EUROPEAN COMMISSION
Community Research

RAPHAEL

Contract Number: 516508 (FI6O)

DELIVERABLE (D-FT4.4)
BN past experience
Synthesis

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Reporting period: 15/04/2005– 14/04/2007
Date of issue of this report: April 2007

Start date of project : **15/04/2005** Duration : **48 Months**

Project co-funded by the European Commission under the Euratom Research and Training Programme on Nuclear Energy within the Sixth Framework Programme (2002-2006)		
Dissemination Level		
PU	Public	
RE	Restricted to the partners of the RAPHAEL project	
CO	Confidential, only for specific distribution list defined on this document	**X**

RAPHAEL ***(ReActor for Process heat, Hydrogen And ELectricity generation)***

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ReActor for Process heat, Hydrogen And ELectricity generation (RAPHAEL)		
Sub-project: SP-FT **Work package:** 4	**RAPHAEL document no:** RAPHAEL-0407-D-FT4-4	**Document type:** Deliverable
Issued by: BN **Internal no.:** 0700181/220		**Document status:** final

Document title

BN PAST EXPERIENCE – FUEL AND IRRADIATION DESCRIPTION, PIE RESULTS

Executive summary

In the first report supplied by BN in the framework of the RAPHAEL-IP project (ref. D FT4-3) several HTR experiments were presented in detail.
This second report makes the synthesis of the past BELGONUCLEAIRE's knowledge and experience on the HTR fuel fabrication and irradiation performance.

Revisions

Rev.	Date	Short description	Author	WP Leader	SP Leader	Coordinator
00		First issue	G. TOURY BN	P. OBRY CEA	V. BASINI CEA	E. BOGUSH AREVA NP

	Type of document:	Reference No / File - "Rev." - Page
		0700181/220-"-" - 1
	Technical Note	Other reference:

Client: EC	Author(s) G. TOURY
Project: Raphael IP – SP Fuel Technology WP-FT4 Performance Modelling	

Subject:

HTR Fuel Irradiation Experiments
BELGONUCLEAIRE past experience

SYNTHESIS

C															
B															
A															
-	17/01/2007	0700181	FIN	GTY	GTY		ssh		SSH				GTY		77
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					Author		Verified by		Approved by		Q.A.		Issued by		

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LIST of MODIFICATIONS		
DATE	**REVISION**	**OBJECT**
17/01/2006	-	FINAL

NOTICE

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This work is performed under the term and conditions of the Consortium Agreement of the RAPHAEL Project supported by the EC under contract n° 516508 (FI6O)

LIABILITY

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1. INTRODUCTION

BELGONUCLEAIRE (BN) and the Belgian nuclear research center SCK•CEN have been co-operating on coated particle development since 1959, when they started investigating the powder agglomeration technique for microspheres fabrication.

The research and development in the fields of the HTR fuel design and technology were carried out with significant collaboration with the DRAGON Project and Euratom (Ispra, Petten and Karlsruhe)

The program was first focused on the thorium cycle and the plutonium utilisation until 1967, when it was reoriented towards the low enriched cycle, this latter being preferred by most of the prospective customers and by influent organisations active in Th fuel cycle. The commercial successes of GGA (Gulf General Atomics) during 1971 and the order of the THTR pebble bed prototype brought back the thorium cycle into fashion.

The fuel manufacturing laboratory has started operation in 1964: all the steps of the fabrication process for thorium, uranium and plutonium coated particles (including C-diluted) were established.

In the same time, an important programme of irradiation experiments has started in several irradiation devices to study the in-reactor performances of different types of fuel coated particles and fuel elements.

Despite the successful continued operation of the existing experimental HTR's (Dragon, AVR and Peach Bottom) which demonstrated the excellent characteristics of the fuel elements and graphite core structures both in respect to fission products retention and in maintenance of integrity in service, the HTR development has been significantly slowed down in 1975, with in particular the shut-down of the Dragon reactor and the termination of the Dragon Project in 1976.

In the first report supplied by BN in the framework of the RAPHAEL-IP project (ref. D-FT4-3), several HTR experiments were presented in detail.

This second report makes the synthesis of the past BELGONUCLEAIRE's knowledge and experience on the HTR fuel fabrication and irradiation performance.

2. FABRICATION OVERVIEW

2.1. Kernels

Two main sources of kernels were utilised by the Belgian industry: kernel prepared by a chemical process (mainly produced by Agip Nucleare) and kernels manufactured following a powder metallurgy route by the Belgian industry. Some characteristics of both materials are compared in Table 1.

Manufacturing method	Powder agglomeration	Gel precipitation	
Density range covered (%TD)	85 – 97	80 - 99	
Reproducibility of average density (%TD)	±2	low density ±3	high density ±0.5
Density variation from kernel to kernel	expected to be important	good	good
Reproducibility of average diameter (µm)	±20	±23	±5
Diameter standard deviation (µm)	±25	21	18
O/U ratio	slightly sub-stoechiometric (1.999)	2.003	2.006
		always over stoechiometric	
Sphericity D_{max}/D_{min} (average)	1.033	1.032	
Standard deviation sphericity	0.024	0.024	
Typical sphericity defects	irregular shape, flats	ovality and egg or drop form	
Surface conditions	orange peel surface, macroporosity; low abrasion resistance	glossy, high abrasion resistance	
Porosity distribution	macroporosity homogeneously distributed to concentrated in the centre	microporosity, very dense surface layer (30 to 60 µm), low density core	
Coating behaviour			
Exploded kernels (%)	0.05 to 1	0.05 to 4 depending of the Cl and H_2O content and on density	
Dimensional stability during coating	Shrinkage 1 to 10 µm	Shrinkage 1 to 25 µm depending on thermal treatment	

Table 1: Typical kernel characteristics
(UO_2 800µm)

Although the potential qualities favoured the gel precipitated kernels, some minor disadvantages were observed:

- the O/U ratio was higher, causing a more important internal CO pressure;
- the different porosity distribution was observed: a dense layer surrounding a porous core could impair the swelling behaviour and fission gas release;
- the difficulty to avoid the presence of proportion of droplet shaped individuals.

2.2. Coating

Tables 2 and 3 indicate respectively the coating conditions and target product specifications that were applied.

Layer	Temperature (°C)	Deposition rate (µm/min)	Gas
Porous PyC	1450	15	C_2H_2 – CO
Transition PyC	1500 – 1700	1.5	CH_4 – CO
Inner HDI PyC	1800 – 1950	1.3	CH_4 – H_2 - Ar
SiC	1550	0.5	H_2 – Silane – Ar
Outer HDI PyC	1800 - 1950	1	CH_4 – Ar –H_2

Table 2: Standard Coating conditions

Layer	Thickness (µm)	Density (g/cm³)	B.A.F.*	Cryst. Size (Å)
Porous PyC	30^{+5}_{-0}	$1.0^{+0.1}_{-0.0}$	-	-
Transition PyC	30 ± 5	$1.6^{+0.1}_{-0.0}$	-	-
Inner HDI PyC	30 ± 5	1.8 ± 0.05	1.00 – 1.08	70 – 140
SiC	35 ± 5	$3.2^{+0.02}_{-0.02}$	-	-
Outer HDI PyC	30 ± 5	1.8 ± 0.05	1.00 - 1.08	70 - 140

* B.A.F: Bacon Anisotropy Factor

Table 3: Typical coating specifications

Since 1975 and the results of the BR2/VC5 experiment, methane was replaced by propylene as carbon source for the PyC layer deposition.

With uranium dioxide fuel, it was necessary to place a PyC transition layer between the porous buffer and the high-density inner PyC to avoid uranium carbide formation during coating operations.

The SiC layer was sometimes surrounded by two flash layers (of a few µm) of porous PyC to compensate differential thermal expansion and prevent cracks propagation during the fabrication and the initial stage of irradiation.

The fluidisation of the kernels was similar for the routes after deposition of the first microns of the coating layers. Nucleation seemed to be slightly more difficult on the surface of the liquid route kernels, resulting in a smaller standard deviation for the inner PyC (7 compared to 12 µm). For some inadequately thermally treated batches, the density of liquid route kernels was insufficiently stabilised and severe shrinkage occurred when deposing PyC at high temperature (up to 25 µm). Even if this effect itself is not harmful for the as-fabricated quality of the products, it could favour the amoeba effect during irradiation under extreme conditions.

2.3. Particle contamination

Both the alpha counting technique and the chemical leaching method were used to evaluate particle contamination. The resulting fraction of visible core (FVC) as calculated by alpha-counting is presented in Table 4. Even if the contamination level was similar on both types of kernels, the source was different: while for the powder metallurgy kernels the contamination was mainly due to abrasion of the kernel surface at the early stage of coating, the contamination of the liquid route particles resulted essentially from kernel explosion after or during deposition of the sealing layer.

Systematic control of the contamination on various fabrication campaigns allowed to draw the following conclusions:

- there were no indications that the contamination was influenced by the number of coating runs;
- the average value on both kernels types after coating is around $6x10^{-7}$ FVC and ranges from 10^{-6} down 10^{-8} FVC ;
- a heat treatment at 2100°C during 1 h has caused a lowering of the contamination by a factor two.

The low contamination level experienced was on the one hand due to the deposition of SiC at which stage the HCl formed reacted with UO_2 and volatised the uranium and, on the other hand, to a thorough cleaning of the furnace after each run.

Concerning the losses during coating, it has been demonstrated that an increase of the chlorine content of the liquid route kernels from 10 ppm to 30 ppm cause an increase in uranium losses (or particle explosion) from 0.05% to 4%. The effect was possibly associated with a simultaneous high H_2O content but the correlation has not been established. As a comparison, the figures range between 0.05 to 1% for the powder agglomeration kernels.

	Kernel process fabrication			
	Liquid route		Powder agglomeration	
	Mean FVC*	Range	Mean FVC	Range
As-coated	5.0×10^{-7}	$6.9 \times 10^{-8} - 2.0 \times 10^{-6}$	6.9×10^{-7}	$1.6 \times 10^{-8} - 3.8 \times 10^{-6}$
Annealed 2100°C	3.5×10^{-7}	$1.1 \times 10^{-8} - 5.6 \times 10^{-7}$	3.5×10^{-7}	$1.1 \times 10^{-8} - 5.6 \times 10^{-7}$

* FVC: Fraction Core Visible

Table 4: Contamination values of coated particles (measured by alpha-counting)

2.4. Fuel body manufacturing techniques

In design calling for heavy metal loadings higher than 1.0 g U/cm3 and /or fractions of broken particles lower than 10^{-4} and/or reduced and better tolerance gaps between fuel and graphite sleeves, different techniques were applied to manufacture fuel bodies.

For the standard compaction method, the coated particles were overcoated by a powder agglomeration technique with a resin impregnated graphite powder. The nominal weight of the overcoating to be laid down on the particle was defined by the heavy metal loading of the compact after manufacturing. Overcoated particles were then loaded in an annular die and hot compacted following a sequence defined by the physical characteristics of the resin (melting point and polymerisation / pressure / time relation). Compaction could be performed by an hydraulic system in which case the matrix density and the diameter were very reproducible, or the material could be compacted until a defined height was reached. In the latter case, the matrix density depended of the amount of coated particles and graphite present; the diametrical tolerance was thus enlarged. The operation was then followed by a carbonisation and out-gasing treatment.

To reduce the mechanical stress supported by the coated particles during compaction, two other techniques were developed: the consolidation and in-situ compaction. For theses two techniques, concessions had to be made to the matrix density, the graphite/carbon ratio in the matrix and the control procedures.

For the consolidation, partially resin and graphite overcoated particles were forced by means of mechanical vibrations or centrifugal force into a slurry of graphite and resin diluted spirits. Spacing of the particles was assured by the overcoating and the matrix was consolidated through the polymerisation of the resin. Overall graphite content of the matrix was 85% for 15% carbon, the overall matrix density was 1.2 g/cm^3. The contamination of the finished body was not higher than the coated particles contamination, i.e. about 10^{-6} FCV. The body could be fabricated directly into the graphite sleeve, the gaps were thus strongly reduced.

