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Project No. 713

May 24, 2002

U.S. Nuclear Regulatory Commission  
Attn: Document Control Desk  
Washington, DC 20555

**Subject:** Submittal of the Pebble Bed Modular Reactor Pty. Document Number 010520-425, Revision 2, "Pebble Bed Modular Reactor Nuclear Fuel."

**Reference:** Letter from USNRC to Exelon Generation Company dated February 6, 2002, "Request for Withholding Information from Public Disclosure, Exelon Generation Letter of November 16, 2001."

Letter from Exelon Generation Company to USNRC dated March 5, 2002 "Withdraw and Re-submittal of the Pebble Bed Modular Reactor Core Design and Heat Removal Presentation Material and the Design and Heat Removal Preliminary Description Document."

By referenced letter dated March 5, 2002 from Exelon Generation Company (EGC) and in response to NRC letter dated February 6, 2002, EGC requested the withdrawal of a Pebble Bed Modular Reactor (PBMR) Pty., Document Number 010520-425, Revision 1, "PBMR Nuclear Fuel." In our March 5, 2002 letter we also committed to provide updated proprietary and redacted versions of the withdrawn "PBMR Nuclear Fuel," document along with corresponding versions of the presentation material submitted to the NRC via a letter dated June 12, 2001. This letter is in response to those commitments.

Portions of the information contained in Revision 2 of the attached document are identified as proprietary, and requested to be withheld from public disclosure on the grounds that these portions contain information in the nature of trade secrets and commercial or financial information that is confidential. We consider, however, that this information will be beneficial to the NRC's understanding of the PBMR fuel design, and this type of information is consistent with the NRC's Policy on Advanced Reactors, NUREG 1226 "Development and Utilization of the NRC Policy Statement on the Regulation of Advanced Nuclear Power Plants."

D064

US NRC  
May 24, 2002

Since EGC has announced that will be concluding the licensing pre-application activities earlier than originally anticipated, we will not provide an updated corresponding version of the presentation material as committed in our March 5, 2002 letter. However, the information presented on June 12, 2001 in the form of presentation slides is contained as text in the "PBMR Nuclear Fuel," Revision 2, document, which will satisfy the intent of this commitment.

We have enclosed an application and affidavit for withholding information from public disclosure pursuant to 10 CFR 2.790 (a) (4) regarding this information.

We have attached proprietary and non-proprietary (i.e., redacted) versions of the "PBMR Nuclear Fuel," Revision 2 (Attachment 1 and Attachment 2 respectively), and have indicated revision changes by providing revision bars on the right hand margins of each effected page for your convince.

If you have any questions concerning this matter, please do not hesitate to contact us.

Sincerely,



Marilyn C. Kray  
Vice President, Special Projects

Enclosures: Affidavit  
Attachments

cc: Farouk Eltawila, Office of Nuclear Reactor Research  
James Lyons, Office of Nuclear Reactor Regulation  
John Flack, Office of Nuclear Reactor Research  
Amy Cabbage, Office of Nuclear Reactor Regulation  
Stuart Rubin, Office of Nuclear Reactor Research

Affidavit of Marilyn C. Kray

Commonwealth of Pennsylvania:

: ss.

County of Chester

:

Marilyn C. Kray being duly sworn, deposes and states as follows:

1. I am Vice President, Special Projects, Exelon Generation Company, L.L.C. (Exelon), and I am authorized to execute this affidavit in support of a request to withhold certain information, described in paragraph (2) below, from public disclosure and in accordance with Section 2.790(a)(4) of the Commission's regulations.
2. The information sought to be withheld is contained in the letter, M. C. Kray (Exelon Generation Company, L.L.C.) to the U. S. Nuclear Regulatory Commission Document Control Desk, Project No. 713.
3. The information which is sought to be withheld from public disclosure is proprietary information of Pebble Bed Modular Reactor (Pty) Limited, a Republic of South Africa corporation ("PBMR Co"), and has been provided to Exelon subject to an agreement that it will be treated as confidential and proprietary information and not be disclosed publicly. Exelon has contributed substantial funds for the development of the information and holds a beneficial ownership interest in PBMR Co.
4. In making this application for withholding of proprietary information, Exelon relies upon the exemption from disclosure set forth in the Freedom of Information Act ("FOIA"), 5 USC Sec. 552(b)(4), and the Trade Secrets Act, 18 USC Sec. 1905, and NRC regulations 10 CFR Section 9.17(a)(4) and Section 2.790(a)(4) for "trade secrets and commercial or financial information obtained from a person and privileged or confidential." The material for which exemption from disclosure is here sought is all "confidential commercial information," and some portions also qualify under the narrower definition of "trade secret," within the meanings assigned to those terms for purposes of FOIA Exemption 4 in, respectively, Critical Mass Energy Project v. Nuclear Regulatory Commission, 975F2d871 (DC Cir. 1992), and Public Citizen Health Research Group v. FDA, 704F2d1280 (DC Cir. 1983).
5. Some examples of categories of information which fit into the definition of proprietary information and which are applicable here are:
  - a) Information that discloses a process, method, or apparatus, including supporting data and analyses, where prevention of its use by the Company's competitors without license from Exelon Generation Company, L.L.C. constitutes a competitive economic advantage over other companies;

- b) Information which, if used by a competitor, would reduce his expenditure of resources or improve his competitive position in the performance of outages or the design, manufacture, shipment, installation, assurance of quality, or licensing of a similar product.
6. The information sought to be withheld is being submitted to the U. S. Nuclear Regulatory Commission ("NRC") in confidence. The information is of a sort customarily held in confidence by Exelon, and is in fact so held. Its initial designation as proprietary information, and the subsequent steps taken to prevent its unauthorized disclosure, are as set forth in (7) and (8) following. The information sought to be withheld has, to the best of my knowledge and belief, is not available in public sources. All disclosures to third parties including any required transmittals to NRC, have been made, or must be made, pursuant to regulatory provisions or proprietary agreements which provide for maintenance of the information in confidence.
  7. Initial approval of proprietary treatment of a document is made by the Vice President, Special Projects, the person most likely to be acquainted with the value and sensitivity of the information in relation to industry knowledge.
  8. The procedure for approval of external release of such a document typically requires review by a Vice President, Exelon Generation, or her/his designee, for technical content, competitive effect, and determination of the accuracy of the proprietary designation. Disclosures outside Exelon Generation Company, L.L.C. are limited to regulatory bodies, customers, and potential customers, and their agents, suppliers, and licensees, and others with a legitimate need for the information, and then only in accordance with appropriate regulatory provisions or proprietary agreements.
  9. The information identified in paragraph (2) is classified as proprietary because it contains nuclear fuel design information related to the Pebble Bed Modular Reactor (PBMR).
  10. Public disclosure of the information sought to be withheld is likely to cause substantial harm to Exelon's and others contributing to the PBMR Project competitive position and foreclose or reduce the availability of profit-making opportunities. The nuclear fuel design issues related to the PBMR provide commercial value to Exelon and its partners. The research, development, engineering, analytical, and NRC review costs comprise a substantial investment of time and money by Exelon and its partners.

Exelon's and its partners' competitive advantage will be lost if its competitors are able to use the design information.

The value of this information would be lost if the information were disclosed to the public. Making such information available to competitors without their having been required to undertake a similar expenditure of resources would unfairly provide competitors with a windfall, and deprive Exelon of the opportunity to exercise its competitive advantage to seek an adequate return on its large investment.

11. She has read the foregoing affidavit and the matters stated therein are true and correct to the best of her knowledge, information and belief.

  
Marilyn C. Kray  
Marilyn C. Kray

Subscribed and sworn to  
before me this 24<sup>th</sup> day  
of May 2002.

Vivia V. Gallimore

Notary Public

Notarial Seal  
Vivia V. Gallimore, Notary Public  
Kennett Square Boro, Chester County  
My Commission Expires Oct. 6, 2003

Member, Pennsylvania Association of Notaries

## **Attachment 2**

**-- Non- Proprietary Version--**

**"PBMR Nuclear Fuel"**

Revision 2

63 pages

Submitted May 24, 2002

## **PBMR NUCLEAR FUEL**

**DOCUMENT NO.: 010520-425**

**Revision: 2**

**Document Status: Approved**

**Applicable Baseline: DDBL**

**Non-Proprietary Version**

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**Management Authorization**

<b>Functionary</b>	<b>Date</b>	<b>Action</b>	<b>Signature</b>
Reactor Experiment and Fuel Qualification Senior Physicist		Reviewed	
			J Venter
Reactor Engineering Manager		Reviewed	
			J Slabber
Product Data Manager		Verified	
			G C Prinsloo
Quality Assurance Manager		Verified	
			D Goode
Power Plant Engineering Manager		Authorized	
			H D Matzner
Fuel Manager		Authorized	
			S Marshall
Nuclear Safety and Licensing Manager		Authorized	
			A P George

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## 1. INTRODUCTION

The purpose of this document is to provide a summary description of the Pebble Bed Modular Reactor (PBMR) fuel design and fuel development history.

Detailed core and fuel calculations have been performed as part of examining the feasibility of the PBMR project using the preliminary reactor design. Changes to design details and parameters are likely to be made as part of the detailed reactor design process. These changes will likely affect the fuel analytical results. Numerical values presented in this report are considered to reflect the order of magnitude of values that will be seen in the final design, but final design calculations have yet to be performed. The status of fuel design described in this document is that which applied in the middle of 2001 and will not change.

**Section 2** of this report describes PBMR fuel element design, manufacture, and specifications.

**Section 3** of this report describes TRISO fuel particle development history and irradiation testing results.

## 2. PBMR NUCLEAR FUEL

### 2.1 Fuel Element Design

The PBMR fuel (**Figure 1**) is based on a high quality German moulded graphite sphere design containing coated fuel particles. A TRISO fuel particle consists of low enriched uranium dioxide (the kernel) coated with four layers: (1) a porous pyrolytic carbon ("buffer") layer (2) a dense pyrolytic carbon layer (3) a silicon carbide (ceramic) layer and (4) another layer of dense pyrolytic carbon. The porous carbon buffer layer accommodates any deformation caused by density changes that the kernel may undergo as a result of fission product build up. It also accommodates gaseous fission products released from the kernel so as to limit the pressure in the coated particle. The dense pyrolytic carbon and silicon carbide layers provide barriers that contain the fission products.

The coated particles [**Proprietary info deleted**] are embedded in a graphite matrix and moulded into a 50 mm sphere, called the fuel zone. Adding a 5 mm thick fuel free graphite matrix zone makes up the fuel sphere, which has an outer diameter of 60 mm. Each sphere has 9g of uranium contained in the coated particles. A total of 330,000 fuel spheres are contained in the core as it existed in the middle of 2001.

### 2.2 Fuel Manufacture

The PBMR fuel manufacturing process steps and principles have been specified to be the same as those that were used in the German NUKEM plant which manufactured fuel for the AVR and THTR. Where important from a process equivalence aspect, the facility designs include the same type and basic design of manufacturing equipment (i.e. the same "working parts") as those that were used in the NUKEM plant. State-of-the-art instrumentation and control equipment and systems have been specified for the PBMR fuel plant to provide better control of processes.

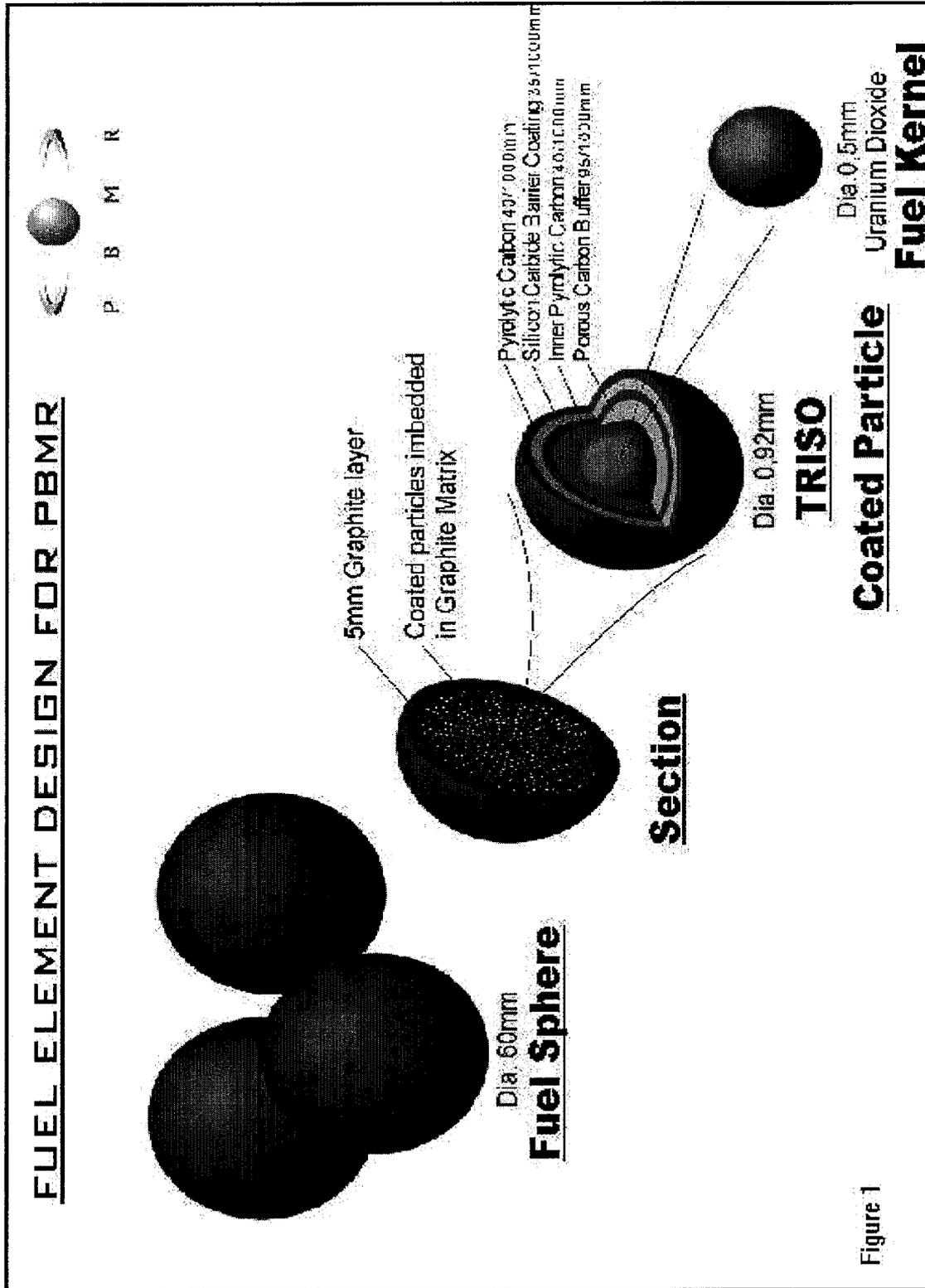


Figure 1: FUEL ELEMENT DESIGN FOR PBMR

## 2.3 Process Description

The fuel manufacturing processes are described in the following sections under the headings of:

- Kernel formation;
- Coating of kernels to make coated particles;
- Mixing of matrix graphite powder;
- Fuel sphere manufacture; and
- Graphite sphere manufacture.

## 2.4 Kernel Formation

Enriched uranium will be delivered to the fuel plant at Pelindaba in the form of  $U_3O_8$  powder. The level of enrichment will be below 10% to start with, and will always be below 20%. This is considered "low enriched uranium", or "LEU". There will be about 3 tonnes of uranium in the core of each PBMR, and about 1 tonne of uranium will be replaced with fresh fuel per year of operation. The LEU will be transported to Pelindaba in special transport containers. These containers will comply with the regulations for the safe transport of radioactive materials published by the International Atomic Energy Agency (IAEA) and adopted by most countries.

The first phase in kernel production is the preparation of the casting solution. This is carried out by first weighing the uranium oxide required for one batch. It is then dissolved in nitric acid, filtered and stored in tanks as uranyl nitrate solution. To make up the casting solution, defined quantities of additives are added to the uranyl nitrate solution.

The casting apparatus consists of a number of small diameter pipes, through which the casting solution is pumped. Each pipe has a vibrator to ensure that uniform small droplets leave the nozzle under gravity. The droplets become spherical while falling through air, then harden on their surface while falling through ammonia gas, and then fall into an ammonium hydroxide solution where the chemical transformation to ammonium diuranate (ADU) continues.

The complexity of the casting process lies in ensuring that each droplet is completely spherical. This depends on careful casting solution make-up and temperature control, consistent pressure at the outlet nozzle, and the avoidance of disturbing air currents during sphere formation.

The kernels are removed from the bottom of the casting vessel and go through separate aging, washing and drying operations.

The last stages of kernel production involve heat treatment. Firstly, the kernels are spread out onto trays and calcined in air in a furnace to convert ADU to  $UO_3$ . Then the kernels are reduced to  $UO_2$  (uranium dioxide) by heating them in hydrogen. Finally, the kernels are sintered to their final density at high temperature.

Kernels which leave the sintering furnace are then sieved and sorted where reject material not conforming to the correct shape or size is removed. The conforming product is transported to dedicated storage rooms to await laboratory analysis and QC acceptance.

## **2.5 Coating of Kernels to make Coated Particles**

Four coating layers are deposited on kernels in a heated furnace by a chemical vapour deposition (CVD) process. Flowing gases in the furnace suspend the kernels so that they form a fluidized bed. Gases are chosen which decompose at high temperatures and deposit certain of their constituents on the surfaces of the kernels. The materials of the layers formed by this process are described as pyrolytic, because they are formed by chemical decomposition that is brought about by heat.

