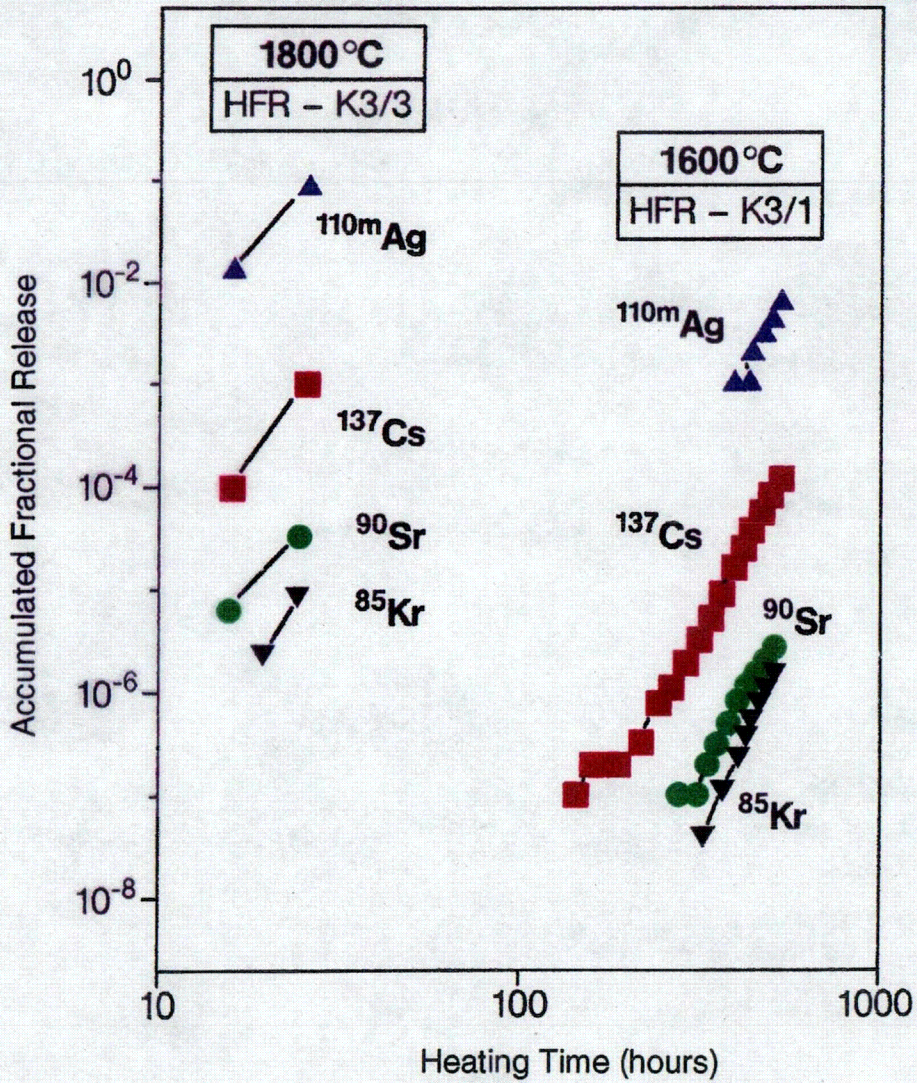


Ceramographic sections through UO_2 TRISO particles.

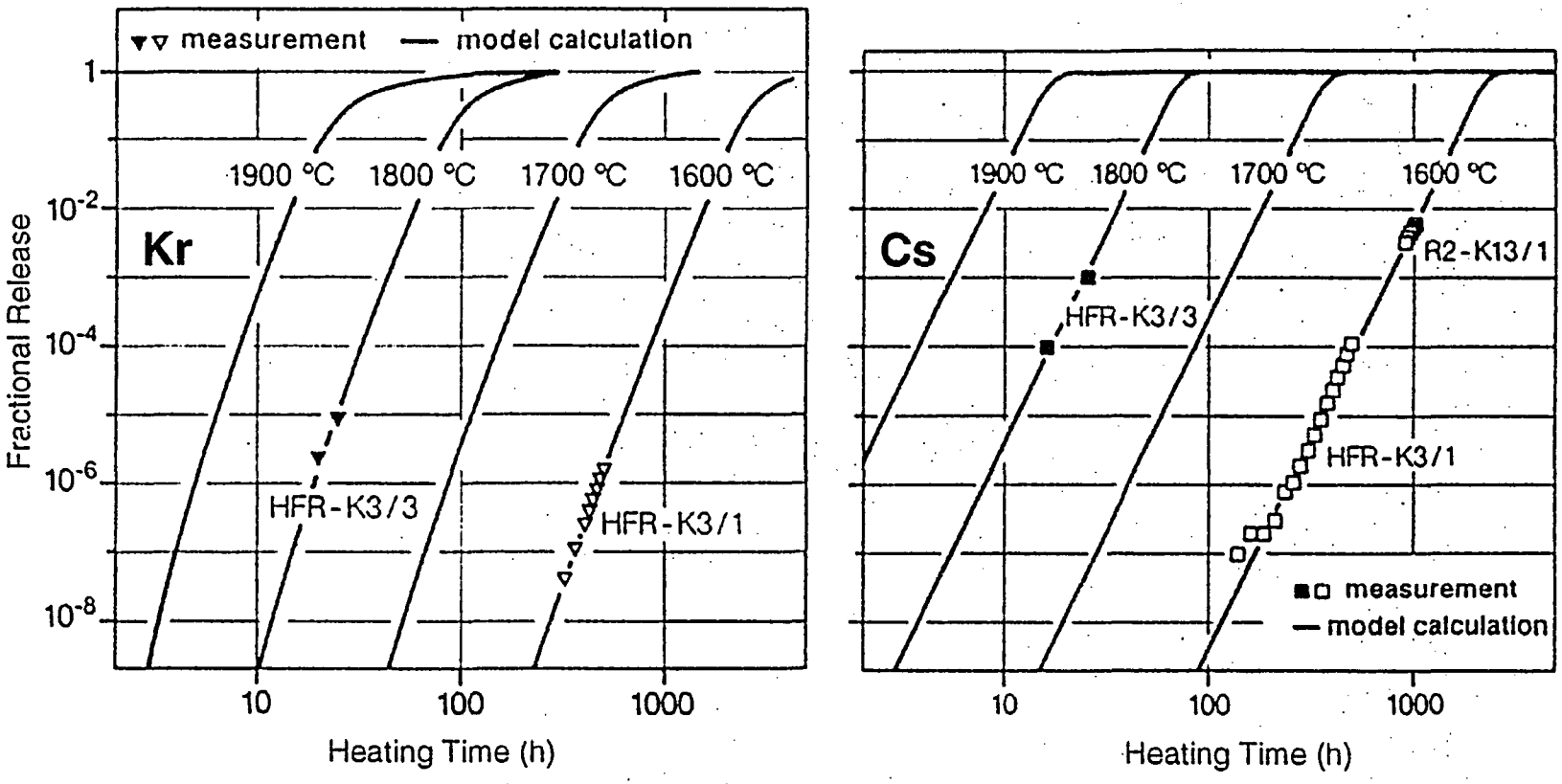
Measured krypton releases from AVR fuel elements 90/2, 90/5 and 85/18 during accident scenario heatup to 1620°C and three PANAMA predictions using various assumed passages through AVR core:



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



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