For the in-situ consolidation, the technique was very similar to the standard compaction method, but the die was replaced by the graphite sleeve itself, thus compensating automatically for dimensional tolerance from graphite machining. This allowed to enlarge such tolerances and reduce the cost of sleeves. Because of the limited resistance of the graphite sleeve, the forming pressure was reduced to 10 kg/cm^2, the amount of resin in the graphite was increased of 35% and an artificial gap had to be created between the inner sleeve and the fuel body. Generally speaking, this technique depended entirely of the utilisation of adequate raw materials.

The characteristics of the three manufacturing techniques are compared in Table 5.

Manufacturing process		Standard compaction	Consolidation	In-situ consolidation
CP volume loading (max)	%	35	50	50
HML[1] (max)	g U/cm^3	1.0 ±0.05	1.6	1.6
Matrix density	g/cm^3	1.6 – 1.7	1.0 – 1.2	1.3 – 1.4
Graphite content of the matrix	%	95	85	92
Fraction of broken particles	-	1×10^{-4}	5×10^{-6}	1×10^{-5}
Nominal gap[2]				
Fuel/outer sleeve	µm	400	100	500
Fuel/inner sleeve	µm	350	10	40
Tolerance on gap				
Fuel/outer sleeve	µm	200	40	100
Fuel/inner sleeve	µm	150	negl.	30
Thermal conductivity	W/cm.°C	0.2	0.06	0.16

[1] Heavy Metal Loading with UO_2 kernels of 800 µm diameter and 90%TD and a 170µm coating

[2] diametrical gap for a compact with 40-60 mm outer diameter and 32-40 inner diameter

Table 5: Characteristics of fuel bodies manufactured according to different processes

3. IRRADIATION EXPERIMENTS

BELGONUCLEAIRE has participated in a few irradiation experiments of HTR particles that were performed in the following reactors:

- BR2 and BR3 located in the Belgian research center in Mol;
- High Flux Reactor (HFR) located in the NRG center in Petten (Netherlands);
- DRAGON reactor located in Winfrith (UK);
- ISPRA reactor located in the EURATOM Joint Research Center of ISPRA (Italia).

The main purpose and characteristics of the experiments are gathered in Table 6.

The main irradiation results for each experiment are described individually hereafter.

U LOOSE PARTICLES							
Irradiation purpose		**Reactor/ Experiment**	**Enrich^{nt} (%U5)**	**Tmax (°C)**	**%FIMA**	**Fast Dose (10^{21} DNE)**
Thermal effect	Thermal shock and amoeba effect under different temperatures and gradients	BR2/ HR-8	Unat	1700	0	0
		BR2/ HR-21	Unat+C	2400	0	0
		BR2/ HR-22	Unat	1550	0	0
		BR2/ HR-29	Unat	2250	0	0
	Low temperature	BR2/ MFBS-6	0.4 - 0.7	700	2.2	1.3
	Intermediate temperature	BR2/ VC 3	0.4 - 9	1100	16	5.3
Irradiation behaviour	Low temperature	HFR/ E-96-2	0.4 - 9	900	8	3
	Coatings of PyC derived from propylene instead of methane	BR2/ VC 5	3 - 5	1400	10	4
	Mean and high temperature	DRAGON/ MET V	3 - 8 - 9	1550	1.35 - 3.0 - 10 - 35	0.36 - 1.08
	Behaviour with and without external porous layer	DRAGON/ Misc Met	12	1250	2	0.5

Table 6: Overview of BN experience in the HTR fuel performance

U COMPACTS						
Irradiation purpose		**Reactor/ Experiment**	**Enrich^{nt} (%U5)**	**Tmax (°C)**	**%FIMA**	**Fast Dose (10^{21} DNE)**
Irradiation behaviour	High temperature	BR2/ VC 1	9	1350	6	1.4
		BR2/ VC 2	4.7 - 9	1350	10	1.4
	Intermediate temperature	BR2/ VC 3	0.4 - 9	1100	16	5.3
Dimensional change	Reference matrix	BR2/ MFBS-6	12	700	2.2	1.3
	Behaviour under irradiation of matrix materials with and without particles	BR3/ 2bis	8 - 12	1200	6	1.7
	Dimensional change of compacts	DRAGON/ DRS 2-5	6	1300	2	5
	Behaviour of annular and cylindrical compacts	DRAGON/ MET-LET	3 - 6	1250	2.5 to 4	0.7 to 3
	Gilsocarbon compact properties	HFR/ Compact 1	dummy CP's	1450	-	5
	Natural graphite (with different polymerisation degree) compact properties	HFR/ Compact 2	dummy CP's	1200	-	4
Mechanical data	Determination of the creep tensile curve of different compacts	HFR/ Creep (E98-3)	dummy CP's	900	-	2.3
	Determination of creep coefficients	HFR/ Creep (E99-3)	dummy CP's	1200	-	1
	Measurement of the creep strain limit and behaviour of broken compact	BR2/ VC4	Unat	1200	-	2.3
Thermal effect	Variation of the thermal conductibility	BR2/ CEB 8	12	1400	9.35	0.6
	Binder effect on the mechanical behaviour of the fuel zone	DRAGON/ SPINES	12	1050		7

Table 6: Overview of BN experience in the HTR fuel performance (cont'd)

U FUEL RODS AND ELEMENTS						
Irradiation purpose		**Reactor/ Experiment**	**Enrich^{nt} (%U5)**	**Tmax (°C)**	**%FIMA**	**Fast Dose (10^{21} DNE)**
Rods	Effect of a low thermal flux radial gradient and an important fast flux radial gradient	DRAGON/ TI35	9	1250	10	4
	Behaviour of natural graphite compacts	DRAGON/ TI51	9	1270	3.9	1.3
	Behaviour under radial gradient of thermal flux	DRAGON/ TI43	9	1250		4
Block element	Long term irradiation of multihole graphite block	DRAGON/ MHGB-1A	8	1250		4
	Short term irradiation of a multihole graphite block	DRAGON/ MHGB-2A	93	1250		1

Pu PARTICLES					
Irradiation purpose		**Reactor/ Experiment**	**Tmax (°C)**	**%FIMA**	**Fast Dose (10^{21} DNE)**
Irradiation behaviour	Sol-gel / agglomeration process kernel	FRJ 2	1200	18	0.1
	Coatings of PyC derived from propylene instead of methane	BR2/ VC5	1400	10	4
	Comparison with U particle	DRAGON/ MET V	1550	1.35 - 3.0 - 10 - 35	0.36 - 1.08

Table 6: Overview of BN experience in the HTR fuel performance (cont'd)

3.1. BR2 Experiments

The BR2 reactor was used extensively for the Belgian HTR irradiation program:

- to test reference particles and compacts under extreme conditions;
- to irradiate new type of fuel;
- to investigate the irradiation induced changes of specific material properties.

Four types of rigs have been utilised in the BR2:

- hydraulic rabbit capsules to perform transient tests;
- the nose core of the MFBS-6 in pile sodium loop to irradiate compacts and particles;
- vented capsules to irradiate particles, compacts and coupons;
- a boiling water capsule to study the evolution of the thermal conductivity of the bonded fuel.

A general survey of the BR2 irradiation devices for high temperature gas-cooled reactors is presented in the following reference:

BR2 irradiation devices for HTGR fuel
P. VON der HARDT
EURATOM Report EUR 4737 e (1971)

3.1.1. BR2/ Hydraulic Rabbit

The main purpose of the irradiations performed in the Hydraulic Rabbit capsules of BR2 was the investigation of the behaviour of the particles under transient conditions and the resulting Amoeba effect. The parameters ruling this effect, i.e. the temperature of the particle, the thermal gradient across the particle and the irradiation time were investigated.

HR8 and HR22 irradiation were dedicated to particles with UO_2 kernels whereas HR21 and HR29 irradiations were carried out to investigate the relative behaviour of UC loose kernels.
Fuel characteristics and irradiation conditions are given in Table 7.

Figure 1 presents a typical cross section of UO_2 irradiated particles that have endured different thermal gradients.
As expected, the particles failed under extreme gradient (up to 1000°C/mm) (Figure 2).

A failure criterion was determined: the particles are at risk when the parameter *Irradiation time x thermal gradient across the particle* exceeds a critical value, which is function of the temperature.
A zone of safe operation was deduced and is illustrated in Figure 3.

A value of the conductivity of a bed of UC kernels has been deduced from HR21 experiment: 1.56×10^{-2} $W.cm^{-1}.K^{-1}$ for the temperature range of 1070-2420 K. After irradiation at temperature ranging from 2000 to 1390 K and under gradients from 890 to 950 $K.mm^{-1}$, the kernels were in good conditions, as shown on Figures 4. Most of them showed a central hole probably due to the fabrication process. Micrographies show a U_2C_3 + C precipitation between the grains and within the grains between the reticular planes (Figure 5).

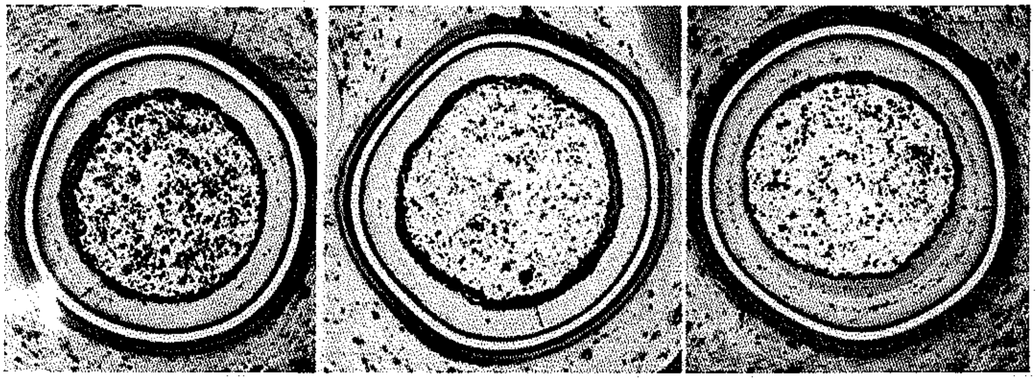
References:

High Temperature Gas Cooled Reactor Fuel
Annual Report BN 7303-10 (February 19723)

M. GAUBE
Coated particle irradiations in the BR2 reactor
Symposium "Results of five years of BR2 reactor utilisation"
MOL, Belgium (December 1973)

HYDRAULIC RABBIT EXPERIMENT					
		HR8	HR22	HR21	HR29
		Particles characteristics			
Kernel	Type	UO_2 nat.		UC (C/U=10)	
	Diameter	800 µm			
	Porosity	10%		10%	
Layer thickness	Buffer	45		-	-
	Inner PyC	45		-	-
	SiC	40		-	-
	Outer PyC	35		-	-
		Peak irradiation conditions			
W/particle		1.1	1.1	0.7	0.7
Time (hour)		10	10	10	1.5
Tmax (°C)		1800	1550	2400	2250
Environment		free	HD matrix	free	free

Table 7: BR2/HR experiments – Fuel characteristics and irradiation conditions



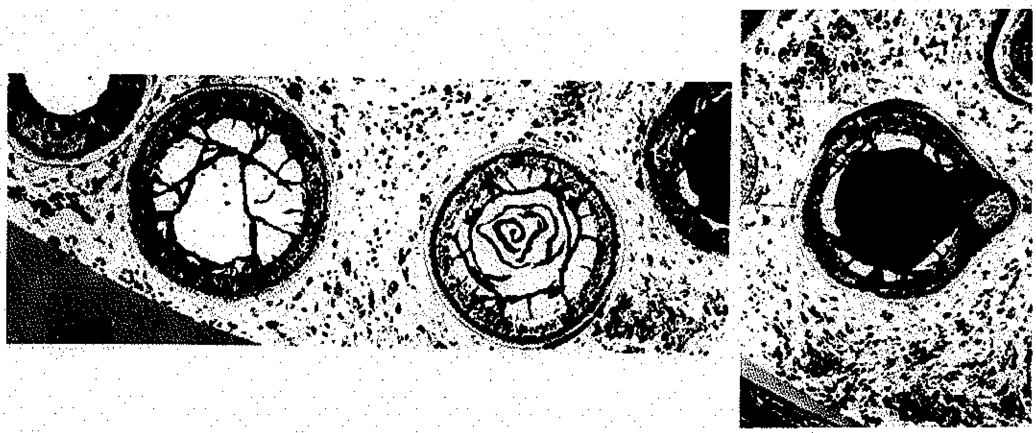
X50

T°= 1870°K
Gradient: 66°/cm
Irradiation time: 10h

T°= 1850°K
Gradient: 79.2°/cm
Irradiation time: 10h

T°= 1840°K
Gradient: 83.8°/cm
Irradiation time: 10h

Figure 1: BR2/HR-8 - Irradiated particles under transient conditions



X 30

Figure 2: BR2/HR-8 – Irradiated particles under high thermal gradient (>1000°/mm)