Coating is carried out on 5kg batches of kernels at a time in a single vessel. The four layers are deposited in succession in one operation. A batch of kernels is introduced into the coater, and the kernels are fluidized and heated to defined temperatures.

The porous carbon buffer layer is deposited using acetylene gas. A mixture of acetylene and propylene is then used to deposit the second, dense pyrocarbon layer. The third layer, silicon carbide (SiC), is deposited using hydrogen as the carrier gas with small quantities of methyltrichlorosilane (MTS).

The last layer is again a dense pyrocarbon layer, deposited using a gas mixture of acetylene and propylene.

At each stage, samples are collected from the coater and sent for analysis to verify that each layer is of correct thickness.

During carbon deposition the off gas from the coater passes through cyclones to remove soot, and the exhaust gases are then filtered before being discharged via the plant ventilation system. The soot is removed from this system and sent to uranium recovery for further treatment.

At the end of each coating cycle, the batch of coated particles is removed, sieved and sorted to remove over and undersized as well as non-spherical product. At this stage each particle is about 920  $\mu\text{m}$  in diameter. The coated particles are easier to handle than the kernels because the uranium has been sealed off, and they do not pose a risk of contamination.

## **2.6 Mixing of Matrix Graphite Powder**

The Matrix Material Production Facility provides a granulated homogenous mixture of natural and electro graphite powder with a resin binder.

One part electro graphite and four parts natural graphite are blended in a mixer. The phenolic resin binder is dissolved in methanol. The graphite and the binder are then proportioned and mixed in a kneader. The product is extruded as smooth spaghetti shaped material, which is then laid on trays and placed in vacuum drying ovens. The methanol solvent is driven off and condensed for partial re-use. After a period of time the trays are removed and the hard "spaghetti" is crushed in a roller mill.

Matrix graphite powder is combined with recycled graphite swarf from the sphere production process and further milled to a powder. It is then placed in storage bins in large batch sizes. This material is used for fuel sphere and graphite sphere production.

A small portion of the matrix graphite powder is milled in a second mill to produce a finer material that is used for over-coating the coated particles.

## **2.7 Fuel Sphere Manufacture**

A fuel sphere is approximately the size of a tennis ball (60 mm) and contains approximately **[Proprietary info deleted]** coated particles in the central core of the sphere. The inner sphere or core is covered with a protective layer of matrix material to ensure a particle-free zone between the spheres.

Batches of coated kernels are first over-coated with matrix graphite powder. This layer minimizes the risk of coated particles touching each other and cracking when the sphere is later pressed under high pressure. The over-coating takes place in a "sweetie barrel" in the same way as confectioners or pharmaceutical manufacturers coat sweets or pills. The particles are then removed and gently dried in ovens to remove any solvents. They are then once again sieved and sorted to remove over and undersized as well as non-round product.

The next phase involves the formation of the inner sphere or core containing a measured quantity of approximately **[Proprietary info deleted]** over-coated particles. The over-coated particles are mixed with a quantity of matrix graphite powder and placed in a sphere-shaped mould. They are then pressed in an industrial press. The resulting sphere can then be easily handled as required in the remainder of the manufacturing process.

The next manufacturing stage takes place in a different set of moulds. A measured quantity of crushed matrix graphite is placed into the lower mould and a hollow shape formed in the center. The inner sphere is then placed into the hollow shape and the upper mould is placed on top. Once a second portion of matrix graphite powder is added through a hole in the top, the inner sphere will be located in the middle of the mould surrounded by a layer of matrix graphite powder. The matrix graphite powder forms a shell of fuel-free matrix graphite powder around the inner sphere. The fuel sphere is compressed at high pressure to form the final sphere).

The next operation involves machining the outer graphite surface on a lathe. X-ray results are used to indicate the position of the inner sphere and these co-ordinates are fed into the lathe, which automatically calculates how to machine the outer surface so as to ensure that the inner sphere is located in the centre.

The last phase in the process is the heat treatment of the fuel spheres. There are two heat treatment processes, one referred to as carbonizing, which involves carbonizing the phenolic resin in the matrix graphite powder, and the other is annealing, where any residual impurities are removed under vacuum at a temperature approaching 2000°C.

The machined spheres are placed into circular steel trays which are stacked to form a columnar shape, and then lifted by overhead crane into a top loading carbonizing furnace. This heat treatment is done in an inert atmosphere.

In the annealing process, the spheres are placed into a top loading annealing furnace and heat treated under vacuum.

The final step is the inspection stage where dimensions are checked; the surfaces are visually inspected for defects and the spheres are weighed for mass control. The spheres are also X- rayed before placed in final storage containers awaiting final QA release.

## **2.8 Graphite Sphere Manufacture**

Moulded or pressed graphite sphere production is essentially the same as fuel sphere production. The inspection, X-raying, lathe machining and carbonizing steps are all followed in exactly the same way as for fuel spheres. In practice, two identical production lines side by side may be provided, so that either line can be used for uranium fuel or graphite spheres.

## **2.9 Manufacturing Defects**

High Temperature Gas-cooled Reactor (HTGR) fuel elements present the following three potential fission product sources:

- Particles with defective TRISO coatings.
- Uranium contamination of the outer pyrocarbon layer.
- Uranium contamination of matrix material.

Defective particles may arise during the cold pressing of spherical fuel elements, when the TRISO coated particles embedded in the matrix material experience mechanical contact, in spite of their protective over-coating, to the degree that the layers crack.

Fabrication-related defects are determined by the burn-leach test. This method measures the free uranium fraction covering the total uranium quantity in the matrix material (tramp uranium), outer pyrolytic carbon layers and defective particles. In this test, the fuel element matrix, including the accessible pyrocarbon layers, is burned at 850 °C. The residue, including all the particles (with or without defective layers), is treated with nitric acid at 95 °C and the uranium content of the acid is determined by fluorimetry.

## 2.10 Uranium Recovery

The uranium recovery plant is designed to recover uranium from uranium containing scrap if economically feasible. Other materials including contaminated soot and graphite coater parts will be incinerated.

All uranium scrap is heat treated to oxidise to  $U_3O_8$ . If the  $U_3O_8$  is contaminated with impurities it is processed by dissolution in nitric acid followed by precipitation as a peroxide. If it is within the impurity specification,  $U_3O_8$  is processed mechanically. Fuel sphere scrap as well as contaminated graphite powder and soot are placed onto trays and fed into an incineration furnace and heated to  $1000^\circ\text{C}$ . This ensures that all carbon is burnt off and the resulting uranium oxide is then stored for further treatment. Coated particle scrap is crushed to destroy the silicon carbide layer so that uranium oxide becomes accessible. Lastly, kernel scrap is heated to  $850^\circ\text{C}$  in an oxidizing furnace to ensure that the uranium is converted to  $U_3O_8$ . The product is then placed into temporary storage and further processing is either via the dissolving route (out of specification  $U_3O_8$ ) or the milling route (in specification  $U_3O_8$ ).

The dissolving route involves dissolving  $U_3O_8$  in nitric acid in a reactor, following which the solution is filtered to remove any solid particles. The uranium nitrate concentrate is then stored in tanks where it is diluted to enhance the subsequent precipitation process. The diluted stream is then pumped into a precipitation reactor where hydrogen peroxide is added. Uranium peroxide is precipitated. The precipitate is filtered, washed, dried and calcined to form  $U_3O_8$ .

Subsequent unit operations involve milling, sieving and the blending of different batches in order to obtain homogeneous  $U_3O_8$  powder suitable for re-cycling.

## 2.11 PBMR Fuel Specifications and Measurements

**Table 1** contains the fuel specifications for PBMR fuel. **Table 2** contains the characteristics to be measured and methodology. **Table 3** contains the characteristics of the fuel to be measured prior to release by Quality Control.

**Table 1: DESIGN SPECIFICATION FOR PBMR FUEL ELEMENT**

Fuel Element Component	Specified Characteristic	Specification
Kernels	Stoichiometry	"[Proprietary info deleted]"
	Diameter	"[Proprietary info deleted]"
	Sphericity	The ratio between the maximum diameter $d_M$ and the minimum diameter $d_m$ shall fulfil the following condition: "[Proprietary info deleted]"
	Density	The average density "[Proprietary info deleted]"
Coated Particles	Coating Thickness	Buffer Layer (PyC): $\bar{x} = 95 \mu\text{m}$ Inner Pyrocarbon Layer (PyC): $\bar{x} = 40 \mu\text{m}$ Silicon Carbide Layer (SiC): $\bar{x} = 35 \mu\text{m}$ Outer Pyrocarbon Layer (PyC): $\bar{x} = 40 \mu\text{m}$
	Coating Density	<i>The average layer densities shall be as follows:</i> Buffer Layer (PyC): $\bar{x} \leq 1.05 \text{ g/cm}^3$ Inner Pyrocarbon Layer (PyC): $\bar{x} = 1.90 \text{ g/cm}^3$ Silicon Carbide Layer (SiC): $\bar{x} \geq 3.18 \text{ g/cm}^3$ Outer Pyrocarbon Layer (PyC): $\bar{x} = 1.90 \text{ g/cm}^3$
	Anisotropy for Layers 2 and 4	The average layer anisotropy shall be as follows: Layer 2 (ILTI Layer): "[Proprietary info deleted]" Layer 4 (OLTI Layer): "[Proprietary info deleted]"
Fuel Elements	Uranium loading per sphere	9 grams
	Particles per fuel sphere	"[Proprietary info deleted]"
	Boron equivalent	"[Proprietary info deleted]"
	Ash content	"[Proprietary info deleted]"
	Lithium content	"[Proprietary info deleted]"
	Free uranium fraction	"[Proprietary info deleted]"
	Carbon content	"[Proprietary info deleted]"
	Diameter	60 mm
	Thickness of fuel free zone	5 mm
	Coated particles in fuel free zone	"[Proprietary info deleted]"
	Drop strength	"[Proprietary info deleted]"
	Crush strength	"[Proprietary info deleted]"
	Abrasion	"[Proprietary info deleted]"
Corrosion rate (mean)	"[Proprietary info deleted]"	
Thermal conductivity	"[Proprietary info deleted]"	
Thermal expansion anisotropy	"[Proprietary info deleted]"	

**Table 2: CHARACTERISTICS MEASURED AND METHODS (KERNELS, COATED PARTICLES)**

<b>Product</b>	<b>Characteristic</b>	<b>Method</b>
UO <sub>2</sub> Kernels	Diameter	Optical particle size analyzer
Coated Particles	Buffer layer thickness	Metallography and image analyzer
	Buffer layer density	Particle size analyzer and weigh-scale
	ILTI layer thickness	Metallography and image analyzer
	ILTI layer density	Gradient column
	SiC layer thickness	Microradiography
	SiC layer density	Gradient column
	OLTI layer thickness	Microradiography
	OLTI layer density	Gradient column

**Table 3: CHARACTERISTICS MEASURED FOR QC RELEASE**

U <sub>3</sub> O <sub>8</sub>	Uranium Enrichment Content
	Isotopic Content
	Impurities
	Stoichiometry
	Uranium Content
	Equivalent Boron Content
	Moisture Content
	Particle Size
UO <sub>2</sub> Kernels	Diameter
	Density
	Sphericity
	Equivalent Boron Content
	Stoichiometry
Coated Particles	Buffer layer thickness
	Buffer layer density
	ILTI layer thickness
	ILTI layer density
	SiC layer thickness
	SiC layer density
	OLTI layer thickness
	OLTI layer density
	Anisotropy of ILTI and OLTI layers
	Unconfined Uranium (Burn-leach)
	Isotopic Content
	Uranium Content
	Uranium Enrichment
Fuel Spheres	Uranium Enrichment (Calculated from coated particle results)
	Uranium Content (Calculated from coated particle results)
	Equivalent boron content of matrix material (plus kernels)
	Ash content of matrix material
	Lithium content of matrix material
	Unconfined Uranium (burn-leach)
	Carbon content
	Sphere Diameter
	Fuel-free zone shell thickness
	Surface Defects
	Drop Strength
	Crushing Strength
	Thermal Conductivity of matrix material
	Anisotropy of matrix material
	Abrasion of matrix material
	Corrosion of matrix material
	Density of matrix material

### 3. FUEL DEVELOPMENT HISTORY & IRRADIATION TESTING

#### 3.1 Fuel Development History

“The investigation of fundamental characteristics of HTGR fuel has been in progress for 30 years. For all countries' reference concepts, the fuel is based on TRISO coated fuel particles with low-enriched uranium. Two directions for the fuel element design have been pursued, the block type in Japan and the US, and the spherical fuel element in Germany, Russia and China. The design of modern HTGR is based on high-quality fuel. The most important goal in the improvement of the manufacturing process of the fuel element is to reduce the coated particle defect fraction and to minimise the uranium contamination. With the burn-leach technique defective particles and the level of contamination can be measured.” (Opening paragraph of [16] ).

This report highlights the history of German HTGR fuel manufacture because of its direct relevance to PBMR spherical type fuel. HTGR fuel programmes in other countries are summarised below:

- The United Kingdom Atomic Energy Authority development programme, which functioned from 1961 to 1974 in support of the Dragon project, produced block type fuel elements. Although it was closed long before the US and German programmes, this programme made important progress in the establishment of processes for the production of spherical carbide and oxide kernels, and of pyrolytic carbon and silicon carbide coatings.
- The fuel manufacturing plant in the USA supplied coated particle fuel initially in rods and later in the block type. It was dismantled when the 2 HTGRs in the USA (Peach Bottom and Fort St. Vrain) were closed.
- The fuel manufacturing equipment in Russia has remained even though the programme was closed. No fuel was supplied to HTGR reactors because none was ever built in Russia. Extensive experience in reactor experiments was accumulated, however, over broad temperature and burn up ranges. The reference Russian particle design consists of an LEU  $UO_2$  kernel with a TRISO coating.
- The only industrial-sized fuel plant that is operating at present is in Japan. It was constructed in 1992, and is relatively small, with a claimed coated particle throughput of 1500 kg(U)/a. The 900 kg (U) of block type fuel elements for the 30 MW (th) High Temperature Test Reactor (HTTR), which was first made critical in November 1998, were manufactured from 1995 to 1997. The HTTR fuel consists of LEU  $UO_2$  kernels (600  $\mu$  m diameter) in TRISO coated particles that are pressed with graphite to form cylindrical compacts. These are assembled into fuel rods, or pins, which are fitted into graphite blocks.

- The Chinese fuel plant is very small (perhaps 120 kg (U)/a) and it manufactures spherical fuel elements of the same design as the German fuel. The Chinese fuel is currently undergoing irradiation tests in the IVV-2M reactor in Russia. The Chinese fuel also consists of LEU  $UO_2$  kernels in TRISO coated particles that are moulded into graphite spheres. The 10 MW (th) HTR-10 reactor became critical for the first time in December 2000.

### 3.2 Manufacturing Experience

The German NUKEM plant supplied in total more than one and a quarter million spherical fuel elements to the 46 MW (th) AVR and the 750 MW (th) Thorium High Temperature Reactor (THTR) before the reactors and the fuel plant all stopped operating in 1988. Excellent product performance and continuous improvement of the fuel element design and manufacture were achieved before the fuel plant was closed [14].

The German HTGR programme started with Highly Enriched Uranium (HEU) in (Th, U)  $C_2$  kernels coated with a low-density, pyrolytic carbon "buffer" layer, and a high -density, isotropic, pyrolytic carbon layer. The outer layer was deposited at high temperature, and was described as "HTI" (i.e. high- temperature isotropic). These coated particles were called "BISO" particles. The first fuel spheres made by NUKEM with HTI BISO particles were loaded into the AVR reactor in October 1968.

The kernels were changed to HEU (Th,U) $O_2$  in 1975 for the German THTR reactor, still with HTI BISO particles. This type of fuel was used throughout the operating life of the THTR, which became critical in 1983, was connected to the grid in 1985, and was stopped in 1988. Approximately 1 million fuel spheres were manufactured in total by NUKEM for the THTR, which operated with a mean coolant outlet temperature of 750 °C [14].

The reference coated particle design in Germany was changed to LT I (low - temperature isotropic) TRISO between 1975 and 1980. TRISO coatings consist of the following layers: pyrolytic carbon buffer, Inner LTI ("ILTI") pyrolytic carbon, silicon carbide, and Outer LTI ("OLTI") pyrolytic carbon. This was found to provide a greater degree of resistance to fast -neutron bombardment and a significantly higher degree of fission product retention than the HTI BISO particle. The silicon carbide layer became the definitive fission product barrier in this design, and contamination of the outer pyrolytic carbon layer with heavy metal was greatly reduced. This change required a long-term manufacturing development and demonstration programme by NUKEM [14].

In 1980, the reference kernel material in Germany was changed to LEU  $UO_2$ . This resulted from non- proliferation considerations and difficulties anticipated in the acquisition of HEU. The switch to

LEU required some changes in the NUKEM manufacturing plant, but these were easier to implement than the change to LTI TRISO particles [14].

All of the foregoing types of fuel manufactured by NUKEM were loaded into the AVR reactor, which operated from 1967 to 1988. The total number of fuel spheres made by NUKEM for the AVR amounted to more than a quarter of a million. The following reloads contained LEU UO<sub>2</sub> TRISO coated particles: No. 19 (24,615 fuel spheres, loaded in July 1982); No.21 (20,250 fuel spheres, loaded in February 1984); and No.21 -2 (8, 740 fuel spheres loaded in October 1987) [16]. The average coolant outlet temperature of the AVR was 950°C from February 1974 until operation of the reactor was stopped at the end of 1988.