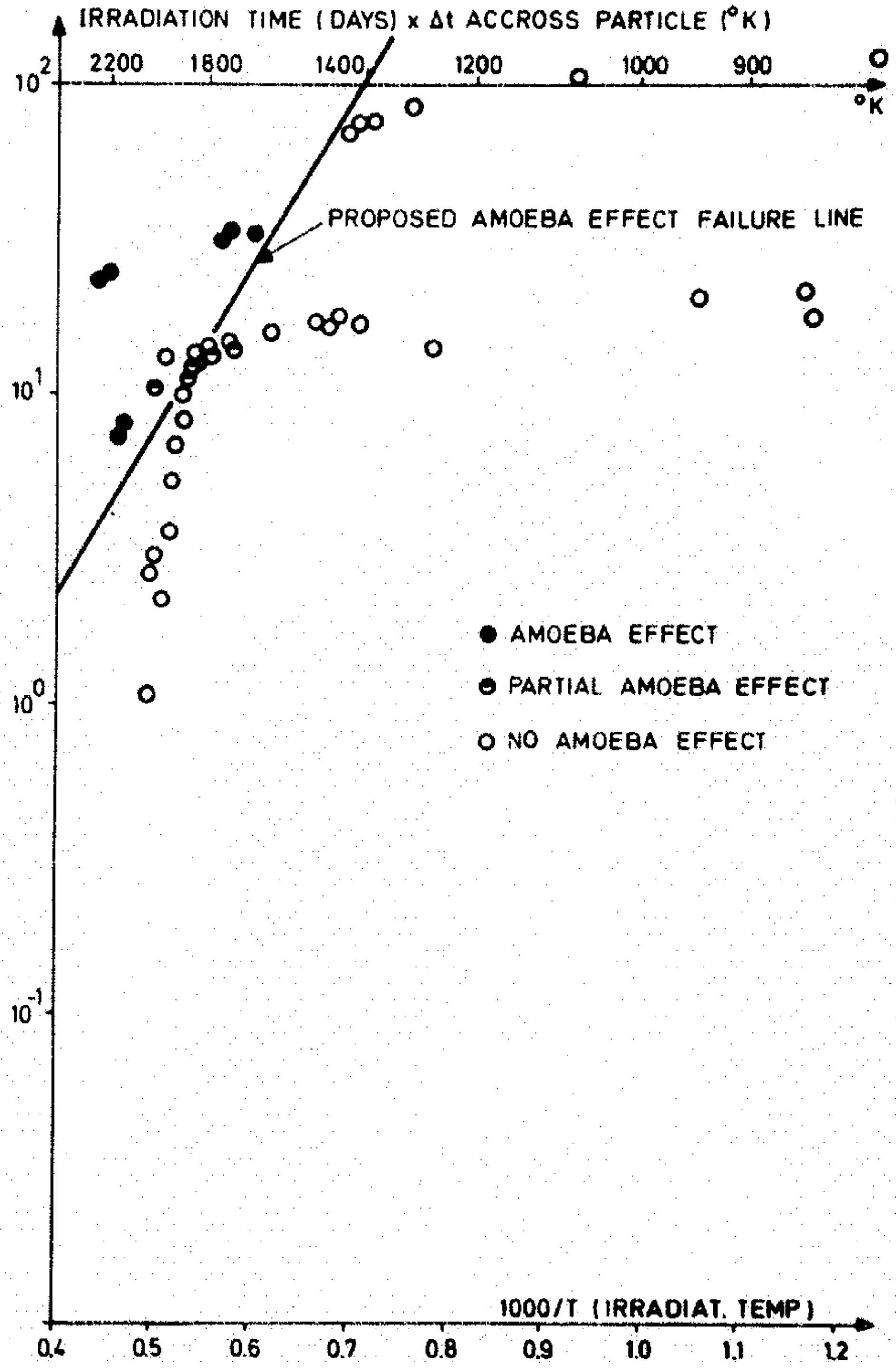


Figure 3: BR2/HR – Amoeba effect failure criteria

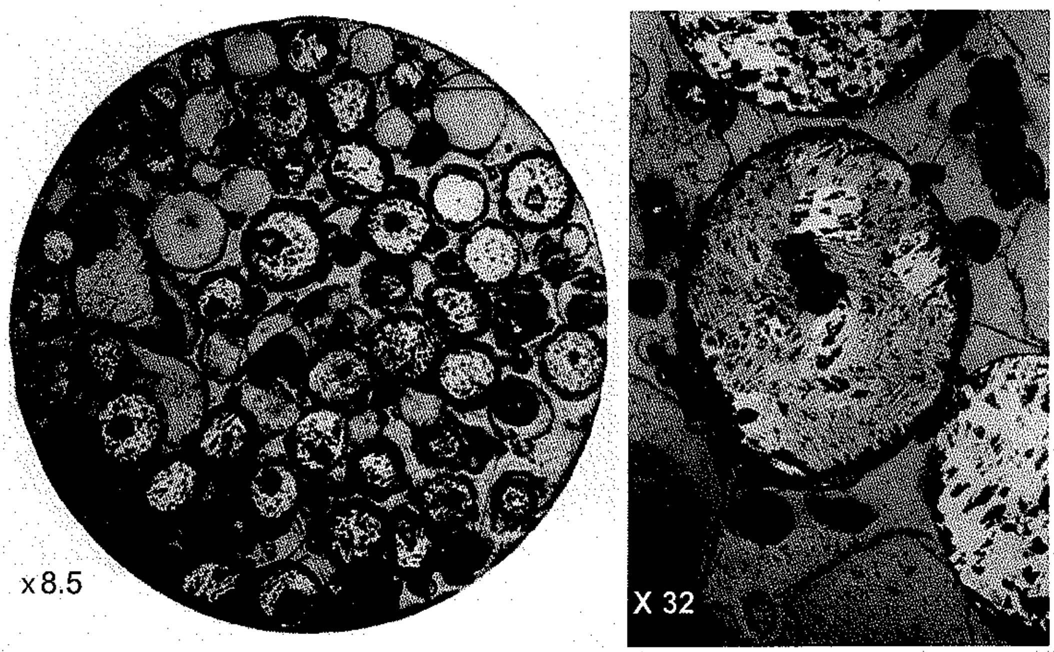


Figure 4: BR2/HR-29– Ceramographies of UC loose kernels

Figure 5: BR2/HR-29 – Details of the grain structure of UC kernel

3.1.2. BR2/ Sodium loop MFBS6

Compacts and particles were irradiated in the lower extension of the MFBS-6 in pile sodium loop of BR2. The objectives of this experiment were to measure the dimensional change of the matrix reference and the behaviour study of different coated particles at low temperature (below 660°C) and under low fast neutrons dose (about $2x10^{20}$ $n.cm^{-2}$ DNE).

The particles and compacts characteristics are given in Tables 8 and 9. The compacts were obtained by mixing two types of overcoated particles.

The post irradiation examinations have confirmed the good behaviour of the compacts and the loose particles. Most of the observed damages resulted from fabrication defect.

About the compacts:

One compact was broken, probably during the PIE handling (Figure 6).

Their external aspect did not change during irradiation, some superficial cracks were observable.

The particle distribution was not homogeneous, the density was higher in the external periphery than in the compact center.

Some circumferential cracks were visible in the porous SiC layer of some particles but have certainly arisen during the compaction step.

About the loose particles:

Batch TN 3/7/4F0: some particles presented the "spearhead effect", a phenomenon attributed to the shrinkage of the buffer resulting from the accommodation of the fission fragments damage; the external layers (SiC, PyC and porous overcoating) remained intact.

Batch TN 3/7/11/F: the particles were in good conditions, only some of them presented a spearhead effect limited to the porous buffer (Figure 7).

Batch TN 3/7/11/FM0C: cracks were visible in the thick overcoating layer; their origin (fabrication, irradiation or preparation for examination) was not clear.

Batch TN 3/12/1/F: the observation has revealed a low cohesion between the SiC and inner PyC layers that could lead to the apparition of a gap during longer irradiation time (Figure 8).

Batch TN S/S/S/S/3/1/1/F/ the SiC layer of some particles presented circumferential cracks (Figure 9); this defect has been corrected later by the increasing of the SiC density.

References:

R.A.U. HUDDLE, K. MAYR
Influence of fabrication parameters on the incidence of spearhead attack in coated particle fuel
IAEA Symp. Radiat. Damage Reactor Mater, 2nd (1969), Volume 2, 481-503

M. GAUBE
Rapport final de l'irradiation de combustible HTR dans l'appendice de l'expérience MFBS 6
BN report 7402-04 (January 1974)

Batch N°	TN/3/7/4/F0	TN/3/7/11/F MOC	TN/3/7/11/F	TN/3/12/1/F	S/S/S/3/1/1/F
Enrichment (%U5/U)	0.4	0.4	0.4	0.4	0.7
Kernel diameter (µm)	753	772	772	748	771
Manufacturing process	Powder agglomeration				Sol-Gel
Layer thickness (µm)					
Buffer	30	28	28	5.5	63.5
Inner PyC	73	63	63	31.5	33
SiC	33	24	24	37.5	38
Outer PyC	38	42	42	28	36.5
Porous layer	61	-	-	-	-
Overcoating	-	312	-	-	-
Particle diameter (µm)	1217	1720	1096	1053	1113

Table 8: BR2/MFBS-6 - Particles characteristics

CP's batch	S/S/S/3/1/1/F	S/S/S/3/1/1/F
CP diameter after overcoating (µm)	1820	1620
Overcoated CP average weight (mg)	7.3	5.24
Hg density (g/cc)	2.32	2.7
%U	30.9	43.2
Density of the overcoating layer (g/cc)	1.57	1.45
Proportion in compact	61.5%	38.5%
Compact external diameter (mm)	37.6	
Compact internal diameter (mm)	26.2	
Height (mm)	14.8	
Uranium weight (g)	8.67	
H.M.L. (g/cc)	1.0	

Table 9: BR2/MFBS-6 - Compacts characteristics

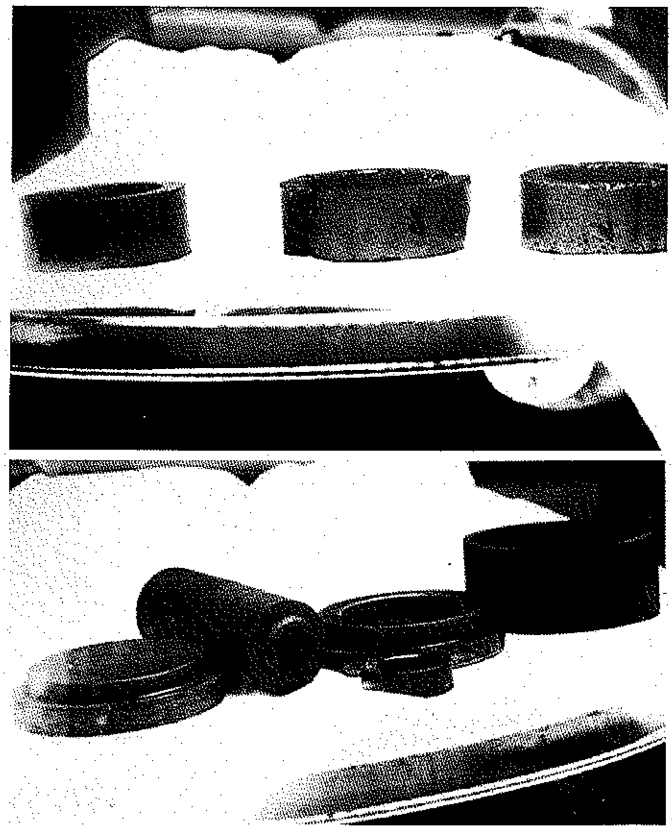


Figure 6: BR2/ MFBS 6 – Graphite components and compacts

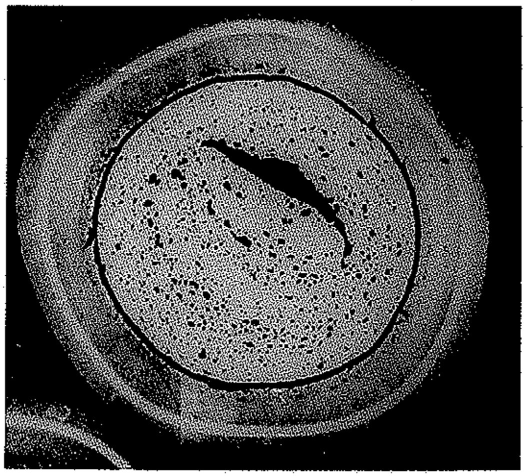


Figure 7: BR2/MFBS 6 – Loose particle showing spearhead effect after irradiation

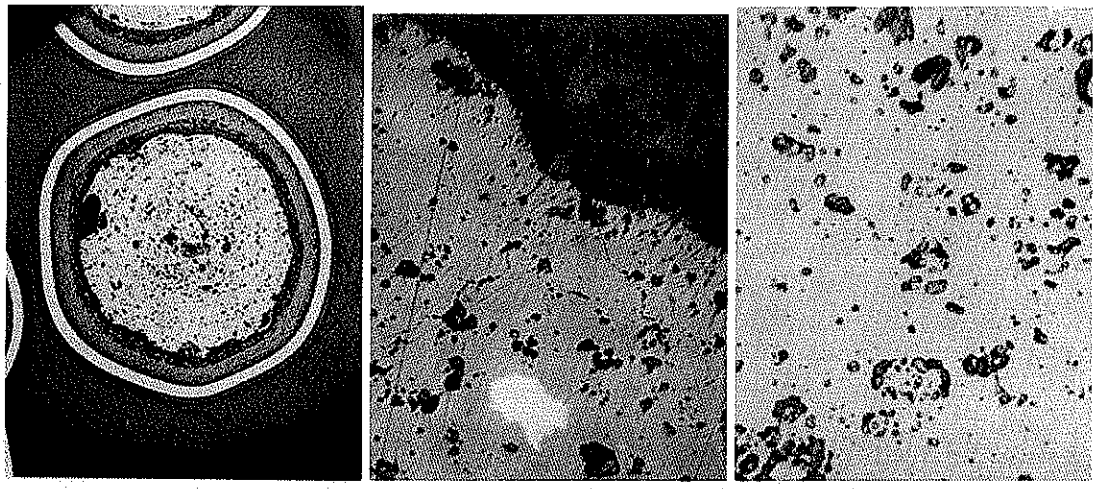


Figure 8: BR2/MFBS 6 – Ceramography of loose particle

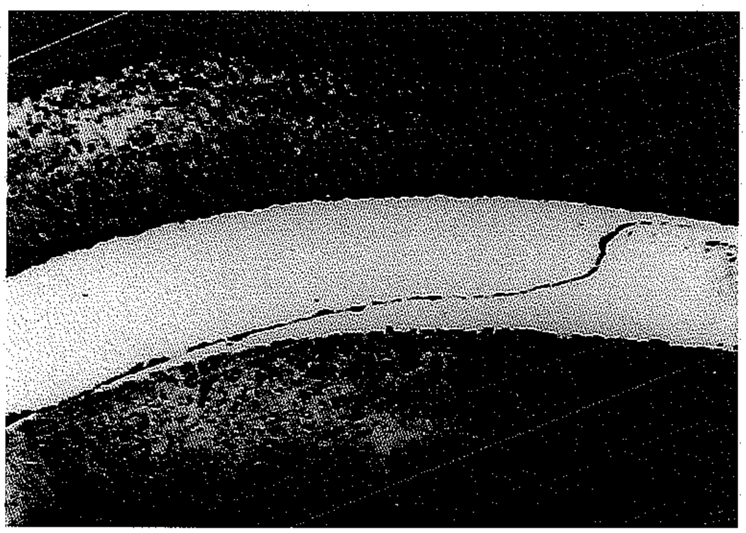


Figure 9: BR2/MFBS 6– Loose particle showing cracks in SiC layer

3.1.3. BR2/ Vented capsules

Among several rigs of the BR2 reactor located in Belgium, the vented capsules have been used extensively for the HTR irradiation programme.

They fulfilled the following requirements:

- allow a large variation of experimental parameters, such as target material, target dimensions, heavy metal loading, fuel enrichment, irradiation temperature and fission rating;
- temperature and power rating could be adjusted in a large range by varying the He_3-Ne mixture (which was used as an absorbent screen to modulate the local power) and by varying the in-pile section position in reactor;
- the fission gas release could be measured continuously.

BR2/VC1-2

The purpose of the two first VC experiments was to study the irradiation behaviour of compacts with coated particles manufactured by different production methods. Compacts were irradiated in BR2 up to a burnup of 8.7%FIMA at a maximum temperature of 1350°C. Fuel characteristics and irradiation data are detailed in the first deliverable D-TF4.3.

The main conclusions of this experiment are summarised below:

- the compacts showed a good stability under irradiation (Figure 10) but most of them showed cracks located in the region of high temperature gradient and some bare kernels could be seen;
- relatively high fission and reaction gases pressures (700 atm) have been measured on the irradiated particles which showed also a significant increase of the PyC anisotropy;
- most of the particles suffered from amoeba effect but this effect did not jeopardize the lifetime of the fuel (Figures 11)
- irradiation induced cracking was also noticed on some particles (Figure 12);
- the compact dimensional change and mechanical behaviour depend strongly on the accumulated fast neutron and irradiation temperature.

<u>References:</u>

Vented capsule VC-1 (MOPS-C1) and VC-2 (MOPS-C2) BR2 manufacturing report
L. CAIRNS, G. BOMBERNA
BN Report 7105–02 (June 1970)

MOPS- C01 (VC-1) Experiment – Final Report
M. GAUBE, J.M. THOMSON
BN Report 7410-02 (October1974)

MOPS- C02 (VC-2) Experiment – Final Report
M. GAUBE
BN Report 7410-03 (October 1974)

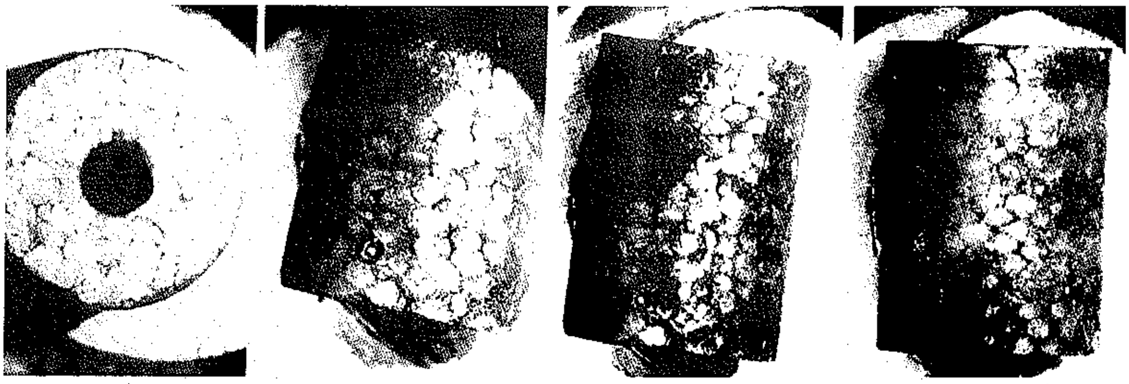


Figure 10: BR2/VC1 Compact 26

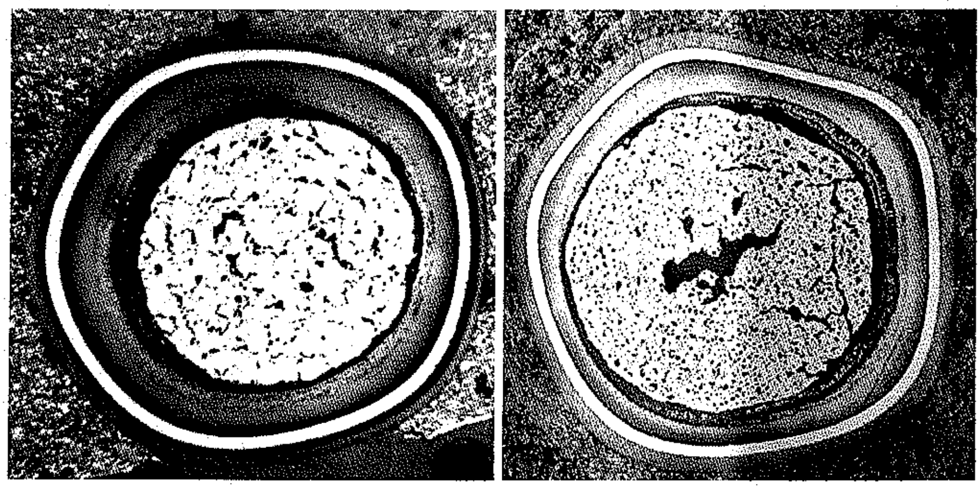


Figure 11: BR2/VC1 Compact 30 – Particle showing amoeba effect
(left compact 30 - right compact 15)

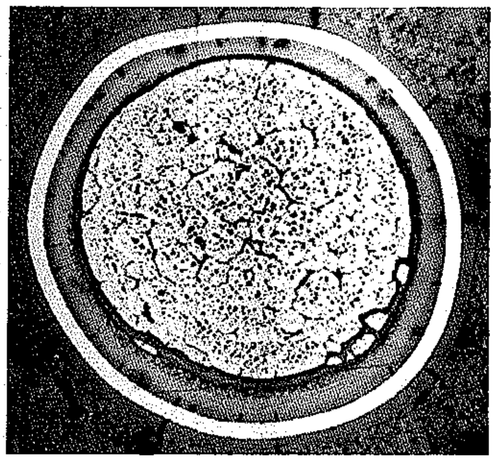


Figure 12: BR2/VC2 - Irradiation induced cracks in particle
(compact n°15)

BR2/VC3

The purpose of the third experiment in the BR2 vented capsule (VC3) was the irradiation of different types of UO_2 particles at 1100°C and up to the power reactor irradiation conditions.

This irradiation has included compacts manufactured by BELGONUCLEAIRE and loose particles supplied by the Dragon Project. The UO_2 kernels were manufactured by a slow growth agglomeration process, with a nominal porosity of 20% and nominal diameter of 800µm. The total layer thickness was about 160 µm.

The fuel reached a peak neutron dose of $3.55x10^{21}$ $n.cm^{-2}$ DNE and for the 9% enriched particle a peak burnup of 16.8% FIMA.

The R/B evolution for Xe^{133}, Kr^{85m}, Xe^{135} and Kr^{87} during irradiation are given in Figure 13. The results of the PIE may be summarised as follows.

The 9% enriched particles showed a very satisfactory survival rate despite the high dose and burn-up achieved, respectively $3.55x10^{21}$ $n.cm^{-2}$ DNE and 16.8% FIMA (Figure 14);

The 3% enriched particles irradiated in the low heavy metal loading compacts did not show any significant damage but those irradiated in the high heavy metal loading compacts suffered very much from the important distortions of the macroscopic temperature distribution; amoeba attack leading to particles destruction can be noticed on many ceramographies;

The 0.4% enriched particles showed less damage than the 3% enriched particles although some PyC cracking and amoeba effect can be noticed.