**Table 4: RESULTS OF BURN-LEACH TESTS ON LEU-TRISO FUEL IN GERMANY**

Fuel Element Population Detail	LEU PHASE 1	AVR 19	AVR 21	AVR 21-2	Proof Test
Year of Production	1981	1981	1983	1985	1988
Number of Fuel Elements Produced	<100	24 600	20 500	14 000	<200
Number of Fuel Element Lots	-	14	11	8	-
Coating Batch Size	5 kg	5 kg	3 kg	3 kg	5 kg
Number of Coating Batches	1	65	54	29	8
Number of Coated Particles per Fuel Element	16 400	16 400	9 560	9 560	14 600
U <sup>235</sup> Enrichment	9,8%	9,8%	16,7%	16,7%	10,6%
Free Uranium Fraction	35 x 10 <sup>-6</sup>	50,7 x 10 <sup>-6</sup>	43,2 x 10 <sup>-6</sup>	7,8 x 10 <sup>-6</sup>	13,5 x 10 <sup>-6</sup>
Number of Burn-Leach Tests	5	70	55	40	10
				Total Elements	> 59,100
				Total Particles	733 million

Two series of fuel spheres containing LEU, UO<sub>2</sub>, TRISO coated particles were manufactured by NUKEM for irradiation testing under controlled conditions in materials testing reactors. The LEU Phase 1 fuel spheres were manufactured in 1981, and were irradiated under a variety of different conditions in several reactors. The so-called "Proof Test" fuel was manufactured in 1988, and 8 spheres were irradiated under conditions simulating the HTR MODUL reactor conditions in the HFR Petten materials testing reactor [14], [16]. The PBMR follows the nuclear design principles, particularly the coated particle design of the German HTR -Modul, which had reached the detailed design phase before work was stopped in the late 1980's.

The fuel sphere moulding process developed during the German programme consisted of pressing over-coated coated particles mixed with matrix graphite powder at room temperature and at high

pressures in rubber moulds. The over-coating of coated particles with matrix graphite powder helped to keep the particles apart during moulding, thus reducing damage to particle layers [15]. Automating the over-coating process was also found to minimise damage to the coated particles during pressing [16].

The last LEU TRISO fuel spheres manufactured by NUKEM were the AVR 21- 2 reload and the Proof Test elements. These exhibited free uranium values, as measured by burn -leach tests of 50 fuel spheres, of approximately 11 parts per million [1]. This result epitomised the high level of quality achieved by NUKEM in the quest "to reduce the coated particle defect fraction and to minimise the uranium contamination".

The coolant outlet temperature of the PBMR is designed to be 900°C, whereas that of the HTR MODUL was 750°C. It is noted, however, that the AVR reactor ran at a mean coolant outlet temperature of 950° C much of the time from 1974 until it was shut down in 1988. The LEU TRISO fuel that was used in the AVR operated well at this temperature, thus confirming that the fuel design of the AVR reload 21-2 and the Proof Tests was adequate under the elevated temperature conditions.

### **3.3 Irradiation Qualification of LEU-TRISO Fuel**

The factors determining the integrity of fuel pebbles in the PBMR core are:

- burn-up;
- fast neutron dose; and
- temperature.

With increasing burn-up of fuel pebbles in the PBMR core, fission products are formed within the kernels of coated particles. The gaseous fission products accumulating within the kernels give rise to an internal pressure, which is contained by the SiC layer that acts as a pressure vessel. As soon as the stress induced in the SiC layer due to the internal pressure exceeds the tensile strength of the layer, the layer will fail.

In order to protect the SiC layer against pressure vessel failure, a buffer layer of pyrocarbon is deposited around each kernel. The buffer layer is porous and provides free volume to accommodate gaseous fission products, thus reducing the internal pressure resulting from these fission products. Fission product release from fuel elements during normal operation is determined by specifying a maximum burn-up value for PBMR fuel elements, and by measuring the burn -up of

each fuel element after each cycle, to ensure that the maximum burn-up value is not exceeded during operation.

Irradiation by fast neutrons ( $E > 0,1$  MeV) causes damage in the pyrocarbon layers, leading to an initial shrinkage of the layers. This effect is beneficial because it induces a net compressive stress in the SiC layer. At higher fast neutron doses the pyrocarbon layers start to expand, eventually reaching and surpassing their original dimensions. This reverses the above process, producing a tensile stress in the SiC layer. The coated particle design for the PBMR is such that tensile stresses that might lead to the failure of the SiC layer occur at fast neutron doses that would only be reached at burn-up values that are higher than the design burn-up for the fuel.

An increase in the temperature of a coated particle will lead to an increase in the internal pressure within the particle, due to the thermal expansion of the gaseous fission products contained in the particle. This will cause an increase in the tensile stresses in the different coating layers that might lead to increased particle failure. At high temperatures ( $>2000$  °C) SiC will dissociate leading to a total loss of the SiC layer's fission product retention capability. Limiting the maximum temperature experienced by the coated particles in the PBMR core under normal operation and under upset event conditions will prevent an unacceptable failed particle fraction in the reactor core.

In Germany, a number of pebble bed reactor concepts were developed around the TRISO fuel design. Irradiation qualification of TRISO fuel elements was therefore done in two phases.

- a) Phase 1 was a generic phase, during which prototype material was irradiated under conditions that covered the whole range of expected parameters envisaged for all reactor concepts that were designed around LEU-TRISO fuel elements. Thus Phase 1 experiments provided the basic data needed to determine fission product release source terms under all foreseeable reactor conditions.
- b) Phase 2 experiments were to be reactor-specific, using typical production material and simulating actual reactor operation conditions as closely as allowed by the materials testing reactor that is used for irradiation. Thus Phase 2 experiments were to demonstrate the transferability of phase 1 data to the reactor concept under investigation, and also widen the statistical database on fission product release.

### **3.4 Phase 1 Experiments**

The main purpose of irradiation measurements is the identification and quantification of all mechanisms that can possibly contribute to the release of fission products from fuel elements. Because of the negligible uranium contamination of TRISO coated particles and the excellent

containment properties of intact-coated particles for most fission products, the most important source is the release from defective coated particles. Coated particles may become defective due to production processes, irradiation under extreme irradiation loads, or exposure to extreme temperature loads.

Quality control measurements, irradiation data, and post-irradiation annealing tests can be used to determine the failed particle fraction resulting from each of the above causes respectively.

If uranium contamination of coated particles and matrix graphite is negligible, the only remaining source of fission product release is diffusion from intact-coated particles, and from defective coated particles. Irradiation experiments were designed to investigate both of these release mechanisms with varying temperature.

The experiments performed for LEU-TRISO reference coated particles are shown in **Table 5**. All tests shown in **Table 5** contained particles of batch EUO 2308 (NUKEM/HOBEG) produced in 5 kg batches in a fluidised bed particle coater.

**Table 5: PURPOSE OF PHASE I IRRADIATION EXPERIMENTS FOR LEU-TRISO FUEL**

Purpose	Irradiation Experiment LEU-TRISO Phase I
Coated Particle Quality (Failed Particle Fraction)	HFR-P4 SL-P1
Fuel Element Quality	HFR-K3
Fission Product Transport from: <ul style="list-style-type: none"> <li>• Intact particles</li> <li>• Defective particles</li> <li>• Fuel elements (AVR small-scale test)</li> </ul>	FRJ2-P27 FRJ2-P28 FRJ2-K13
AVR large-scale test	AVR-19 AVR-21

For technical reasons, not all irradiation experiments were conducted with full-size fuel elements. In tests HFR-P4 and SL-P1, so-called 'small spheres' were used. These spheres were produced using materials and processes identical to those used for normal fuel elements. However, the spheres had a fuel containing zone of only 20 mm diameter, and a fuel-free zone of thickness 20 mm. Cylindrical test samples that would fit into the test rigs were machined from the small spheres. The irradiation rigs used in irradiation experiments in Jülich (FRJ2) were designed for the irradiation of compacts. Cylindrical compacts with a diameter of 28 mm and height of 30 mm, with a higher coated particle density than fuel elements, were hot-pressed in matrix graphite in a steel die. Because of these deviations in geometry, production method and materials from fuel elements,

the bedding of particles in the test samples was not strictly representative of that in fuel elements. Nevertheless, the effect of these deviations was considered to be minimal and was neglected.

The most important irradiation target, after temperature, is burn-up. This is because coated particle failure during reactor operation is determined by tensile stress in the SiC layer caused by the burn-up dependent fission product pressure within the coated particles. It was also known from irradiation experiments on HTI-BISO coated particles that fast neutron fluence in combination with temperature effects caused failure of high-density pyrolytic carbon coatings. It is unavoidable that a certain number of pyrolytic carbon coatings are produced during the coating process will not be round. Irradiation by fast neutrons causes further anisotropic structural changes in these particles, which induces tensile stress and causes failure. In the PBMR fuel production process, particles that are not round are removed by sorting before going to the next production step.

For Phase 1 experiments, the target burn-up was set at 10 to 12% Fissions per Initial Metal Atoms (FIMA), which corresponds to 90 000 to 107 500 MWd/tU. A fast neutron fluence of  $6 \times 10^{21}$  n/cm<sup>2</sup> ( $E > 0,1$  MeV) was specified for those experiments designed to measure failed particle fraction, i.e. HFR-P4, SL-P1 and HFR-K3.

To investigate the temperature dependence of particle failure, irradiation temperatures of 1 000 °C and 1 200 °C were specified for most of the tests, although values of 800 °C and 1 300 °C were specified for a few tests. The irradiation temperature was kept constant for the whole duration of the test in most cases. An exception was test FRJ-P28, that contained a number of artificially introduced defective particles. The temperature for this test was varied in order to determine the temperature dependence of fission gas release in more detail.

The Phase 1 experiments is discussed under the following headings:

- Irradiation tests.
- Post-irradiation annealing tests.
- Derivation of failed particle design base.

### **3.5 Phase 1 Irradiation Tests**

The Phase 1 irradiation programme consisted of six experiments performed in three different reactors. Experiments were allocated to different reactors on the basis of the technical capabilities available at each reactor.

Experiments requiring full burn-up and fast neutron fluence were performed in the HFR Petten (HFR-P4 and HFR-K3) and in SILOE Grenoble (SL-P1).

Experiments requiring priority investigations regarding fission product release from intact particles (FRJ2-P27) and fuel elements (FRJ -K13) as well as defective particles (FRJ2-P28) were performed in FRJ2 in Jülich. For these irradiation experiments, it was not essential to reach the full target value for fast neutron fluence.

Coated particles of batch EUO 2308 manufactured by NUKEM/HOBEG were used in all Phase 1 experiments. The defective particles introduced artificially in experiment FRJ2-P28 consisted of fuel kernels similar to those used to produce EUO 2308-coated particles taken from kernel charge UOS 331. The defective particles were coated with only a buffer layer, and then removed from the coater.

By providing kernels with a buffer layer, it was ensured that fission products released from the kernel by recoil are slowed down and transported in exactly the same way as fission products produced in kernels whose high -density coatings have become defective. Experiment FRJ2-P28 is not considered in the statistical analysis, because it contained deliberately introduced defective particles, which is not representative of LEU-TRISO coated particles.

All irradiation rigs were provided with gas loops that were used to measure the Kr-85m release from each capsule in a rig continuously. The birth rate of Kr -85m in each capsule was calculated and from the Release-to-birth (R/B) ratio and knowledge of the R/B ratio for a single defective particle (obtained from test FRJ2 -P28), the number of defective particles in a capsule could be calculated. Initially, broken particles will be detected at start of irradiation.

In parallel with the irradiation tests, a number of compacts, small spheres and fuel elements were also analysed using the burn-leach test, to determine the number of failed particles in unirradiated fuel elements.

The results of Phase 1 tests are shown in **Table 6** . Measured values for Kr-85m R/B ratio and Cs-137 release fractions at the end of irradiation are shown in **Table 7** .

From the data presented in **Table 6** and in **Table 7** , it is clear that, of the 211 834 particles contained in eight fuel elements, 36 small spheres, nine compacts and six coupons, not a single particle failed as the result of irradiation. The tests covered a range from 7.5 to 14.7% FIMA for burn-up, 0.2 to  $8 \times 10^{21}$  n/cm<sup>2</sup> for fast neutron fluence, and 800 to 1 320 ° C for irradiation temperature.

In **Figure 2**, the Burn-up vs. Fast Neutron Fluence graphs calculated for HTR-Modul [3] and PBMR [4] are shown in relation to the Phase 1 values for the same parameters taken from **Table 7**. In the graph, values obtained from the irradiation of complete fuel elements are identified by means of labels showing the test numbers of the fuel elements. Data points without labels were obtained from small spheres, compacts and coupons. From the graph it is clear that PBMR irradiation loads fall well within the burn-up and fast neutron fluence envelope of Phase 1 measurements. Thus it is not intended to perform any generic testing similar to Phase 1 tests for PBMR fuel, based on the premise that locally manufactured fuel will be similar to German fuel.

**Table 6: RESULTS OF PHASE 1 IRRADIATION TESTS ON LEU-TRISO COATED PARTICLES**

Experiment Number	Capsule Number	Sample Number and Form	Temperature (°C)		Burn-up (% FIMA)	Fast Neutron Fluence E>0.1MeV (n/cm <sup>2</sup> ) (x 10 <sup>21</sup> )	Number of Coated Particles in Capsule	Number of Defective Particles		Parallel Test (Burn-leach)		
			Surface	Fuel Element				BOL	EOL	Samples	Number of Particles	Number of Failed Particles
HFR-P4	A/01	12 small spheres	915	940	11.1 to 14.7	5.5 to 8.0	19 572	0	0	5 small spheres	8 155	0
	C/03	12 small spheres	1 050	1 075	9.9 to 14	5.5 to 8.0	19 572	0	0			
SL-P1	-	12 small spheres	780	800	8.6 to 11.3	5.0 to 6.7	19 572	5	5			
HFR-K3	1/A	1 fuel element	1 020	1 200	7.5	4.0	16 400	0	0	5 fuel elements	82 000	3
	2/B	2 fuel elements	700	920	10.0	5.8	32 800	0	0			
	3/C	1 fuel element	1 020	1 220	10.6 9.0	5.9 4.9	16 400	0	0			
FRJ2-K13	1	2 fuel elements	985	1 125	7.5; 8.0	0.2	32 800	0	0			
	2	2 fuel elements	980	1 120	7.9; 7.6	0.2	32 800	0	0			
FRJ2-P27	1	3 compacts, 2 coupons	880	1 080	7.6	1.42	7 272 34	5	5	5 compacts	12 120	0
	2	3 compacts, 2 coupons	880	1 320	8.0	1.67	7 272 34	3	3			
	3	3 compacts, 2 coupons	880	1 130	7.6	1.29	7 272 34	0	0			
						TOTAL	211 834					

**Table 7: Kr-85 R/B VALUES AND Cs- 137 RELEASE FRACTIONS AT END OF LIFE FOR PHASE 1 TESTS [2]**

Experiment Number	Temperature Surface/Centre	Burn-up (% FIMA)	Fast Neutron Fluence E>0.1 MeV ( $\times 10^{21}$ n/cm <sup>2</sup> )	End of Irradiation	
				R/B Kr-85m	Fractional Release Cs-137
HFR-P4/1	915/940	11.1 to 14.7	5.5 to 8.0	8E-08	5E-6
HFR-P4/3	1050/1075	9.9 to 14.0	5.5 to 8.0	9E-08	2E-05
SL-P1	780/800	8.6 to 11.3	5.0 to 6.7	1E-06	5E-06
HFR-K3/1	1020/1200	7.5	4.0	2E-07	9E-06
HFR-K3/2	700/920	10.0	5.8	2E-07	2E-05
HFR-K3/3	700/920	10.6	5.9	2E-07	2E-05
HFR-K3/4	1020/1220	9.0	4.9	3E-07	1E-05
FRJ-K13/1	985/1125	7.5	0.2	2E-08	2E-05
FRJ2-K13/2	990/1150	8.0	0.2	2E-09	2E-05
FRJ2-K13/3	990/1150	7.9	0.2	7E-09	6E-06
FRJ2-K13/4	980/1120	7.6	0.2	7E-09	6E-06
FRJ2-P27/1	880/1080	7.6	1.4	2E-06	2E-05
FRJ2-P27/2	1220/1320	8.0	1.7	1E-05	1E-04
FRJ2-P27/3	1080/1130	7.6	1.3	1E-07	1E-05

### 3.6 Phase 1 Annealing Tests [1], [5]

The cumulative release fraction of Kr-85 as a function of time for fuel elements from Phase 1 tests and some fuel elements that were irradiated in the AVR is shown in **Figure 3** for different annealing temperatures. The annealing was carried out at constant temperature and the results are representative of full-size fuel elements with approximately 16 400 coated particles per element.

The Kr-85 release fraction graphs for annealing at 1 600 ° C (AVR 71/22, HFR-K3/1, FRJ2-K13/2 and FRJ2-K13/4) show that fission gas release rates stayed at very low levels for long annealing times (more than 100 h). This is due to the very low natural uranium contamination of the graphite matrix. It is important to note that not a single particle failed during annealing of irradiated fuel elements at 1 600 ° C. The sudden increase in the release rate for FRJ2 -K13/4 after 138 h is due to the fact that the annealing temperature was raised to 1 800 ° C after being at 1 600 ° C for 138 h. The fuel element was then annealed for a further 100 h at the higher temperature.

The broken curve designated as AVR 74/11 shows the results of annealing irradiated fuel at 1 700 °C. The sudden rise in the curve after 85 h is characteristic of coated particle failure during annealing.

The release curves for annealing at 1 800 °C (AVR 70/33 and HFR-K3/3) show the effect of multiple coated particle failure (failure of SiC pressure vessel). The curve for fuel element AVR 70/33 indicates the failure of approximately 40-coated particles (out of 16 400). The heating curve for AVR 74/10 was very similar to the AVR 70/33 and HFR -K3/3 curves, but is not shown for reasons of clarity.

It should be noted that the Kr-85 release fraction curves for Phase 1 fuel elements irradiated under controlled conditions, and AVR fuel elements taken from an operating reactor, are very similar.

Annealing experiments on samples containing simulated defective particles from test FRJ2 -P28 showed that the behaviour of iodine was similar to the behaviour of krypton and xenon. This means that iodine is almost completely contained within particles with intact coatings, and that the release of iodine is proportional to the defective particle fraction.

Results from annealing tests measuring the release of Cs-137 and carried out on the same fuel elements as those used for Kr-85 release measurements are shown in **Figure 4**. The graphs show that the release pattern for Cs-137 is similar to that of Kr-85. However, the temperature dependence is not so marked as in the case of Kr-85. The Cs-137 release from failed particles is superimposed on the release due to thermally activated diffusion of Cs-137 through intact SiC layers.

It is important to note that in all annealing tests carried out at 1 600 ° C, not a single LEU-TRISO coated particle failed. However, irradiation loads of fuel elements annealed at 1 600 ° C did not simulate burn-up target values for the HTR-Modul very well. On the whole, burn-up values achieved during irradiation tests were lower than target values. However, the fast fluence values for most of the tests were appreciably higher than target values. One example is test element HFR-K3/1, with a fast fluence of  $4 \times 10^{21}$  n/cm<sup>2</sup>, which was markedly higher than the target value for the HTR-Modul.

The results of fuel element tests FRJ2-K13/2 and 4, HFR-K3/1, and AVR 71/22 were used to derive the particle failure fraction at 1 600 ° C. Irradiation and annealing details for these experiments are shown in **Table 8**.

**Table 8: IRRADIATION AND ANNEALING DATA FOR FUEL ELEMENTS USED TO DERIVE PARTICLE FAILURE FRACTION AT 1 600 ° C**

Experiment	Capsule Number	Nominal Irradiation Temperature (°C)		Burn-up (% FIMA)	Fast Neutron Fluence (>0,1 MeV) (n/cm) x 10 <sup>21</sup>	Number of Particles	Annealing History	Number of Failed Particles
		Surface	Fuel Element					
HFR-K3	1	1 020	1 200	7.0	4.0	16 400	500 h at 1 600 °C	0
FRJ2-K13	1/2	990	1 150	8.0	0.2	16 400	160 h at 1 600 °C	0
	2/4	980	1 120	7.6	0.2	16 400	138 h at 1 600 °C	0
AVR 71/22	-	-	-	3.5	-	16 400	500 h at 1 600 °C	0
					Total	65 600		0

### 3.7 Observations Made

The following observations were made from a study of ceramographic images of coated particles from different Phase 1 tests and annealing programmes [1]:

- The appearance of the SiC layer of a coated particle from an irradiated fuel element (6 .5% FIMA) was not much different from that of a coated particle from an unirradiated fuel element. It showed the same amorphous, dense white surface with very few small and dark pores.
- The SiC layer of a coated particle from a fuel element irradiated at 1 200 °C to a burn-up of 8% FIMA and annealed for 138 h at 1 600 °C, was for all practical purposes the same as the SiC layer of an unirradiated coated particle.
- Some evidence of degradation of the inner surface of the SiC layer was visible for coated particles from a fuel element that had been irradiated to 3.5% FIMA and then annealed at 1 600 °C for 500 h.
- Under the same conditions (annealing at 1 600 °C for 500 h), the SiC layer for coated particles from a fuel element with higher burn -up and fast neutron fluence (8% FIMA at 1 200 °C; fast fluence 3.9 x 10<sup>21</sup> n/cm<sup>2</sup>) showed increased degradation. The decrease in density of the layer was much more visible.
- An element of average burn-up (6.2% FIMA) annealed at 1 700 °C for 185 h showed some damage which penetrated the SiC layer.

- Less severe, although penetrating, damage of the SiC layer was observed for a fuel element of low burn-up (1.8% FIMA) annealed at 2 000 °C for 100 h.
- Severe penetrating damage of the SiC layer was observed for a fuel element of average burn-up (5.6% FIMA) annealed for 30 h at 2 100 °C.

### 3.8 Qualitative Conclusions

The following qualitative conclusions were drawn from ceramographic investigations:

- The SiC layer of coated fuel particles, and thus the fission product retention capability of fuel elements, will remain intact under all foreseen normal operating conditions, and also during upset event conditions leading to fuel element heating for long time spans.
- At 1 600 °C, heating for extremely long time spans (longer than 500 h) will result in damage to the SiC coated particles.
- Changes in the fission product retention capability of coated particles become apparent during annealing at 1 600 °C, when the radiation load becomes very high, i.e. irradiation temperatures of 1 200 °C and fast neutron fluence of  $4 \times 10^{21}$  n/cm<sup>2</sup> or more.
- It is not clear which of the mechanisms, burn -up, fast neutron fluence or fuel element temperature is dominant in the degradation of the SiC layer at temperatures of 1 600 °C and higher.
- At heating temperatures of 1 700 °C and higher for relevant time spans, some SiC damage can be expected. Annealing tests on full-size fuel elements from Phase 1 experiments were supplemented with annealing tests on small spheres from HFR-P4 and SL -P1 [6]. The irradiation loads on these tests spanned the expected burn-up values for HTR-Modul a lot better than the fuel element tests.

The irradiation conditions for small spheres from tests HFR -P4 and SL-P1 that were later annealed at different temperatures, are shown in **Table 9**.

**Table 9: IRRADIATION AND ANNEALING CONDITIONS FOR SMALL SPHERES FROM HFR-P4 AND SL-P1**

Test Number	Small Sphere Number	Burn-up (% FIMA)	Fast Neutron Fluence (>0,1 MeV) ( $n/cm^2$ ) x $10^{21}$	Irradiation Temperature (°C)	Annealing Temperature (°C)	Annealing Time (h)
HFR-P4	1.12	11.1	5.5	900	1 600	304
	1.8	13.8	7.2	900	1 600	304
	2.8 (51 $\mu$ m SiC layer)	13.8	7.2	900	1 600	304
	3.7	13.9	7.5	1 050	1 600	304
SL-P1	9	10.69	6.3	790	1 700	304
	10	10.32	6.0	790	1 700	304
HFR-P4	3.12	9.9	5.5	1 050	1 800	279

Four small spheres from HFR -P4 were irradiated to burn-ups of between 11.1 and 13.9% FIMA, fast neutron fluence from  $5.5$  to  $7.5 \times 10^{21}$   $n/cm^2$  and a maximum temperature of  $1\ 050$  °C, and then annealed at  $1\ 600$  °C. The fractional releases of Kr-85, Cs- 134, Cs-137, Sr-90 and Ag110m for these small spheres annealed at  $1\ 600$  °C are shown in **Figure 5**. The Kr-85 release for small sphere 1.12 is very similar to the curves for HFR- K3/1 and AVR 71/22 shown in **Figure 3**, i.e. remaining practically constant between  $10^{-7}$  and  $10^{-6}$  for 304 h. However, the curves for the other small spheres, which had higher burn -ups, show Kr-85 release fractions increasing with time and a pressure vessel failure (prompt jump in release) occurring in the case of small sphere 3.7, after 47 h. The steady increase in release fractions for small spheres with high burn -up is attributed to structural damage occurring in SiC layers, probably due to the diffusion of Cs into the SiC layer. Kr-85 is then released either through transport through the Pyrolytic Carbon (PyC) layers of many particles, or as a consequence of damage to PyC layers in a few individual particles.

Two small spheres from SL-P1 were irradiated to burn-ups of 10.32 and 10.69% FIMA, fast neutron fluence of  $6.0 \times 10^{21}$  and  $6.7 \times 10^{21}$   $n/cm^2$ , and maximum temperature of  $790$  °C and annealed at  $1\ 700$  °C. The Kr-85 release fraction for the test on SL -P1/9 is shown in **Figure 6** . The results were very similar to the fuel element results shown in **Figure 3**.

In **Figure 7** and **Figure 8**, Kr-85 and Cs-137 release curves for the five small spheres annealed at  $1\ 600$  °C are compared with those from six fuel elements with burn-ups in the range 3.5 to 9% FIMA. It is apparent from these graphs that particle coatings only become permeable to fission

products in fuel with burn-ups greater than 11% FIMA. In contrast to Kr-85 release, Cs release increases continuously with time for small spheres with higher burn-up.

A single small sphere from HFR-P4 irradiated to a burn-up of 9.9% FIMA and fast neutron fluence of  $5.5 \times 10^{21}$  n/cm<sup>2</sup> at a temperature of 1 050 °C was annealed at a temperature of 1 800 °C. The Kr-85 and Cs release curves are shown in **Figure 9**. At 1 800 °C, the release fraction for Kr-85 and Cs-137 increases continuously with time. The release is not due to pressure vessel failure, which would produce a prompt jump in release fraction, but to coating failures in a large number of coated particles.

The Cs-137 release from small spheres with burn-ups in the range 10 to 11% FIMA and annealed at temperatures of 1 600, 1 700 and 1 800 °C is shown in **Figure 10**. While the retention at 1 600 °C was good, releases rose rapidly at 1 700 °C, and even more at 1 800 °C during the 300 h of isothermal annealing.

### 3.9 Derivation of Failed Particle Design Base

The most important source for the release of fission products of radiological importance for the HTR-Modul during normal operation and for upset event conditions was the 'free uranium fraction' or 'failed particle fraction' in the fuel elements. In order to quantify free uranium/failed particle fraction, the following three fuel conditions were distinguished:

- Failed particle fraction due to manufacture.
- Failed particle fraction induced by radiation.
- Failed particle fraction induced by high fuel temperatures.

### 3.10 Failed Particle Fraction Due to Manufacture

In the fuel specification for the HTR-Modul, the average value for the free uranium fraction as determined by the burn-leach method for a lot consisting of 10 000 fuel elements was specified as  $6 \times 10^{-5}$ . This value was used as the design value for HTR-Modul fission product release from fresh fuel.

A single defective particle in an HTR-Modul fuel element containing 11 600 coated particles, would represent a failed particle fraction of  $8.6 \times 10^{-5}$ . Thus the specified failed particle fraction corresponds to 0.7 failed particles per HTR-Modul fuel element. (For the PBMR with approximately **[Proprietary info deleted]** fuel particles per sphere, this translates to about one failed particle per fuel sphere).

The actual free uranium values for AVR loading AVR -19, 21/1 and 21/2 fluctuated between  $1 \times 10^{-5}$  and  $5 \times 10^{-5}$ . An expected value of  $3 \times 10^{-5}$  was accepted for new fuel elements.

### 3.11 Failed Particle Fraction Induced by Radiation

Disregarding experiment FRJ2-P28 where failed particles were artificially introduced, the continuous monitoring of fission gas release during irradiation for all other experiments indicated that not a single particle of the 211 834 particles irradiated, failed during irradiation.

In applying the experimental result of 'no particle failure for 212 000 particles tested' to finding a design value for failed particle fraction at EOL for the HTR-Modul, the following was considered:

- In Phase 1 experiments, coated particles from a single coating charge were used, while in the HTR-Modul equilibrium core, particles from approximately 250 to 500 coating charges would be present in the reactor at any one time. The uncertainty introduced by this factor was not considered to be very large. During the initial operation phase of AVR, nine LEU-TRISO fuel elements from the AVR-19 loading were annealed at temperatures between 1 050 and 1 250 °C, and no indication of failed particles was found. The AVR-19 fuel elements contained coated particles from 65 coating charges. Phase 1 experiments were performed in several different reactors, and the Modul target burn -up was not fully reached in some of the experiments, while they were exceeded in other experiments. The same is true for the fast neutron fluence. All coated particles irradiated during Phase 1 tests were taken as a single batch in calculating a radiation-induced failed particle fraction.
- All Phase 1 experiments, except FRJ-P28, were performed at constant temperature and power levels that were mostly higher than the expected values for the HTR-Modul. In the HTR-Modul design, fuel elements are circulated on average 15 times through the core during their lifetime, undergoing continuous changes in temperature and power in the process. This fact was not considered to contribute much to the uncertainty of applying Phase 1 results to the HTR-Modul. The already mentioned AVR annealing tests were performed on fuel elements that had undergone such cycles, although not as many cycles as in the HTR-Modul.

Although 212 000 particles, which is a large but finite sample, were irradiated without a single failure, the expected value calculated from this result will be a value for a finite sample. In the light of the above-mentioned qualitative simplifications, the expected value for the failed particle fraction due to irradiation is calculated with a 95% confidence level, instead of the usual 50% confidence level. The expected failed particle fraction value found from statistical considerations is  $2 \times 10^{-5}$ .

From this conservatively calculated expected value for the failed particle fraction, the design value was calculated by multiplying the expected value by a factor 10. Thus the design value for the failed particle fraction due to fuel irradiation under normal operating conditions for the HTR-Modul is  $2 \times 10^{-4}$ .

The expected and design values for radiation-induced failed particle fraction for the HTR-Modul,  $2 \times 10^{-5}$  and  $2 \times 10^{-4}$  respectively, are valid for fuel elements that have been irradiated to maximum burn-up. No relationship between burn-up and failed particle fraction could be distinguished from the results of Phase 1 experiments. Thus for the HTR-Modul design, the conservative assumption was made that the irradiation-induced failed particle fraction increased linearly with fuel burn-up, starting at zero for fresh fuel and reaching the above value at maximum burn-up. The design value for irradiated fuel for the HTR-Modul was fixed at a point halfway between zero and  $2 \times 10^{-4}$ , (i.e. at  $1 \times 10^{-4}$ ).

### **3.12 Failed Particle Fraction Induced by High Fuel Temperatures**

The results of annealing tests at  $1600^\circ\text{C}$  for irradiated fuel spheres FRJ2-K13/2, FRJ2-K13/4, HFR-K3/1, and AVR 71/22 were used as a basis for the determination of a design value for temperature-induced failed particle fraction for the HTR-Modul.

No increase in fission gas release was noted during annealing at  $1600^\circ\text{C}$  of any one of these fuel elements. In the same way as described above, an expected value for temperature-induced failed particle fraction of  $5 \times 10^{-5}$  with 95% confidence level was derived, taking into account the number of coated particles contained in the fuel elements (65 600). Using the same conservatism as above, a design value of  $5 \times 10^{-4}$  for temperature-induced failed particle fraction was found.

The design value of  $5 \times 10^{-4}$  at  $1600^\circ\text{C}$  is valid for all burn-up and fast neutron fluence values occurring between loading and unloading of the fuel. There is no evidence of any relationship between upset event temperature-induced particle failure and HTR-Modul operational conditions such as burn-up, fast neutron fluence, fuel temperature, and the course of an upset event. In fact, it is clear from annealing tests on experiments HFR-P4 and SL-P1 that even for irradiation loads beyond HTR-Modul loads, and by inference PBMR limiting loads, the temperature-induced particle failure fraction is still small.

In three of the tests used to determine the design value for temperature-induced failed particle fraction, the burn-up did not reach HTR-Modul target burn-up values. The AVR fuel element with a burn-up of 3.5% FIMA was clearly well below the target value. Fuel element 1 from HFR-K3, with a fast neutron fluence of  $3.95 \times 10^{21} \text{ n/cm}^2$ , exceeds the target value of  $2.1 \times 10^{21} \text{ n/cm}^2$  by far. As mentioned before, the design value for temperature-induced failed particle fraction is valid for all

HTR-Modul load conditions. The volume fraction of the core that reaches a temperature of 1 600 °C is very small, and the fraction of fuel elements that have burn-up values exceeding the test values is even smaller. Thus the conservatism (10 times the 95% confidence interval value) in the temperature-induced failed particle fraction will take account of the statistical uncertainties discussed above.

During an abnormal event, there will be a temperature distribution among the fuel elements in the reactor core. Fuel element temperatures will vary between the normal operating temperature and the abnormal event temperature of 1 600 °C. As no particle defects were detected at normal operation conditions or under abnormal event conditions, some conservative but realistic assumptions are called for. It was assumed that particle failure due to temperature effects commences at a temperature of 1 200 °C. The basis for this assumption is the fact that no particle defects were detected for Phase 1 experiments irradiated at temperatures of 1 200 °C. Furthermore, no particle defects were found in fission gas release studies from LEU -TRISO fuel elements annealed at 1 250 °C. It is assumed that the mean failed particle fraction varies exponentially between 'no temperature induced particle failure at 1 200 °C' and the design value of  $5 \times 10^{-4}$  at 1 600 °C. For the purposes of HTR- Modul design calculations, it was assumed conservatively that the failed particle fraction increases instantly with an increase in temperature, not taking into account any failure mechanism that might be time dependent.

The failed particle fraction as a function of fuel temperature that was used as the design base for radiological design for the HTR-Modul is shown in **Figure 11**. The three curves represent fresh fuel, fuel with intermediate burn-up, and fuel that has reached full target burn-up. The curves were generated using some measured values for failed particle fraction, some overly conservative assumptions, and some previous experience. The following three failed particle fraction values were derived from measurements:

- $6 \times 10^{-5}$  for the failed particle fraction for fresh fuel from the fuel specification.
- $2 \times 10^{-4}$  for irradiation induced failed particle fraction- derived statistically from the fact that no particle failures occurred during irradiation of 212 000 particles as  $2 \times 10^{-5}$  (95% confidence level) and multiplied by a factor 10.
- $5 \times 10^{-4}$  for temperature-induced failed particle fraction – derived statistically from the fact that no particle failures occurred during annealing of 65 600 particles as  $5 \times 10^{-5}$  (95% confidence level) and multiplied by a factor 10.