<u>References:</u>

High Temperature Gas Cooled Reactor Fuel
Annual Report BN 7602-07 (February 1976)

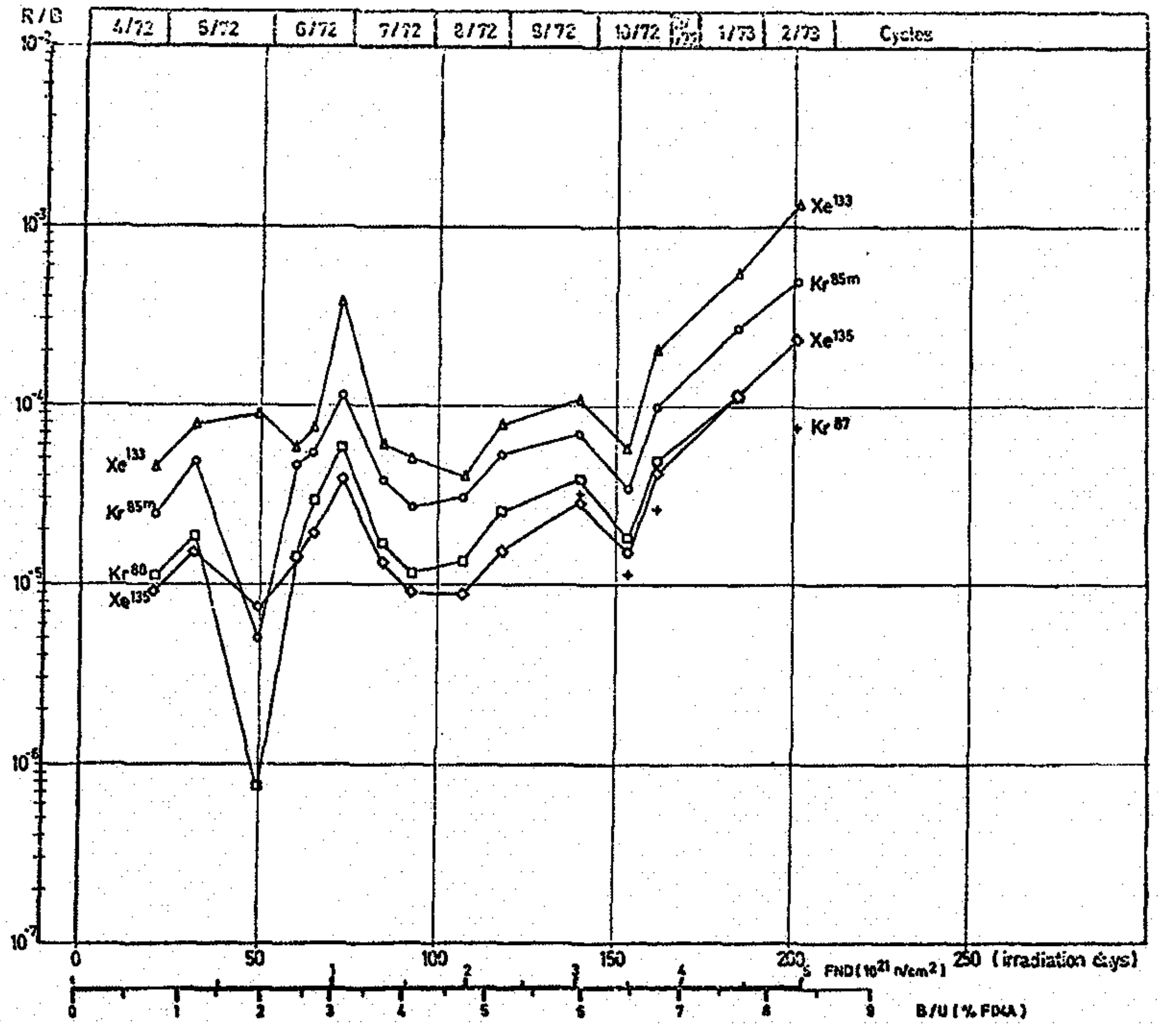


Figure 13: BR2/VC3 – R/B versus irradiation time

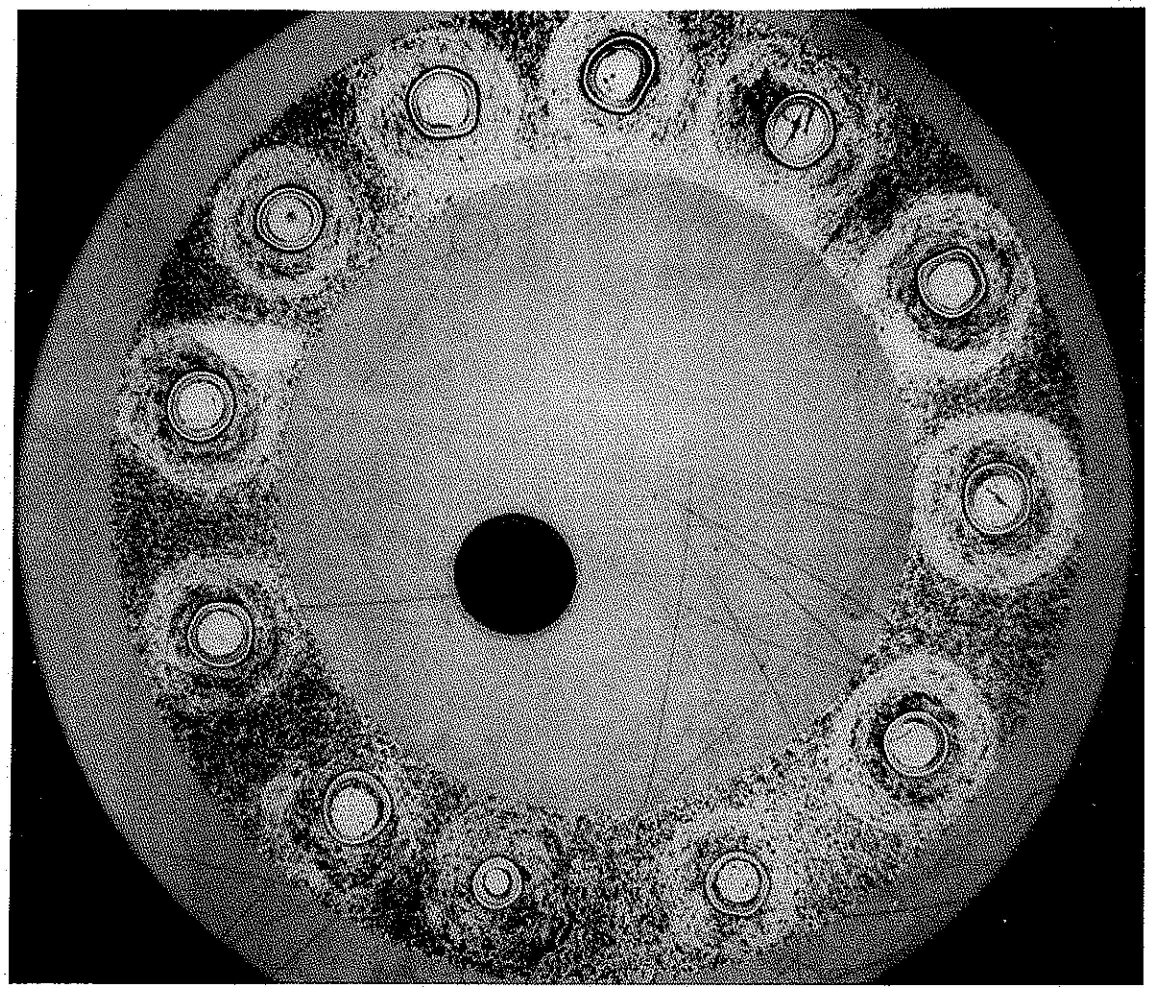


Figure 14: BR2/VC3 – 9% enriched particles after irradiation in a coupon

BR2/VC4

The fourth vented capsule experiment included compacts manufactured by BN/SCK and the Dragon Project and was part of the Euratom sponsored program in BR2 and HFR.

The purpose of the experiment was the investigation of the behaviour of annular compacts under peak stresses and peak strain conditions and the study of the failure mechanism.

Three series of three BN and DRAGON compacts with different gaps with an inner graphite spine leading to different degrees of interaction have been irradiated in the BR2 reactor. The BN compacts characteristics and irradiation conditions are described respectively in Tables 10 and 11.

The irradiation-induced shrinkage of the intact compacts was deduced from dimensional measurement before and after irradiation.

The code TOUCAN was used to calculate the peak hoop stresses evolution in the six BN compacts which had interacted with the inner spine during irradiation.

The input data were:

- Compact dimensions before irradiation
- Initial gap between compact and inner spine
- Temperature and dose evolution during irradiation
- Compact shrinkage rate

The calculations and the post-irradiation examinations lead to the following conclusions:

- All the broken compacts showed one single fracture (Figure 15) and remained in contact with the inner spine. Generally the rupture did not lead to the particle breakage. Leaching tests have revealed that the fraction of broken particles in compacts N°711 and 712 were respectively of the order of $3x10^{-4}$ and $0.7x10^{-4}$;
- For each series of three compacts:
  - the compact with the largest gap rupture (n° 742, 696 and 728) did not interact with the spine or did interact late in life so that the hoop stress remained significantly below the UTS, and did not therefore lead to any rupture;
  - the compact with the medium size gap interacted with the spine during irradiation and the resultant hoop stresses, relaxed mainly by the secondary creep, went through a maximum later in life before starting to decrease; the failure of the concerned compacts (n°711, 709 and 729) supposed that this maximum exceeded the UTS of the compacts.
  - the third compact with the smallest gap did interact during the first heating up and broke immediately, the stresses generated by this interaction being far above the UTS.

Typical evolutions of the hoop stress in such compact are given in Figure 16.

References:

The creep strain limit experiment
M.R. EVERETT, M. GAUBE
Journal of Nuclear Materials, 65 (1976) 116-130

Mechanical design of VC4
M. GAUBE
BN Note 560.70/390/2/127M (November 1970)

Kernel	
	Natural UO_2 Porosity: 10.22% Diameter: 797 µm
Coatings	
Inner PyC layer	Thickness: 103 µm Density: 1-1.65 g/cm^3
SiC layer	Thickness: 35 µm Density: 3.2 g/cm^3
Outer PyC layer	Thickness: 37 µm Density: 1.73 g/cm^3
Total coating thickness:	175 µm
Overcoating	
Material resin bonded (14%) natural graphite	
CP/ Overcoated CP: 58.14%	
Compacts	
Outer diameter:	21 mm
Inner diameter:	12 mm
Length:	18 mm
Material:	natural graphite (LCL, 14% resin)
UTS:	5.9 to 7.8 MPa
E:	7.7x10^3 MPa
Volume loading:	35%

Table 10: BR2/VC4 - Fuel characteristics

COMPACT N°	Initial Diametral gap between compact and spine (µm)	Fast fluence (10^{21} n.cm^{-2} DNE)	Burnup (% FIMA)	Mean irradiation Temperature (°C)	Observation after irradiation
742	299	1.91	3.41	980	No crack
711	123	2.10	3.64	1000	Axial crack
712	34	2.16	3.77	1030	Axial crack
696	311	2.54	4.45	1120	No crack
709	130	2.54	4.59	1130	Axial crack
721	51	2.54	4.59	1140	Axial crack
728	304	2.41	4.41	1075	No crack
729	125	2.35	4.13	1050	Axial crack
741	50	2.23	3.91	1020	Axial crack

Table 11: BR2/VC4 - Irradiation parameters of compacts

	IRRADIATION INDUCED DIMENSIONAL CHANGE (%)			
Compact N°	Inner diameter	Outer diameter	Height	Poco-spine outer diameter
742	-1.33	-0.83	-0.61	+1.01
696	-2.38	-1.47	-0.17	-0.38
728	-1.9	-1.3	-0.72	+1.01