The following assumptions were made:

- There is a linear relationship between failed particle fraction and burn -up.
- Temperature-dependent particle failure commences at a temperature of 1 200 °C and particles fail immediately on reaching a specific temperature, i.e. there is no delay between reaching a specific temperature and particle failure occurring.
- There is an exponential relationship between failed particle fraction and temperature between 1200 and 1600 °C.

Previous experience from annealing tests on AVR fuel elements at temperatures up to 1 250 °C showed no relationship between failed particle fraction and temperature in the temperature range below 1250 °C.

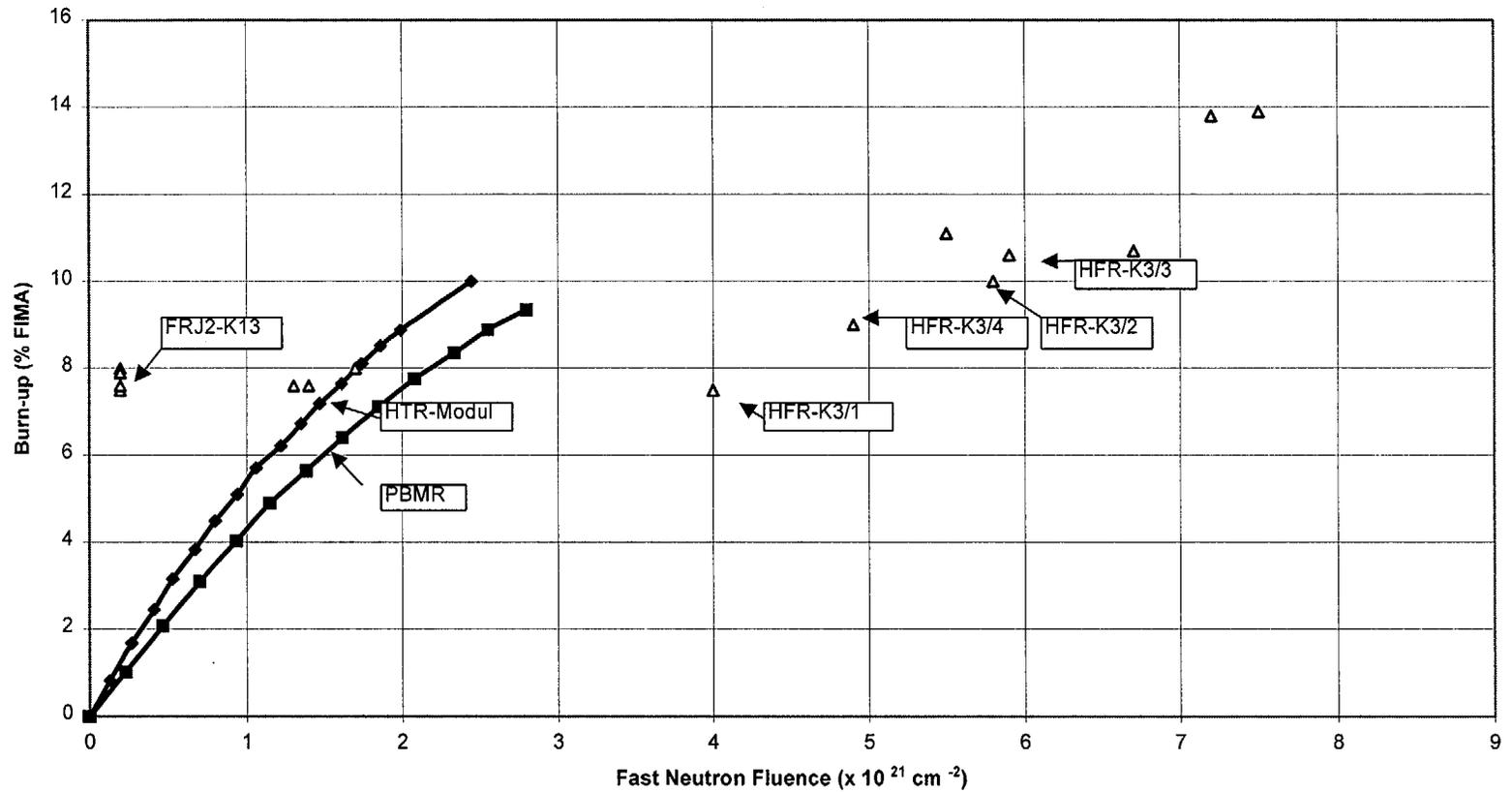


Figure 2: COMPARISON OF PBMR, HTR-MODUL AND PHASE 1 TESTS BURN-UP vs. FAST FLUENCE

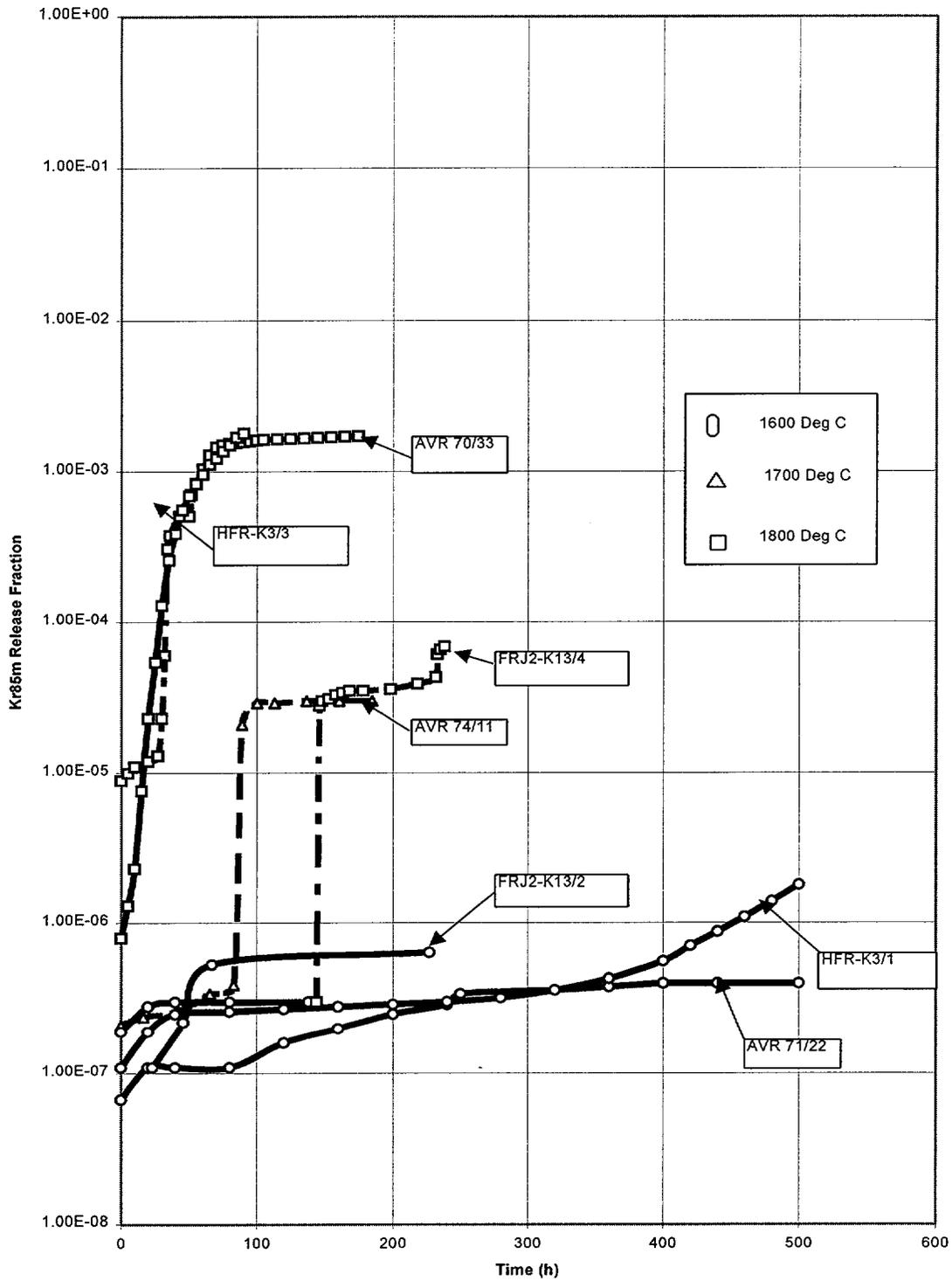
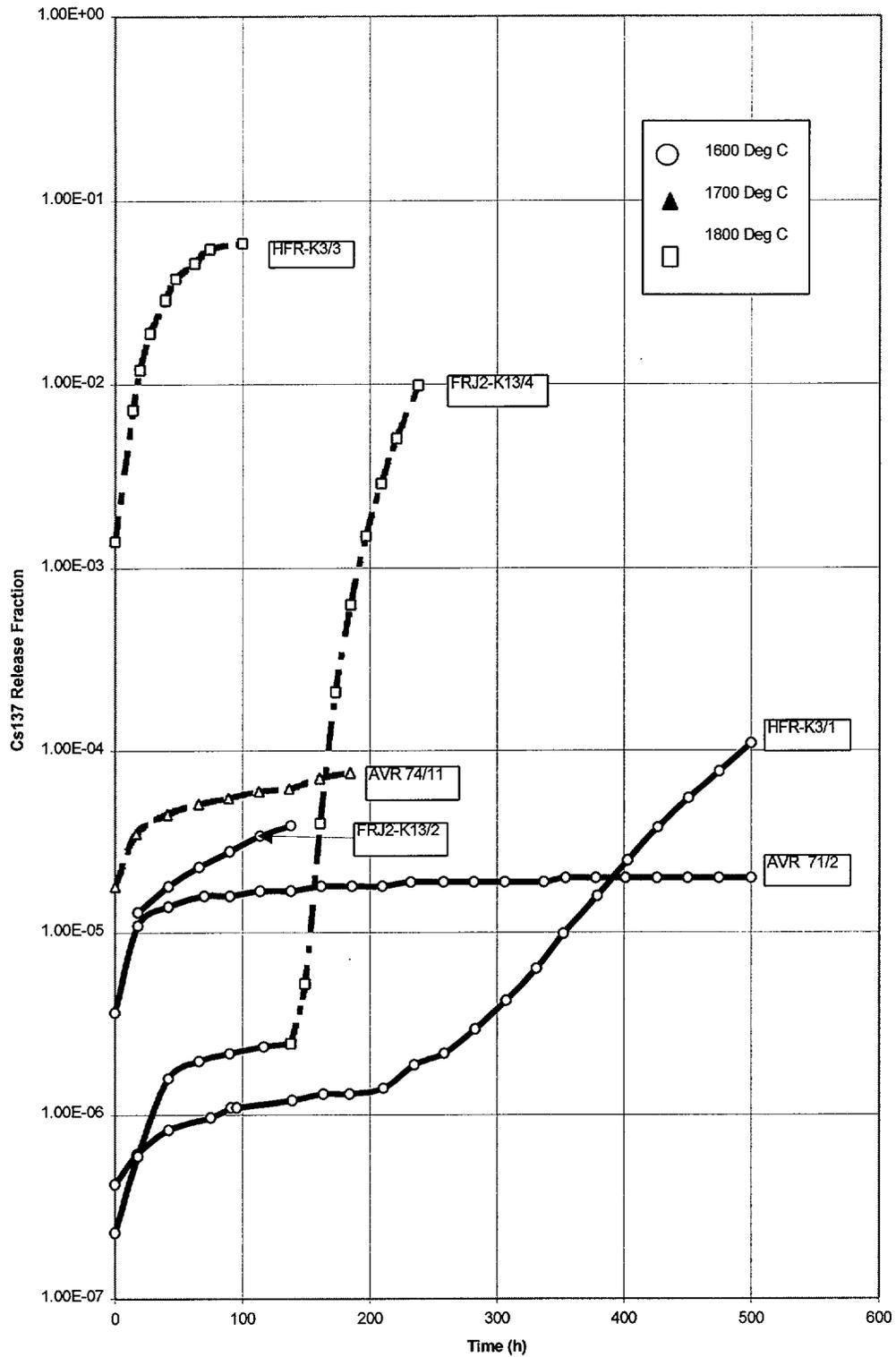


Figure 3: Kr-85 RELEASE FRACTION FOR ANNEALING TESTS ON PHASE 1 AND SOME AVR FUEL ELEMENTS



**Figure 4: Cs-137 RELEASE FRACTION FOR ANNEALING TESTS ON PHASE 1 AND SOME AVR FUEL ELEMENTS**

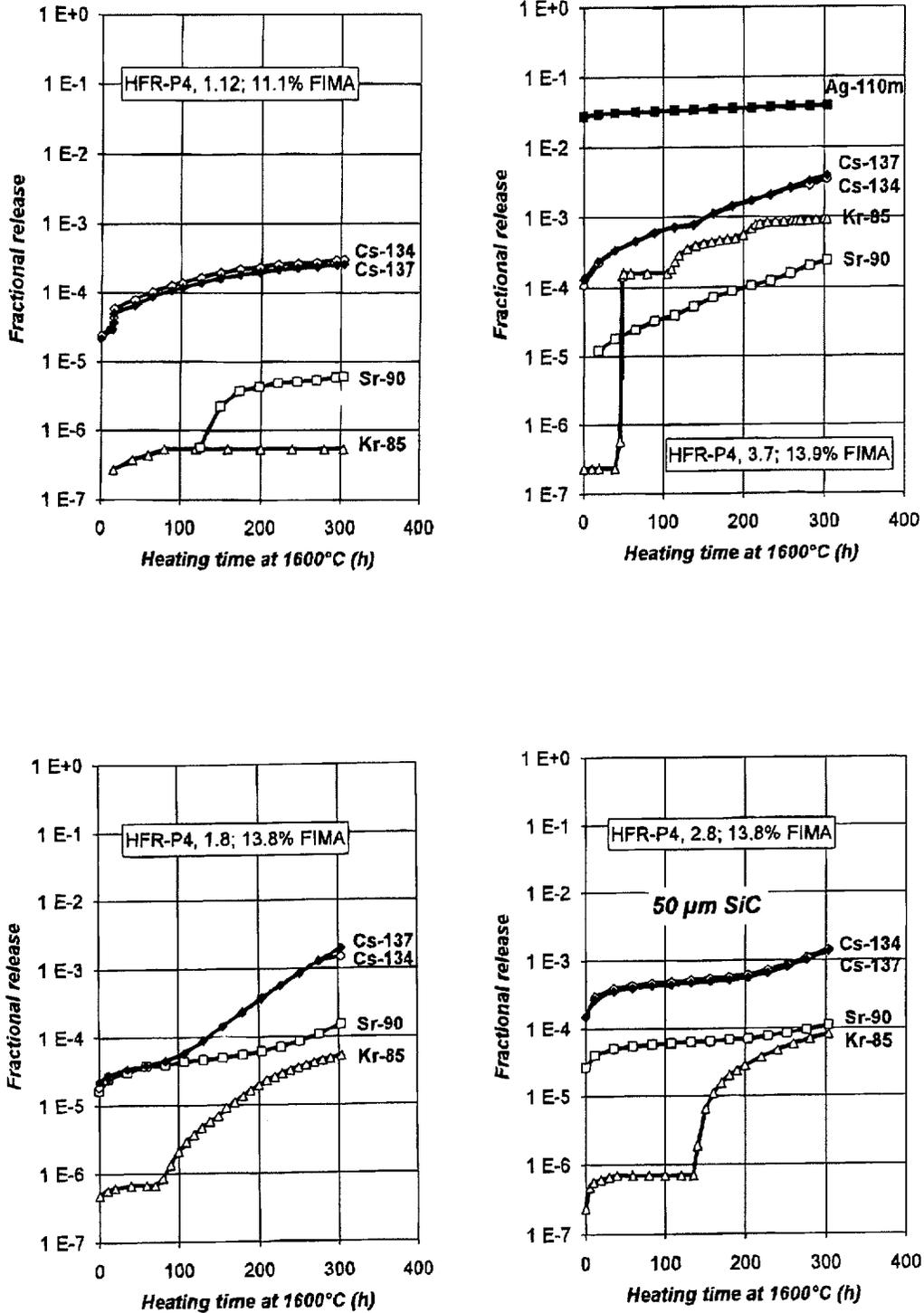


Figure 5: RELEASE FROM COMPACT, (1.12, 3.7, 1.8, 2.8) DURING HEATING TEST AT 1 600 °C

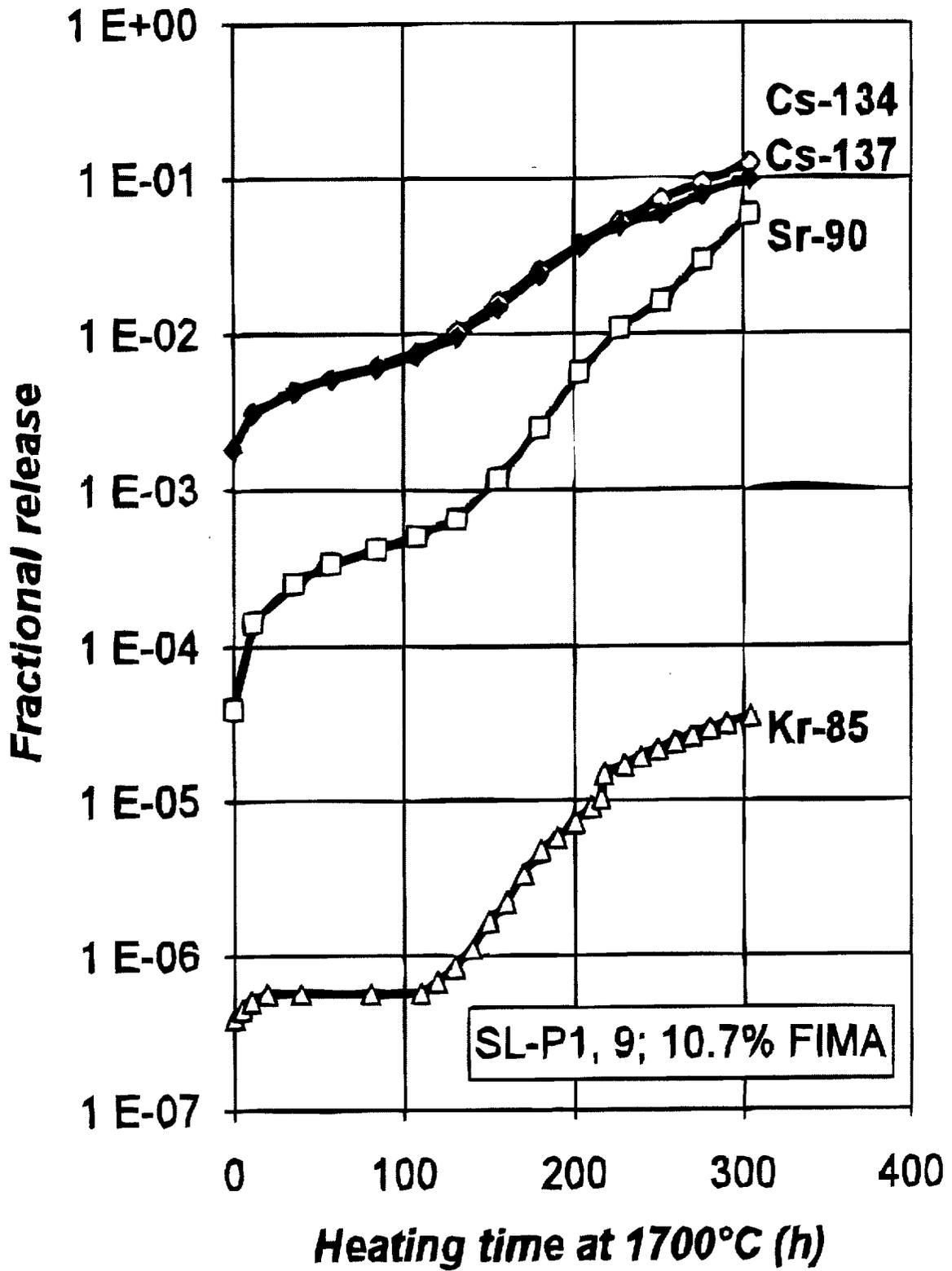


Figure 6: RELEASE FROM COMPACT SL-P1, 9 DURING THE HEATING TEST AT 1 700 °C

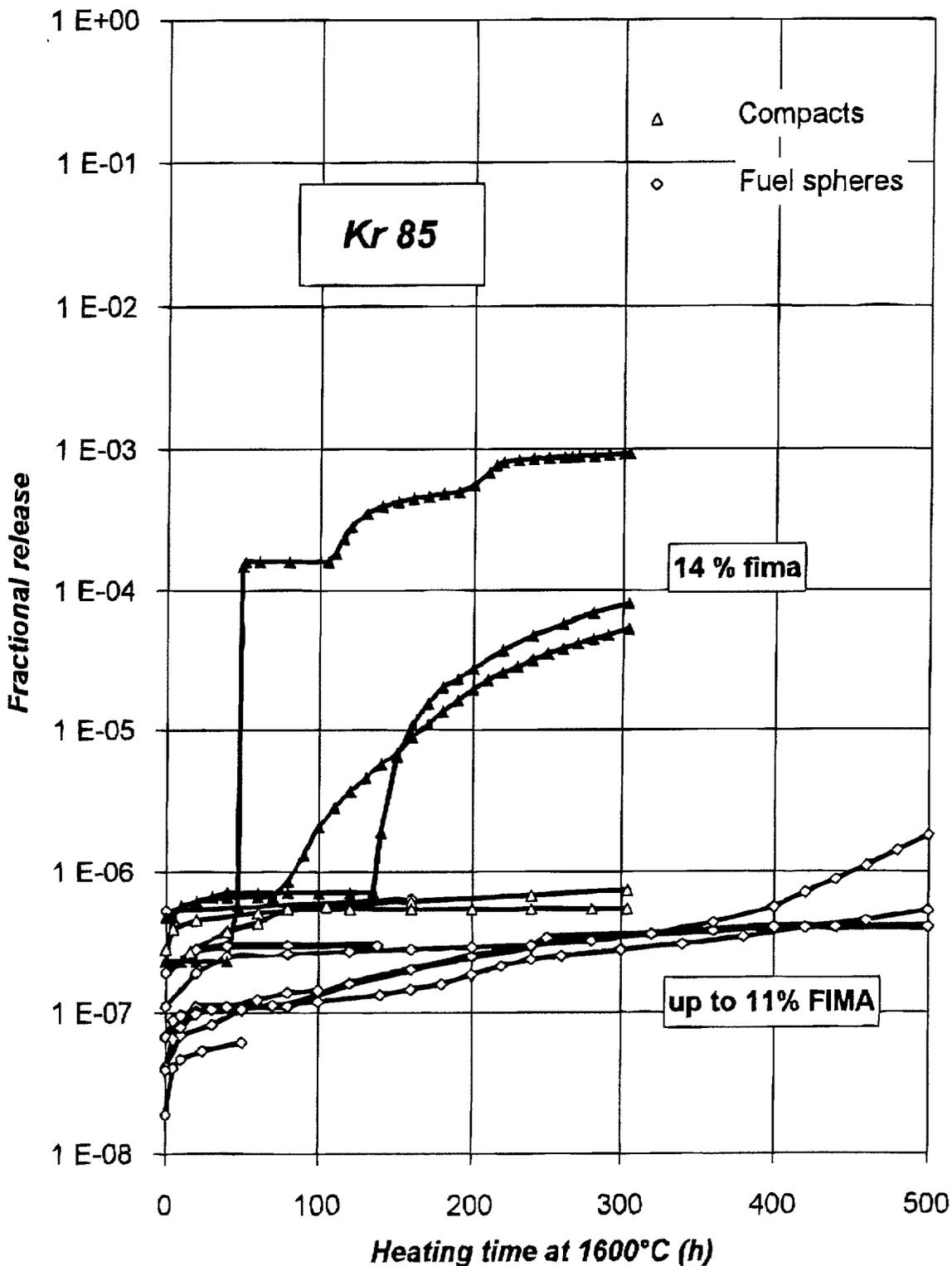


Figure 7:  $^{85}\text{Kr}$  RELEASE AT  $1600^\circ\text{C}$  FROM COMPACTS (10.7 TO 13.9% FIMA) AND FUEL SPHERES (3.5 TO 9% FIMA)

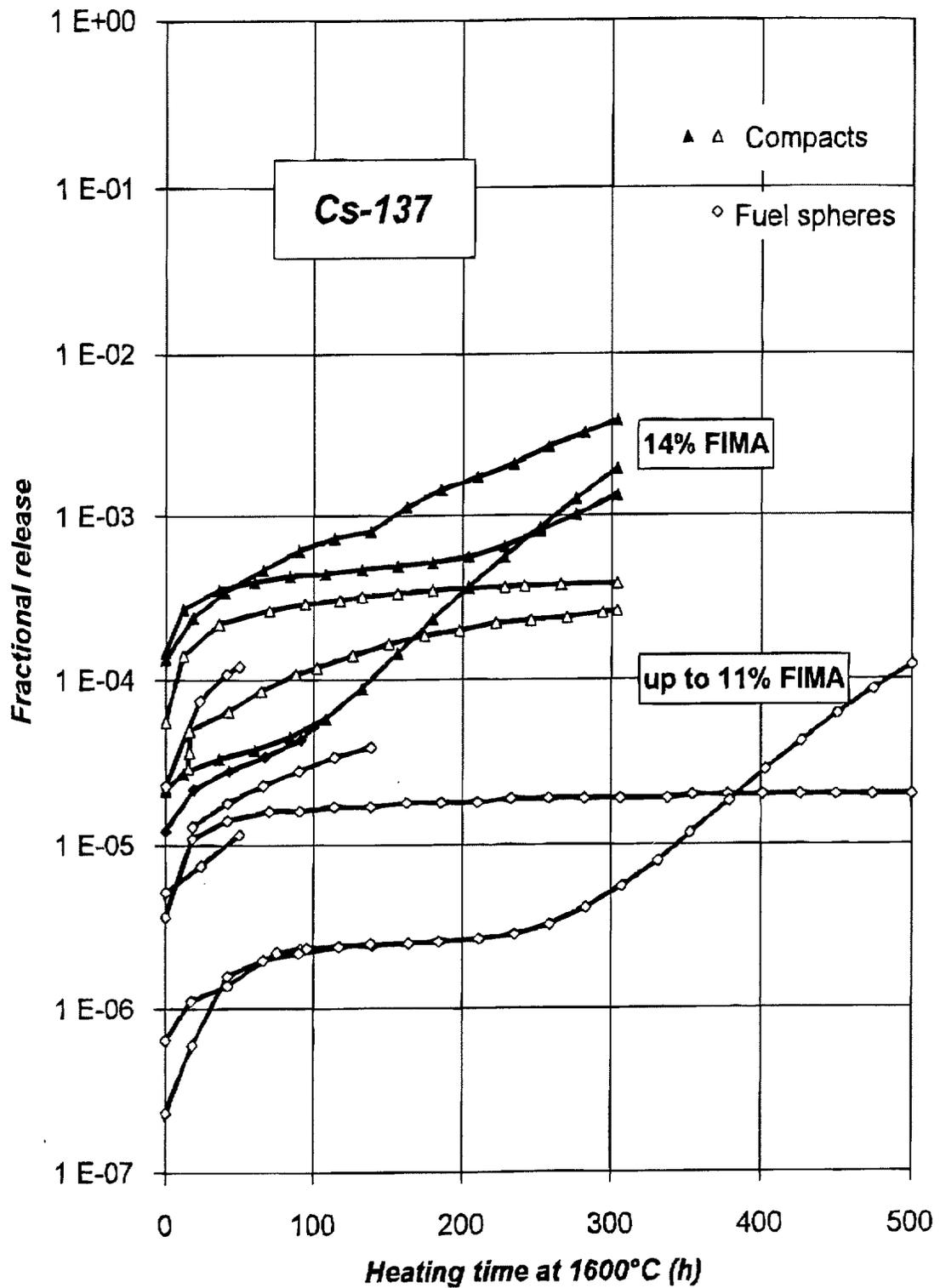


Figure 8: <sup>137</sup>Cs RELEASE AT 1 600 °C FROM COMPACTS (10.7 TO 13.9% FIMA) AND FUEL SPHERES (3.5 TO 9% FIMA)

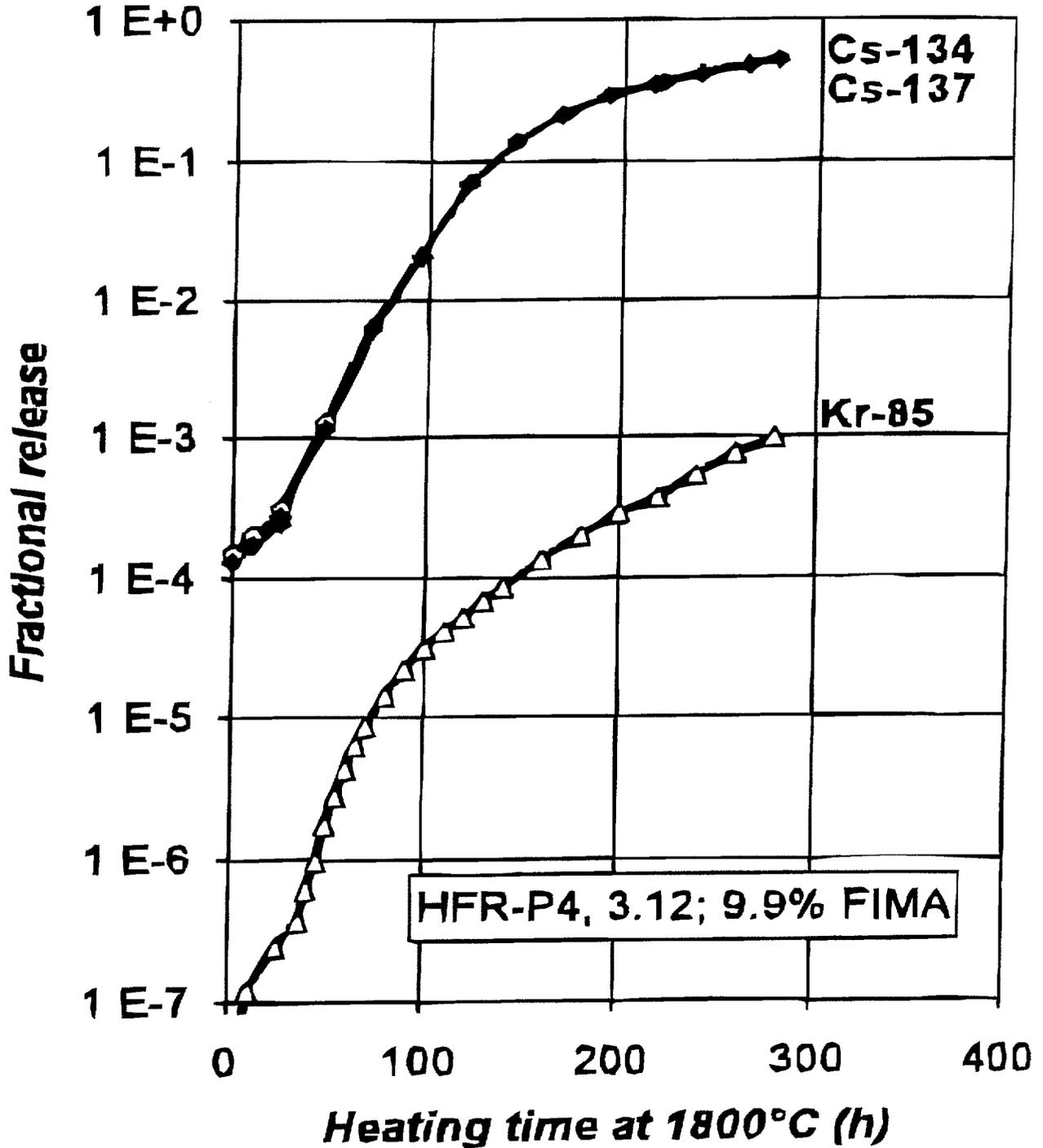


Figure 9: RELEASE FROM COMPACT HFR-P4, 3.12 DURING THE HEATING TEST AT 1 800 °C

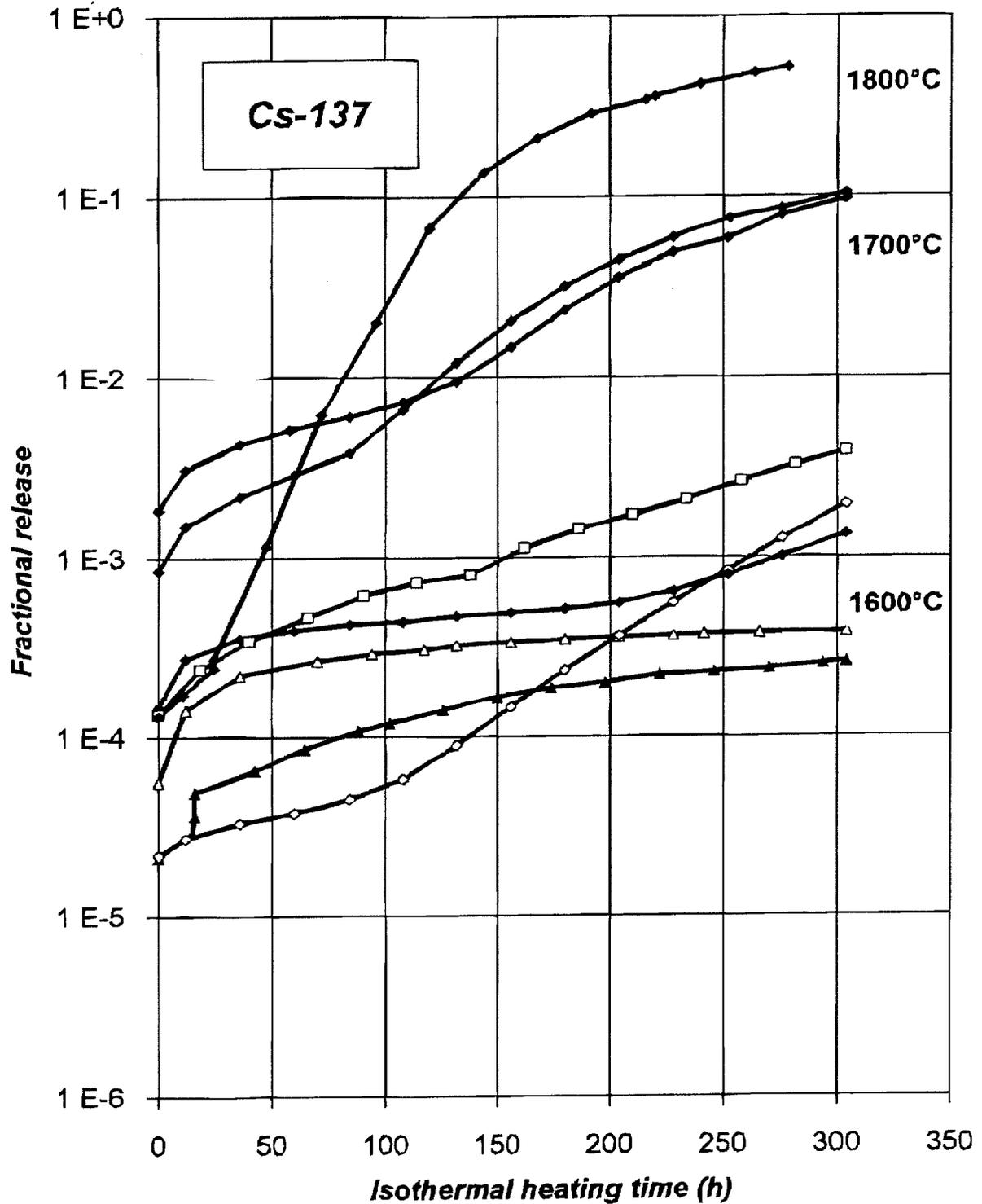


Figure 10: <sup>137</sup>Cs RELEASE FROM COMPACTS WITH BURN -UP OF 10 TO 11% FIMA AT 1 600, 1 700 AND 1 800 °C