Table 12: BR2/VC4 - Dimensional change of compacts after irradiation

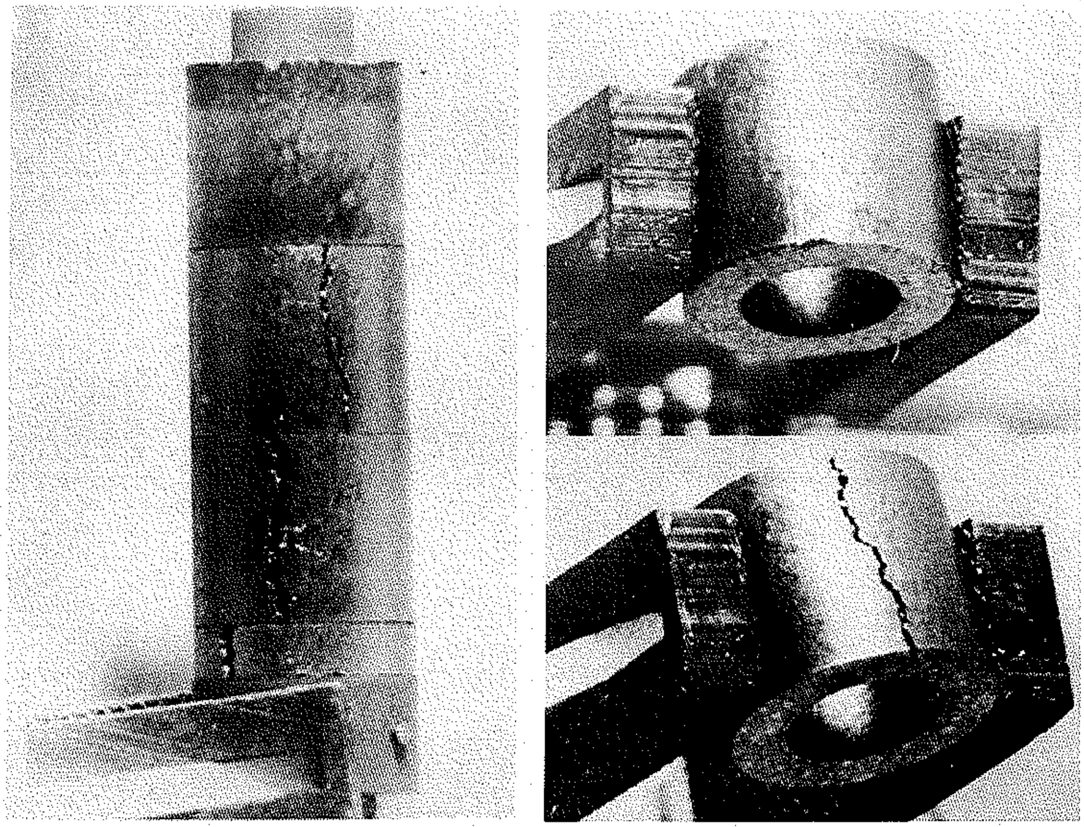


Figure 15: BR2/VC4 – Irradiated compacts

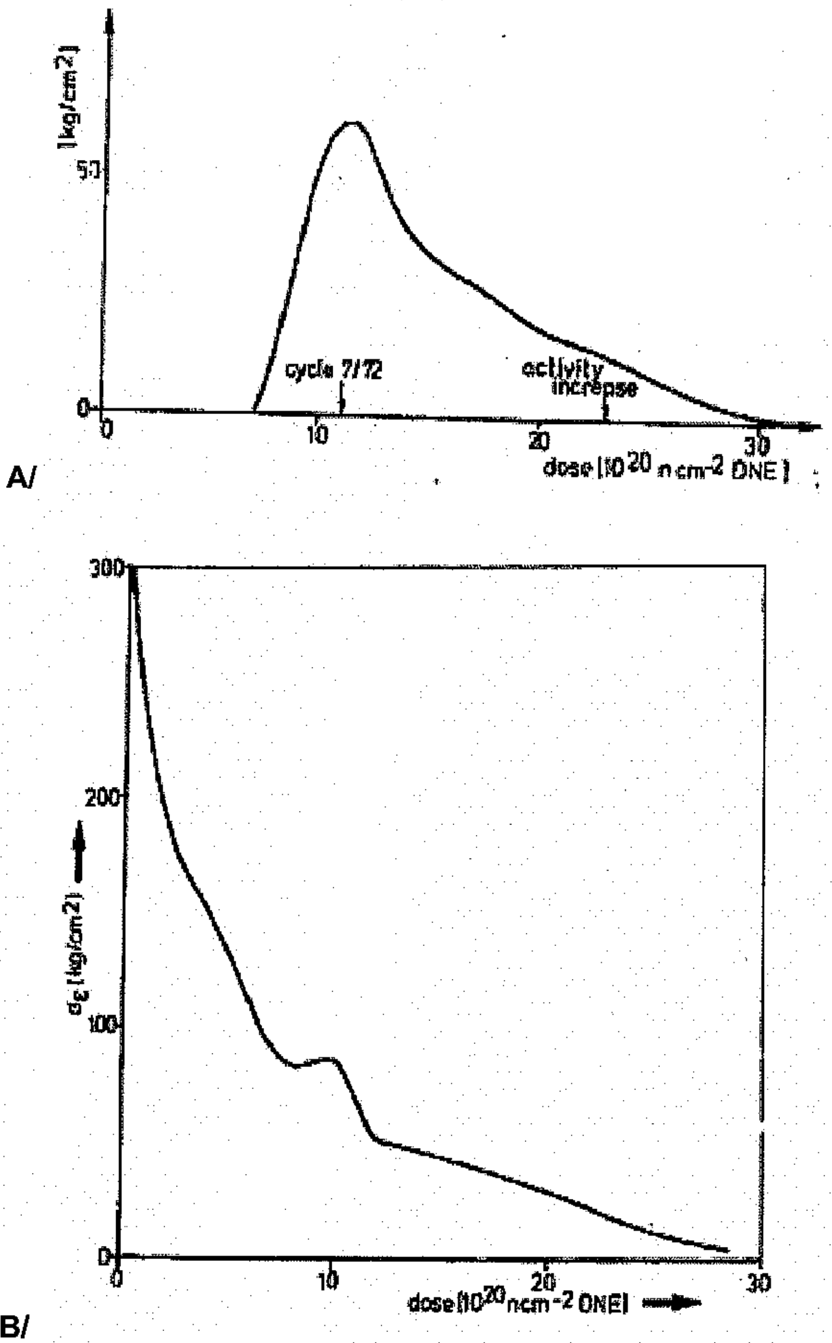


Figure 16: BR2/VC4 – Maximum hoop stress evolution in compact
A/ with gap compact/spine = 130µm
B/ with gap compact/spine = 50µm

BR2/VC5

This experiment was the last of the Euratom sponsored program in BR2 and HFR. Its purpose was the comparison of the behaviour of methane and propylene coated particles when irradiated up to a temperature of 1400°C.
The objective to perform coating derived from propylene was to overcome the mechanical failure or at least to delay it to very much higher dose than the coating derived from methane.
The fuel was supplied by Dragon Project and BELGONUCLEAIRE; the UO_2 (with different U5 enrichments) and PuO_2/20C (type D) kernels were coated at Mol. A series of tests was carried out in the 75 and 40mm coaters. The major problem encountered during coating was the temperature control: to obtain deposits with acceptable characteristics, the temperature changes should not exceed 25°C compare to 50° for the methane. For a good reproducibility of the coating characteristics, an automatic temperature control of the furnace was necessary.
Some metallographies of some loose U particles are shown in Figure 17.

The experiment was unloaded in April 1975 after having reached a peak fast neutron dose of 6.6×10^{21} $n.cm^{-2}$ DNE and a peak burnup of 13.9%FIMA for the 5% enriched particles and about 90%FIMA for the Pu particles.

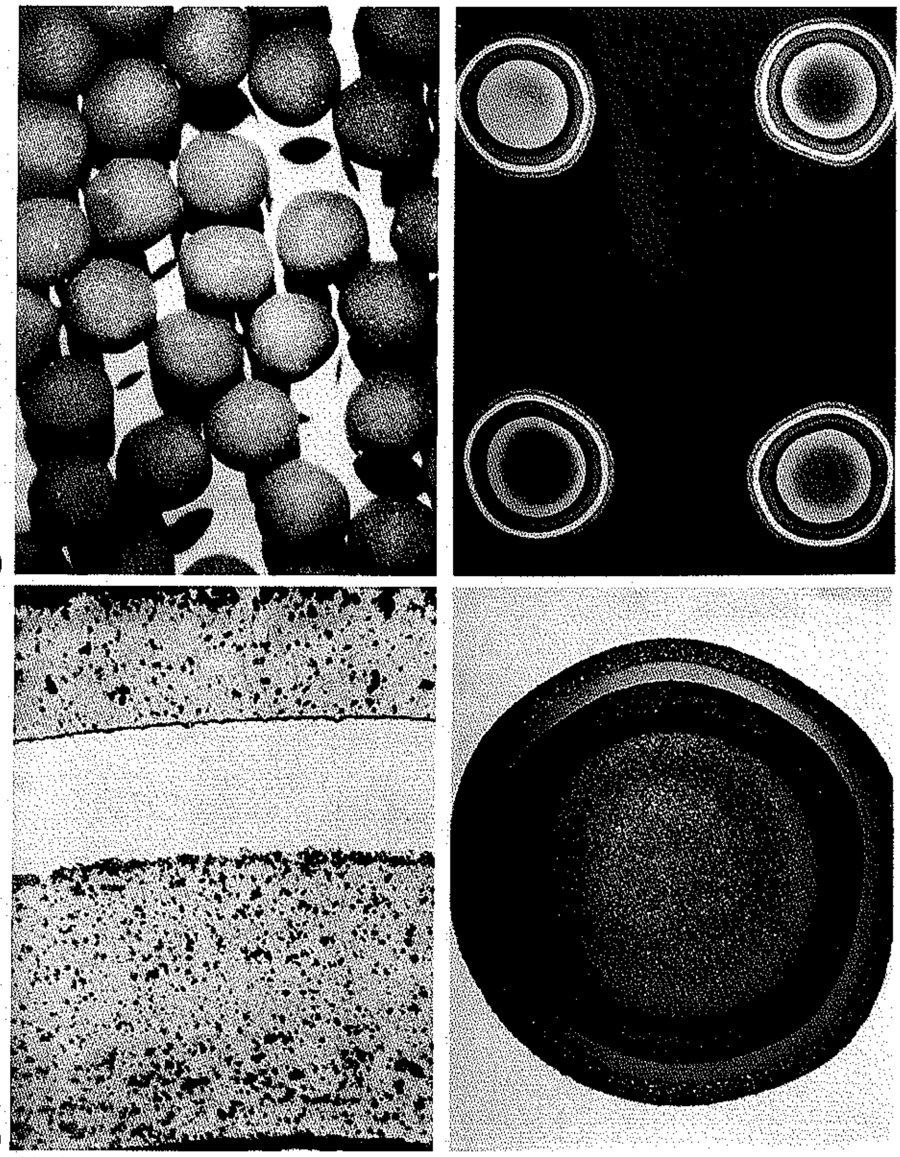
The post-irradiations examinations, carried out at Petten, have confirmed the better behaviour of the propylene-coated particles, which show high survival rates (Figure 18)). The only observed damage was restricted to the inner coating and to kernels. Propylene was consequently used for the deposition of the pyrocarbon layers.

The visual examinations of the tray with Pu loose particles has revealed that all the particles but two had exploded under these far too severe irradiation conditions (Figure 19-A). These two intact particles were picked up for ceramographic analysis which revealed a high reaction of the kernel with the inner PyC layer (Figure 19-B).

<u>*References:*</u>

Post Irradiation examination of a 1300°C HTR fuel experiment project J96-M3
J. de BUEGER, H. ROTTGER, Th. SCHOOTS
EUR Report 5841 E (1977)

X10 X15



X330 X50

Figure 17: BR2/VC5 - Particle with coating derived from propylene

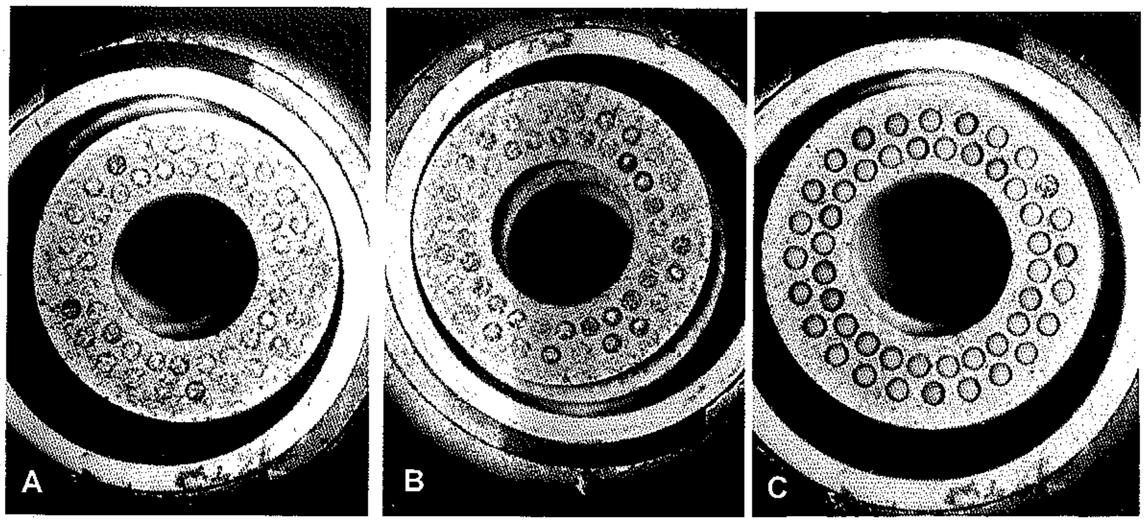


Figure 18: BR2/VC5 - Methane and propylene coated particles after irradiation
A/: 3% enriched methane coated particles
B/: 7% enriched methane coated particles
C/: 5% enriched propylene coated particles