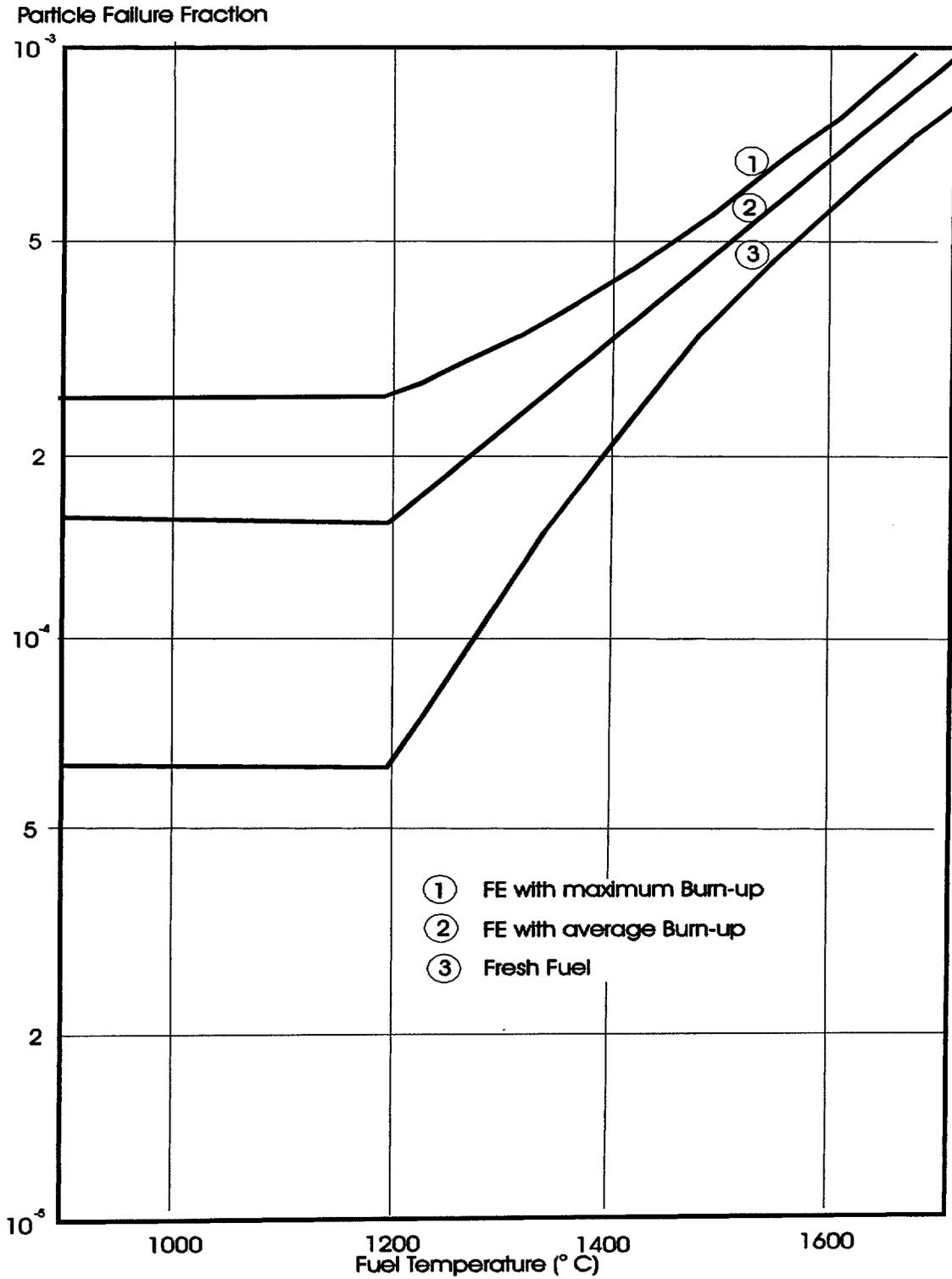


Figure 11: FAILED PARTICLE FRACTION AS FUNCTION OF FUEL TEMPERATURE (DESIGN VALUES)

### 3.13 Operational Experience with LEU -TRISO Fuel

Two reactors using pebble type fuel have been designed and built; both in Germany. The AVR was a 46 MWth reactor intended as a test bed for fuel element development while the THTR-300 was a 756MWth reactor intended as a prototype for a medium sized pebble bed reactor. The fuel elements of the THTR-300 consisted of HTI -BISO type coated particles using (Th,U)O<sub>2</sub> kernels.

The AVR was initially designed to have a coolant outlet temperature of 850 °C, but this was raised to 950 °C during 1974. During its lifetime, a total of 289 789 spherical fuel elements from 26 fuel batches were fed into the AVR core. Of particular interest to the PBMR are reloads 19 (24 615 fuel elements), 21 (20 250 fuel elements), and 21 -2 (8 740 fuel elements). These three reloads consisted of spherical fuel elements containing LTI-TRISO coated particles with UO<sub>2</sub> kernels of 10% and 17% enrichment. Approximately 600 of the fuel elements taken from the AVR core during the reactor lifetime underwent detailed post-irradiation examinations, including heating tests at different temperatures above 1 600 °C.

**Table 10** shows the detail for AVR reloads of fuel that utilized LTI -TRISO coated particles containing UO<sub>2</sub> kernels [7].

**Table 10: AVR RELOADS USING LTI-TRISO COATED PARTICLES WITH UO<sub>2</sub> KERNELS**

Reload Number	Insertion Date	Fuel Element Type	Number Of Fuel Elements	U-235 Enrichment (%)
19	7/82	GLE-3	24 615	9.6
21	2/84	GLE-4	20 250	16.7
21-2	10/87	GLE-4	8 740	16.7

Average burn-up values of 9.6% FIMA and 13.1% FIMA were reached in these tests for 10% enriched GLE-3 fuel and 16.7% enriched GLE-4 fuel elements respectively.

Fuel elements irradiated in the AVR were very similar to reference fuel elements used for irradiation tests of Phase 1. **Table 11** is a comparison between AVR-GLE 3 fuel element characteristics and reference fuel characteristics taken from reference [5].

**Table 11: COMPARISON BETWEEN AVR-GLE 3 FUEL AND REFERENCE FUEL**

Experiment	AVR 19	HFR-K3/FRJ2-K13
Particle batch	HT 232-245	EUO 2308
Kernel composition	UO <sub>2</sub>	UO <sub>2</sub>
Kernel diameter (μm)	500 ± 2%	497 ± 3%
Kernel density (g/cm <sup>3</sup> )	10.80	10.81
<b>Coating Thickness (μm)</b> Buffer layer Inner pyrocarbon layer SiC layer Outer pyrocarbon layer	93 ± 14% 38 ± 10% 35 ± 6% 40 ± 9%	94 ± 11% 41 ± 10% 36 ± 5% 40 ± 6%
<b>Coating Density (g/cm<sup>3</sup>):</b> Buffer layer Inner pyrocarbon layer SiC layer Outer pyrocarbon layer	1.01 1.86 3.19 1.89	1.00 No Value 3.20 1.88
Fuel element type	AVR-GLE 3	HFR-K3/FRJ2-K13
Graphite type	NUKEM A3-27	NUKEM A3-27
<b>Fuel Loading:</b> U-235 (g/FE) Heavy metal (g/FE) U-235 enrichment (%) Particles/FE	1 10 9.82 16 400	1 10 9.2 16 400
<b>Free Uranium Fraction:</b> (Burn-Leach) Particles Fuel element	No value 5 x 10 <sup>-5</sup>	< 5 x 10 <sup>-8</sup> 3.5 x 10 <sup>-5</sup>
Year of manufacture	1981	1981

The biggest problem with measurements in the AVR was that it was difficult to estimate the temperatures seen by the fuel elements that were extracted for further testing. Temperatures were measured by inserting probe fuel elements containing meltwires spanning the temperature range from 655 to 1 280 °C. After the probe fuel elements had passed through the reactor core, they were separated and the maximum temperature determined by noting which meltwires had melted. The temperature distribution for 106 probe fuel elements is shown in **Figure 12 [7]**. Of the 106 probe elements used, all meltwires had melted in 17 of them, and they were added to the 1 300 to 1 350 °C bin in the diagram. The gas outlet temperature for this test was 950 °C. Although the temperature distribution of AVR fuel elements is known, the actual temperatures for individual spheres that passed through the core is not known.

High temperature annealing tests were performed on 18 fuel elements of type GLE -3 that had been irradiated in AVR to a wide range of burn -up values. A comprehensive range of Post-irradiation Evaluations (PIE) was performed on some of these fuel elements. The results of all

these tests can be found in reference [5]. **Table 12** shows some detail of the AVR fuel elements and the PIE performed on them. During Phase 1 of German reference fuel irradiation tests; a number of reference fuel elements were irradiated under strictly controlled irradiation and temperature conditions. These fuel elements are also included in **Table 12**, under their original test identification numbers.

The measured Cs-137 release fractions as a function of annealing time obtained from annealing tests at different temperatures are plotted in **Figure 13**. Similar graphs of measured Sr-90 and Kr-85 release fractions as a function of annealing time at different temperatures are shown in **Figure 14** and **Figure 15**.

It is clear from the graphs that there is no evidence that fuel elements irradiated in the AVR behaved any differently from fuel elements irradiated under controlled temperature conditions in materials testing reactors. Thus, results from Phase 1 tests will be applicable in pebble bed reactors, provided that the irradiation load conditions in the reactor fall within the envelope provided by test results.

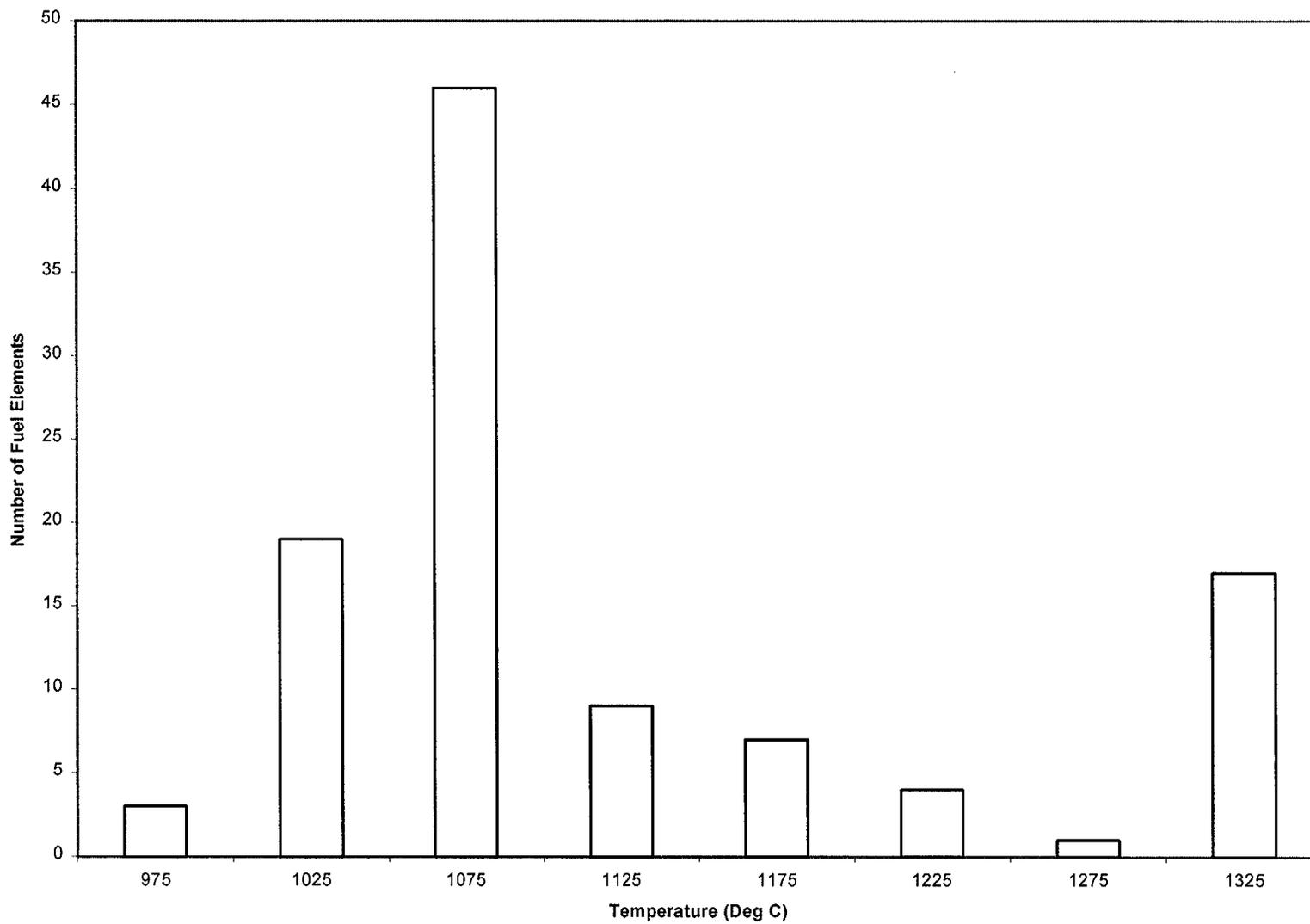
Kr-85 release fractions obtained after annealing of irradiated fuel elements at different temperatures for 100 h are shown in **Figure 16**. Annealing time at temperatures higher than 1 800 °C was only 30 h, and the 30 h data for these temperatures is included in the graph. The data was fitted using an exponential function, which is also shown on the graph. From this graph it is clear that at 1 600 °C, the release of fission products from fuel elements is determined solely by the natural uranium contamination of the graphite matrix, and that there is no leakage of fission products from coated particles. At higher temperatures, the fractional release increases exponentially with temperature.

Table 12: PIE DETAILS OF AVR FUEL ELEMENTS

Element Number	Burn-up (% FIMA)	Annealing Temperature (°C) (K)=KUFA (A)=A-TEST	Annealing Duration (h)	Fission Product Release Fraction	Fission Product						Ceramo-graphy
					Profile in Fuel-free Zone	Concentration in Matrix	Inventory in Particles	Distribution in Coatings (Micro-probe)	Distribu-tion in Particle	Balance	
AVR 71/22	3.5	1 600(K)	500	Y	Y	Y	Y	Y	Y	Y	N
HFR-K3/1	7.7	1 600(K)	500	Y	Y	Y	Y	Y	Y	Y	N
FRJ2-K13/2	8.1	1 600(K)	160	Y	Y	N	Y	Y	Y	Y	N
AVR 82/20	8.6	1 600(K)	100	Y	N	N	N	N	N	N	N
AVR 82/9	8.9	1 600(K)	500	Y	N	N	N	N	N	N	N
AVR 74/11	6.2	1 700(K)	185	Y	Y	Y	Y	Y	N	Y	N
AVR 76/18	7.1	1 800(K)	200	Y	N	Y	N	N	N	Y	Y
FRJ2-K13/4	7.6	1 600(K) 1 800(K)	138 100	Y	Y	Y	N	N	N	Y	Y
HFR-K3/3	10.2	1 800(K)	2 x 100	Y	Y	Y	Y	Y	Y	Y	Y
AVR 74/10	5.5	1 800(A)	3 x 30	Y	N	N	N	N	N	N	N
AVR 71/33	1.6	1 800(A)	3 x	Y	Y	N	N	N	N	N	Y
AVR 73/12	3.1	1 900(A)	2 x 50	Y	N	N	N	N	N	N	N
AVR 76/19	7.3	1 900(A)	30	Y	N	N	N	N	N	N	Y
AVR 80/22	9.1	1 900(A)	30	Y	N	N	N	N	N	N	Y
AVR 71/7	1.8	2 000(A)	2 x 50	Y	N	N	N	N	N	N	Y
AVR 80/16	7.8	2 000(A)	30	Y	N	N	N	N	N	N	Y
AVR 74/6	5.6	2 100(A)	30	Y	N	N	N	N	N	N	Y

PBMR Nuclear Fuel  
Non-Proprietary

Element Number	Burn-up (% FIMA)	Annealing Temperature (°C) (K)=KUFA (A)=A-TEST	Annealing Duration (h)	Fission Product Release Fraction	Fission Product						Ceramo-graphy
					Profile in Fuel-free Zone	Concentration in Matrix	Inventory in Particles	Distribution in Coatings (Micro-probe)	Distribution in Particle	Balance	
AVR 76/28	6.9	2 100(A)	30	Y	N	N	N	N	N	N	N
AVR 76/27	7.4	2 100(A)	30	Y	N	N	N	N	N	N	Y
AVR 70/19	2.2	2 400(A)	-	Y	N	N	N	N	N	N	N
AVR 74/8	2.9	2 500(A)	-	Y	N	N	N	N	N	N	Y
AVR 80/14	8.4	2 500(A)	-	Y	N	N	N	N	N	N	Y