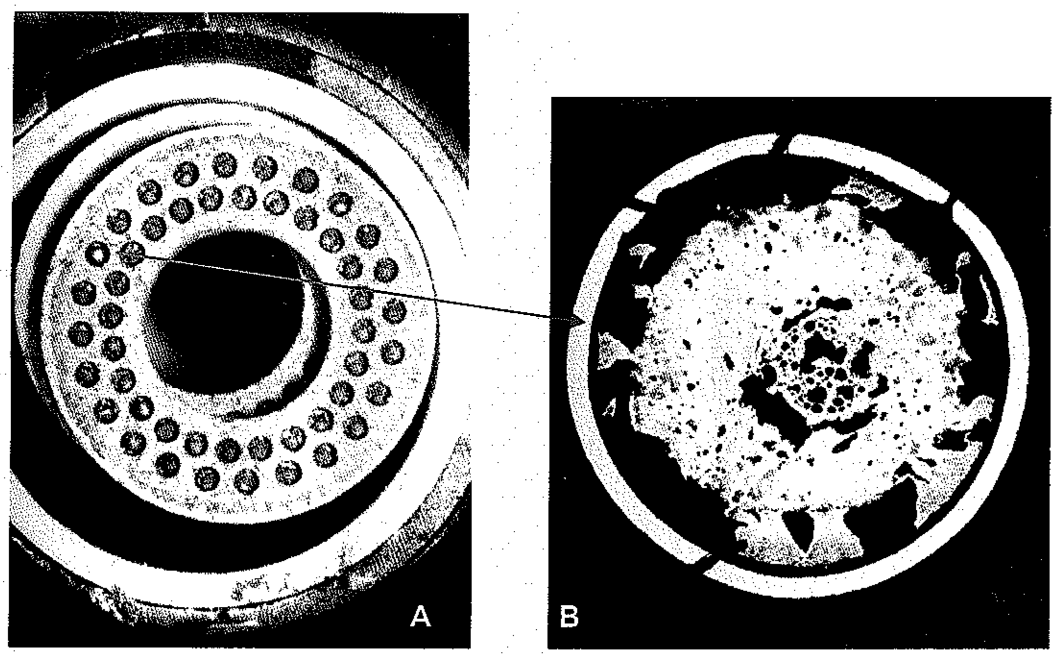


Figure 19: BR2/VC5 – Pu particles after irradiation
Remark: the cracks in the SiC layer were not due to the irradiation but to the use of a too coarse grinder during ceramography preparation.

3.1.4. BR2/CEB-8

The evolution of the thermal conductivity of the compact was very important especially in view of the utilisation of low density matrices. The first experiment on the irradiation induced change of the thermal conductivity was performed in the boiling water capsule CEB-8 of the BR2 reactor in 1969, the test was devoted to consolidated fuel with a rather low initial thermal conductivity.

The main characteristics of the fuel segment are given in Table 13.

External diameter of the cladding (mm)	14.5		
U5 enrichment (%)	12		
Fuel segment identification	AB	HG	EF
In pile position	lower	median	upper
Total length (mm)	250	100	250
UO_2 Linear weight (g/cm)	1.03	0.32	1.02
Volume loading (%)	50	50	50

Table 13: BR2/CEB-8 - Characteristics of the fuel segment

The main irradiation parameters of the experiment were:

- A peak irradiation temperature of 1400°C
- Average linear power of 200 W/cm
- A peak burn-up of 93500 MWd/$t_{eq.}$U
- Peak fast neutron dose: $0.6x10^{21}$ n./cm^{-2} DNE

The results of this experiment are summarised in Figure 20: the thermal conductivity was found to be of the order of 0.05 W/cm°C at 700°C and to be strongly temperature dependent. No effect of fast neutron dose was detected and the post irradiation examinations have indicated the particles to be intact.

<u>References:</u>

Rapport final de l'experience CEB-8
M. GAUBE, B. VAN OUTRYVE D'YDEWALLE, A. LHOST
BN Report 7307-01 (July 1973)

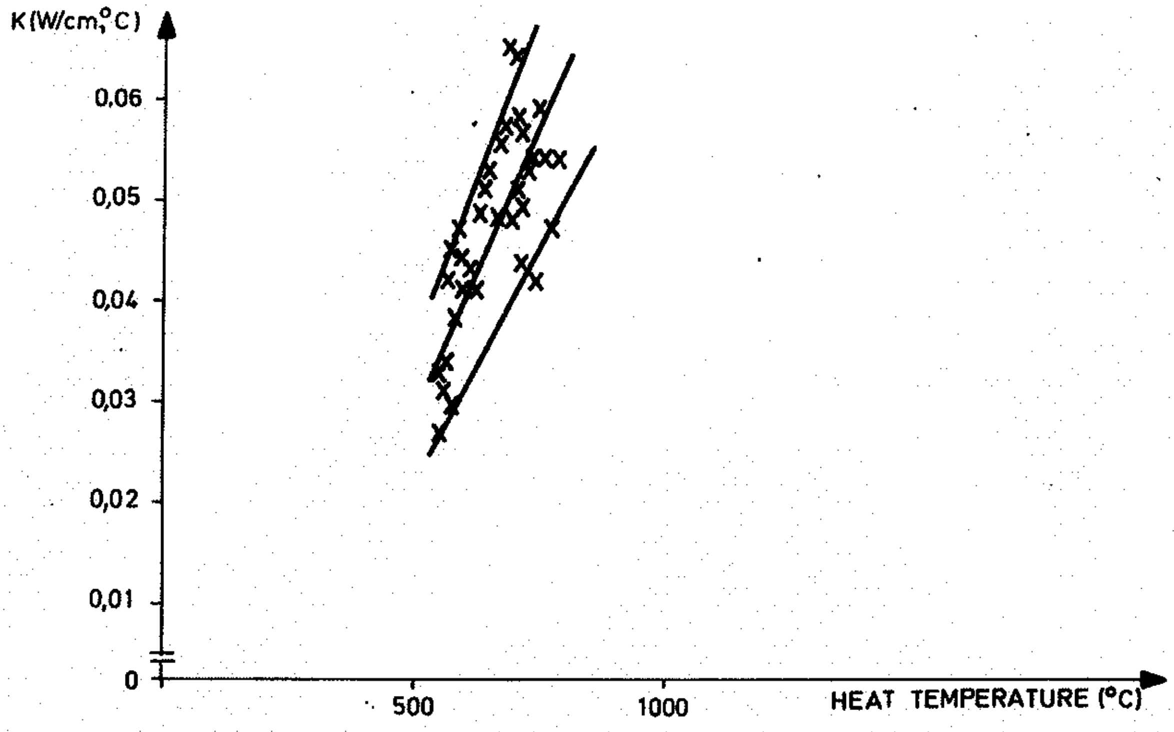


Figure 20: BR2/CEB-8 – Bonded fuel conductibility versus temperature

3.2. HFR Experiments

The experiments that were performed in the HFR Petten had essentially dealt on the HTR fuel creep measurement.

3.2.1. HFR/J98

This experiment was part of the EURATOM sponsored programme in HFR and BR2. Its purpose was the determination of the irradiation induced primary and secondary creep coefficients of fuelled compacts under tensile load.

Three identical creep assemblies, shown in Figure 21, were fabricated and loaded with samples delivered by Dragon Project, KFA and BELGONUCLEAIRE. The BN samples were dummy coated particle compacts (with carbon kernels), that had dumbbell shaped with a rectangular cross-section. Their characteristics are given in Table 14.

Their irradiation was carried out in the third capsule (J98-3) in the HFR at a temperature of 900°C, under an average fast flux of 0.77×10^{14} $n.cm^{-2}.s^{-1}$. A constant tensile stress between 40% and 50% of the ultimate tensile stress, i.e. about 1.3 MPa was maintained.
The irradiation was stopped after a reached dose of 6.3×10^{20} $n.cm^{-2}$ DNE by the rupture of the central sample.

The recording of the stress, of the sample columns elongation and of the temperature are given at Figure 22 as function of time and DNE dose for the BN samples.
The results of the three J-98 experiments are summarised below.
The irradiation creep of matrix and compact material consists of a transient and a steady state part. For a given temperature, the total creep strain can be expressed as follows:

$$\dot{\varepsilon} = P\frac{\sigma}{E}\left(1-e^{-aN}\right)+K\sigma N$$

where: $\dot{\varepsilon}$: creep strain
σ : stress
E: Young's modulus
N: Dido nickel dose
P, a, K: coefficients

The comparison of the values of the coefficients a, K and P deduced from the three experiments J-98 with the values published in the literature, has lead to the following remarks:

- In the three experiments, the (E/P) values were close to one another. The values of P deduced by taking the Young's modulus of 5.9×10^{3} MPa measured on compacts of the E-99 experiment range from 2.0 to 2.5. they should be compared with the value of about 1.0 given for graphite: they are of the same order of magnitude.

- The measured values of the primary creep coefficient range from 1.5 to 6.4; the mean value was of the same order of magnitude as the given value foe graphite (about 4.0×10^{20} $n.cm^{-2}$ DNE).
- The secondary creep coefficient deduced from the E-98-3 experiment was 60% higher than the value obtained from the E-99 experiment.

Finally, these remarks have lead to the following conclusions:

- the above mathematical expression used for the irradiation induced creep in graphite seems to be convenient for the matrix material and fuelled compacts;
- the calculated coefficients were in agreement with the data published in the literature.

References:

Joint Programme of uni-axial creep measurements of HTR compacts and matrix material under tensile load at 900°C
M.R. EVERETT, R. LÖLGEN, G. POTT, J.M. THOMSON
Journal of nuclear materials 65 (1977) 107-115

Matrix material	Resin bonded natural graphite
Particle type	PyC/SiC/PyC on carbon kernel
Overall particle diameter (µm)	1065
Volume loading (%)	32
Density (g/cm^3)	1.69 – 1.71
$Section_{matrix}/Section_{sample}$	0.53
Length of the column under tension (mm)	47.70
Young's modulus (MPa)	8.9×10^3
UTS (MPa)	1.6 – 4.2
Rupture stress (MPa)	3.1 – 7.9