**Figure 12: TEMPERATURE DISTRIBUTION FOR AVR FUEL ELEMENTS**

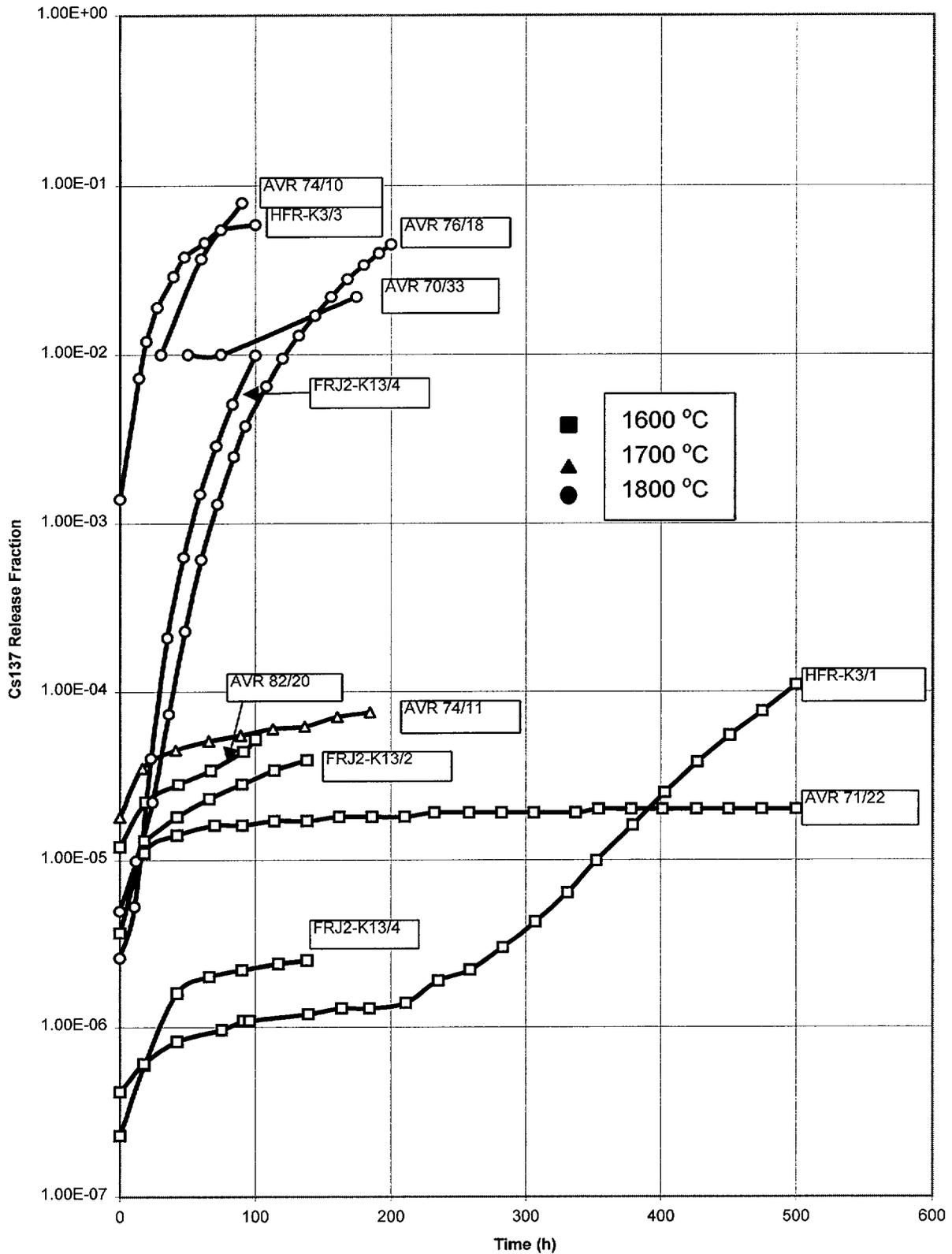


Figure 13: Cs-137 RELEASE FRACTIONS AT 1 600, 1 700 AND 1 800 ° C

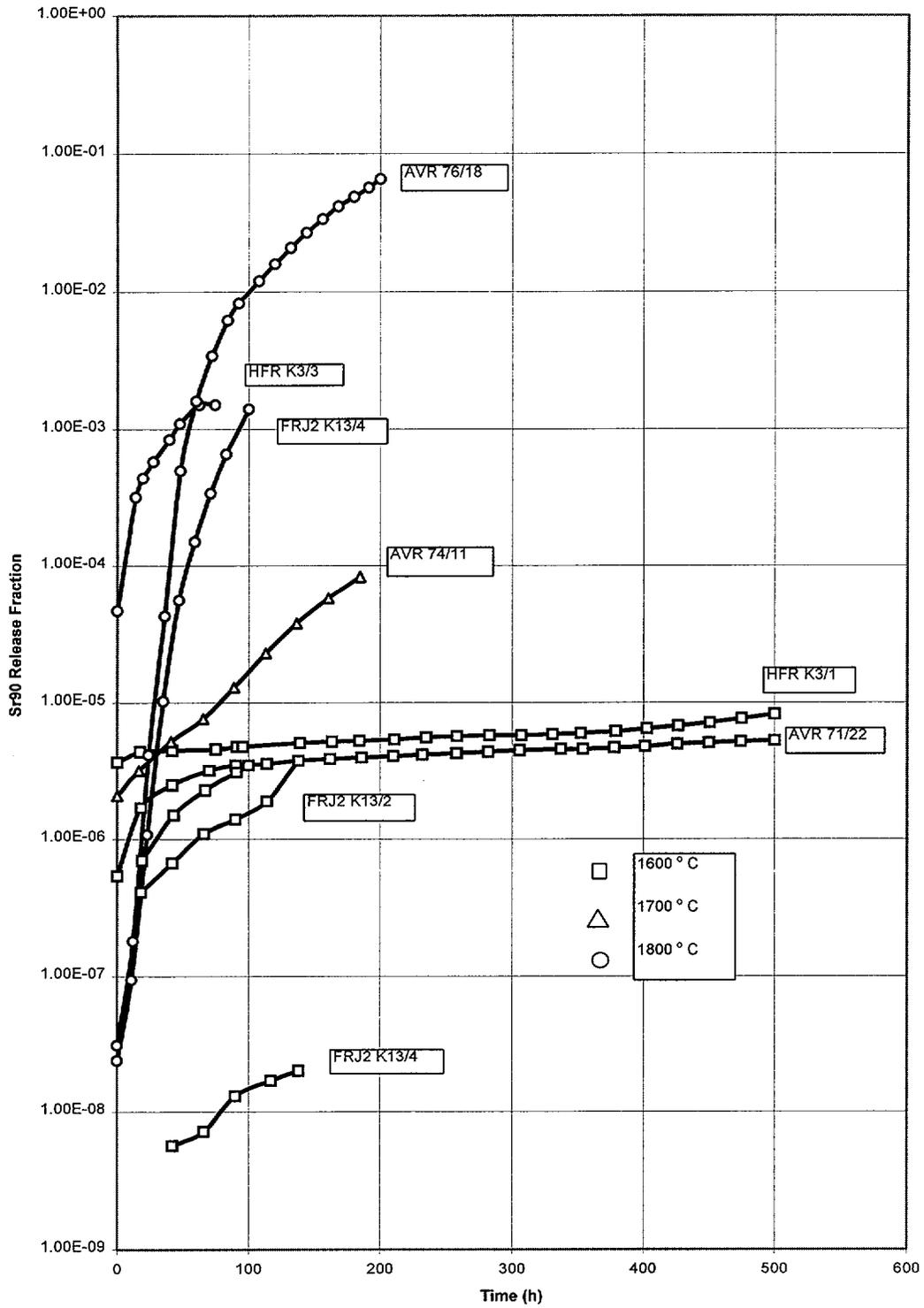


Figure 14: Sr-90 RELEASE FRACTIONS AT 1 600, 1 700 AND 1 800 °C

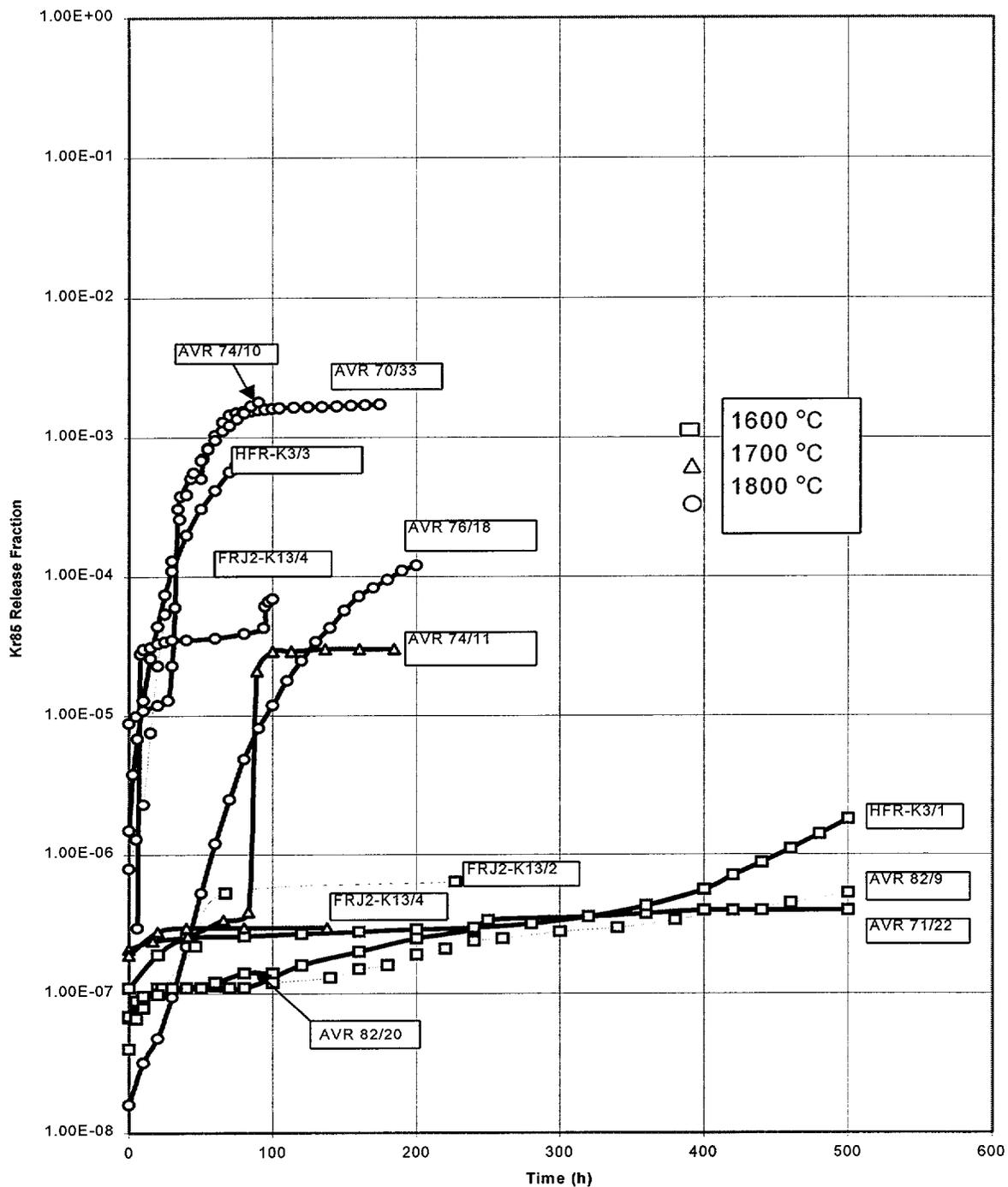


Figure 15: Kr-85 RELEASE FRACTION AT 1 600, 1 700 AND 1 800 ° C

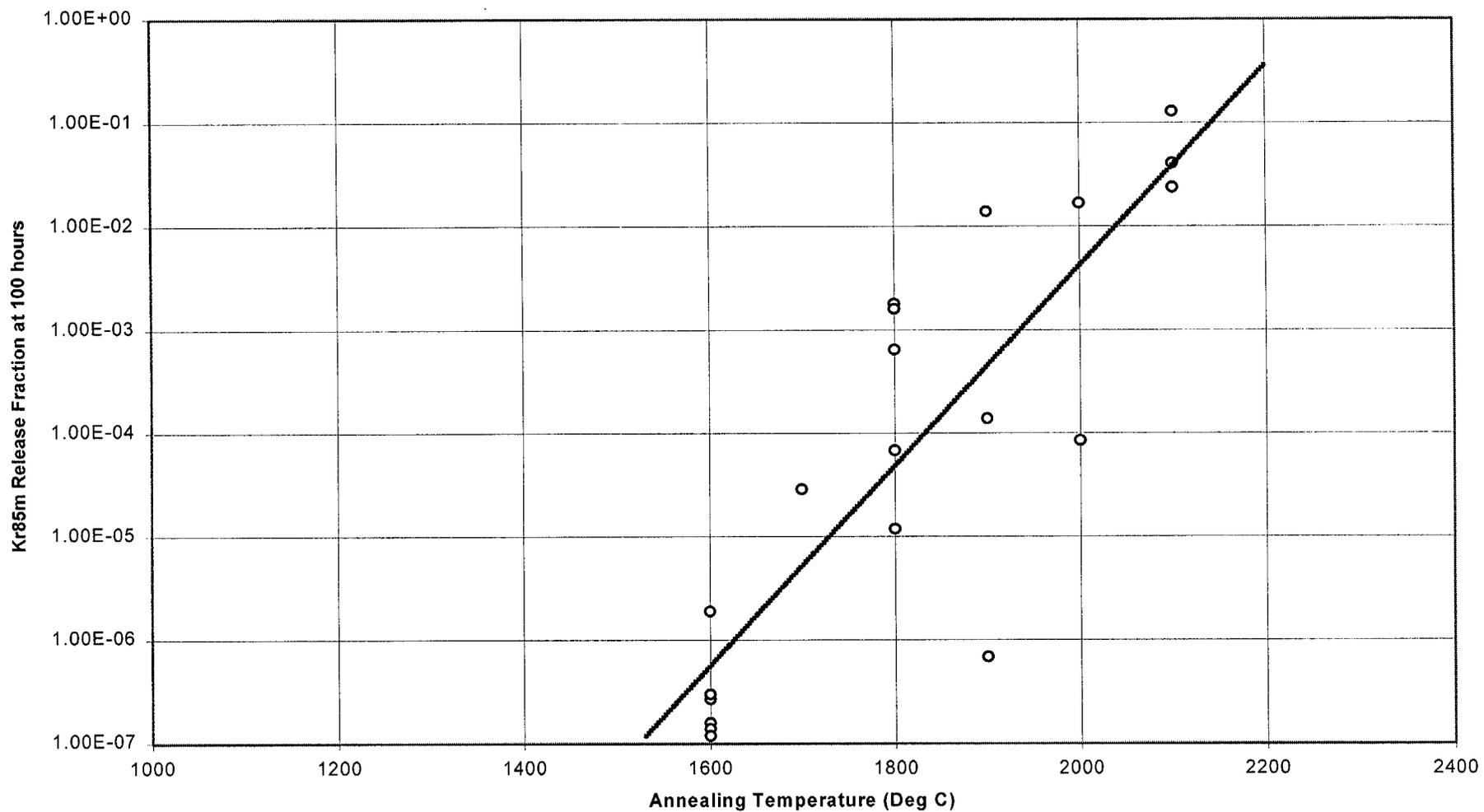


Figure 16: Kr-85 RELEASE FRACTION AS FUNCTION OF ANNEALING TEMPERATURE FOR PHASE 1 AND AVR FUEL ELEMENTS

## 4. SUMMARY

### 4.1 Manufacturing Process

1. The manufacturing process is well known and proven to produce high quality fuel with failed particle fractions less than the conservative PBMR specification of  $6 \times 10^{-5}$  (~1 particle per sphere). The last LEU-TRISO fuel spheres manufactured by NUKEM were the AVR 21-2 reload and the Proof Test elements. These exhibited free uranium values, as measured by burn-leach tests of 50 fuel spheres, of approximately 11 parts per million [13].
2. PBMR technology is based on technology developed by NUKEM who made over a quarter of a million spheres for the AVR and approximately 1 million fuel spheres for the THTR.

### 4.2 TRISO Fuel Design

1. The TRISO fuel design is the result of 30 years of developing, testing and in reactor operation of spherical particles with almost 54,000 spheres, (680 million particles) produced and inserted into the AVR reactor (1981-1988).
2. As a result of the Phase I irradiation tests 211,834 fuel particles were irradiated with no failures covering a range from 7.5 to 14.7% FIMA (burn-up from 67,500 to 132,300 MWD/MTU), a range of 0.2 to  $8 \times 10^{21}$  n/cm<sup>2</sup> for fast neutron fluence, and a temperature range of 800 to 1,320 °C. The PBMR target burnup is 80,000 MWD/MTU
3. The expected value for the irradiation induced failed particle fraction calculated with a 95% confidence level is  $2 \times 10^{-5}$ .
4. No particle failed during annealing of fuel spheres FRJ2 -K13/2, FRJ2-K13/4, HFR-K3/1, and AVR 71/22 containing 65,600 particles at 1,600 °C confirming the 1,600 °C maximum temperature for PBMR during abnormal conditions.
5. The expected value for temperature -induced failed particle fraction is  $5 \times 10^{-5}$  with 95% confidence level, taking into account the number of coated particles contained in the fuel elements (65,600) [refer to point 4 above].

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