Table 14: HFR/J98-3 - BELGONUCLEAIRE compact characteristics

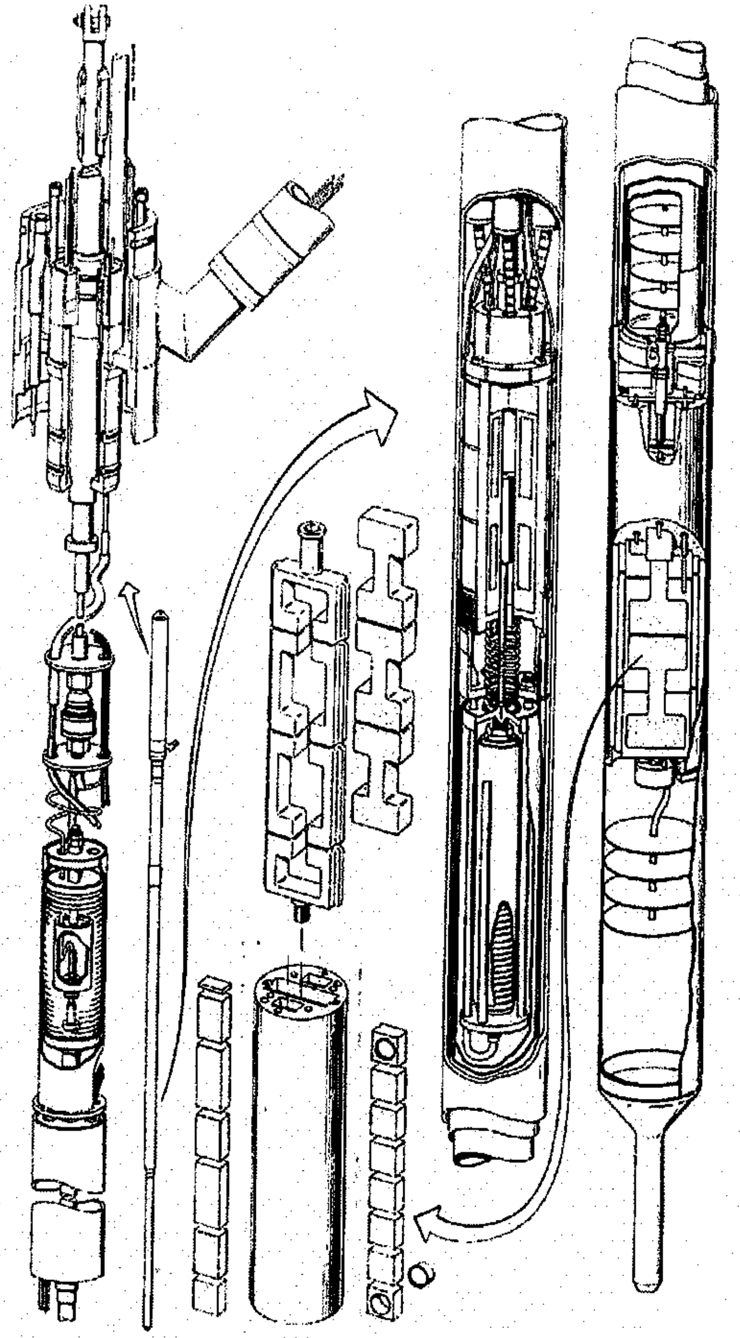


Figure 21: HFR/J-98 - General assembly of the experimental device

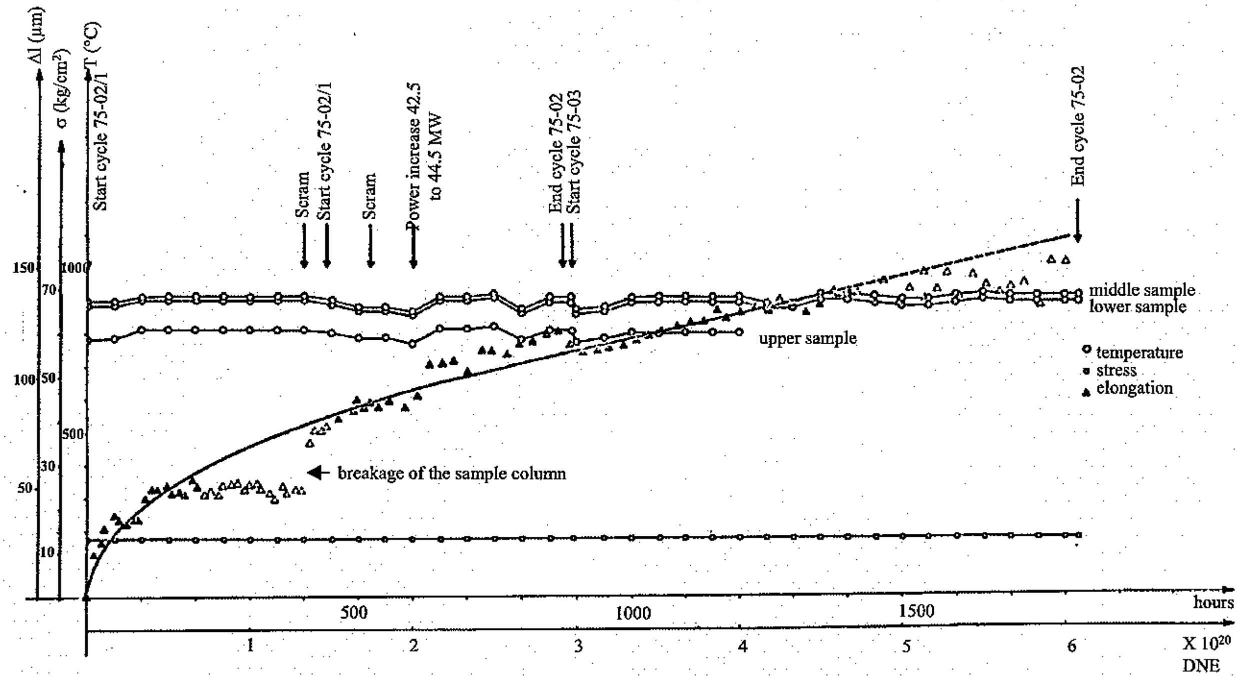


Figure 22: HFR/J98-3 – Temperature, stress and elongation during irradiation

3.2.2. HFR/E99

The purpose of this experiment was the investigation of the steady state creep coefficients of HTR compacts.

A serie of 3 restrained shrinkage experiments were performed in the HFR Petten on various dummy coated particle compacts, at temperature of 600°C, 900°C, 1050°C and 1200°C up to a neutron fluence of maximum $3x10^{21}$ $n.cm^{-2}$ DNE. The samples were provided by 3 different manufacturers (NUKEM, DRAGON, BELGONUCLEAIRE), and were identical to those of experiment J-98 (dumbbell shape).

It was found that the creep behaviour of these materials, as deduced from the observed stress build-up and restrained dimensional changes, could be described satisfactorily with a mathematical expression, based on the same simple assumptions that are normally used to describe the radiation creep of reactor graphites.

No evidence was found for any significant systematic difference between the various materials (dummy coated particles compacts with carbon kernel or coated carbon kernel, pure matrix material).

Creep strains up to 2.5% were observed, but there was no evidence that this represented a real creep strain limit of these materials.

The radiation creep coefficients of these materials are of the same order of magnitude as those of reactor graphite. The values range from 4 to $8x10^{-12}$ $(N.m^{-2})^{-1}(10^{20}\ n.cm^{-2})^{-1}$ at 600°C, 8 to $30x10^{-12}$ $(N.m^{-2})^{-1}(10^{20}\ n.cm^{-2})^{-1}$ at 1200°C. The values for the compacts, like those for the graphites thus form a relatively wide band. Part of the band width is due to experimental uncertainties. These bands for compacts and graphites partially overlap, that for the compacts being on the upper side. The observed temperature dependence of the creep is similar to that of graphite creep.

The creep process was not found to have a large influence on the irradiation induced property change.

In some restrained specimens, it was observed at 1100°C that irradiation induced cracks around the coated particles were oriented perpendicular to the direction of the stress whereas they were randomly distributed in unrestrained compacts. These cracks led to an increase of the creep constant and therefore to stresses lower than those calculated.

<u>*References:*</u>

High Temperature Gas Cooled Reactor Fuel
Annual Report BN 7503-03 (March 1975)

3.2.3. HFR/Small Compacts 1 and 2

The purpose of these irradiations was the investigation of the evolution of different matrix mechanical properties under irradiation in the HFR Petten.

The first irradiation concerned Gilsocarbon based matrices compacts with dummy particles (vitreous carbon) with one or two PyC layers. It covered a wide temperature range (400-1400°C), and reached a peak dose of 5.3×10^{21} $n.cm^{-2}$ DNE. The evolution of the shrinkage, Young's modulus and coefficient of thermal expansion evolutions were recorded. As example of results, the axial shrinkage evolution of Gilsocarbon compacts is given in Figure 23.

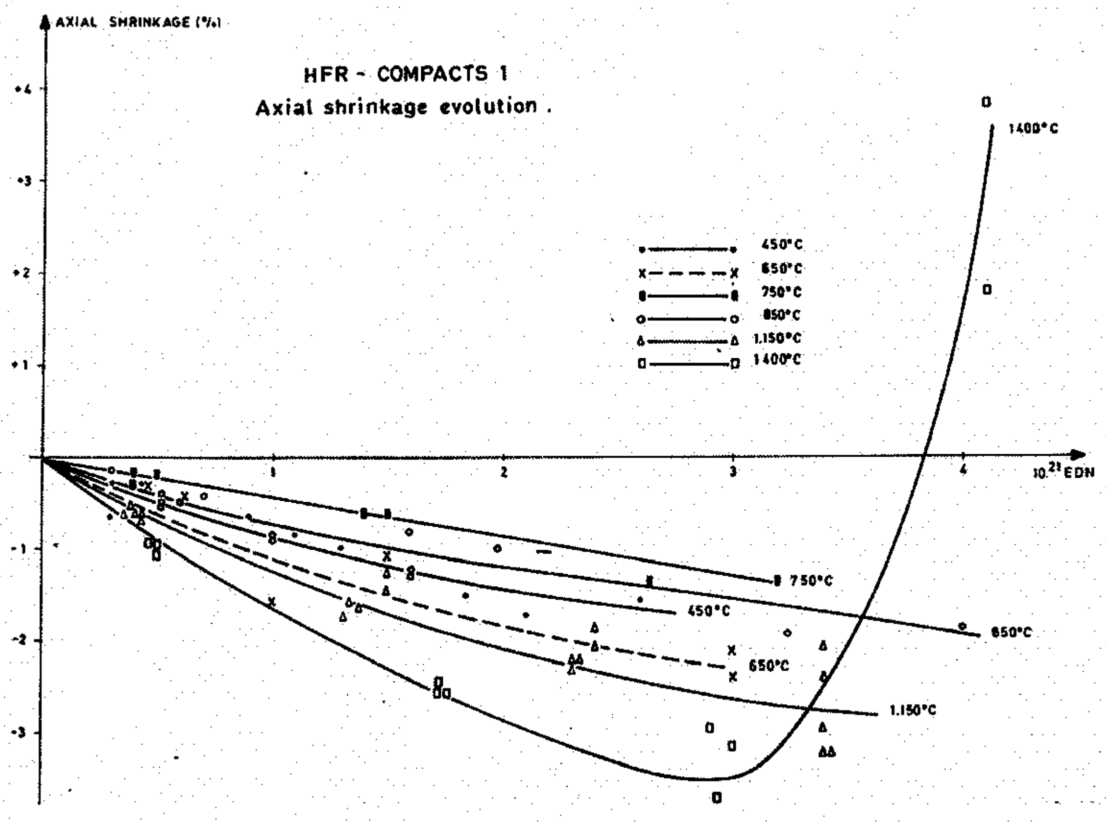


Figure 23: HFR/Small Compacts 1 - Axial shrinkage evolution

The second experiment was the repetition of the previous one with natural graphite matrices presenting different degrees of polymerisation.

The evolution of the compact density, Young's modulus, electrical resistivity, axial and radial dimensions, was determined.

These measurements can be summarised as follows:

- the densification increased with the irradiation temperature but no significant effect of the compaction time was noticed;
- the Young's modulus changes were inversely proportional to the compaction time;
- the electrical resistivity increased with the fast neutron dose but no effect of the compaction time nor the irradiation temperature has been noticed.

The limited number of measurement points and the early termination of the Dragon Project who was a co-sponsor of this experiment, gave preliminary information on the property changes of natural graphite compacts up to a dose of 2.5×10^{21} $n.cm^{-2}$ DNE.

<u>*References:*</u>

Rapport final de l'experience d'irradiation Small Compacts 1
M. GAUBE
BN Report 7401-04 (January 1974)

Small Compacts 2 Experiment Final Report
J.M. THOMSON
BN Report 7606-01 (June 1976)

3.2.4. HFR/E96-1

This experiment was part of the Euratom sponsored programme in HFR and BR2. Its purpose was the investigation of the performances of different loose triso-coated particles with UO_2 and $(Th,U)O_2$ kernels and compacts over a range of burn-up and fast neutron doses with fuel surface temperatures of about 900°C.

This irradiation was stopped after 27 days when a serious leakage of the irradiation device was detected: the peak fast fluence was $1.2x10^{21}$ $n.cm^{-2}$ DNE and a peak burn-up of 6% FIMA has been reached. The R/B values for Kr85m and Xe133 remained respectively at10^{-7} and $2.5x10^{-6}$.

Post-irradiation examinations were carried out at Petten, and the main results are summarised as follows:

- the visual examination showed that all the compacts and the coupons were intacts (Figure 24);
- all the compacts and coupons shrank under irradiation from 0 to 0.5%;
- the particles were in good conditions; a kernel sintering was noticed in some of them.

References:

The first joint 900°C HTR fuel irradiation experiment in the HFR Petten Project E96-01
H. ROTTGER
EURATOM Report 5147 E (1974)

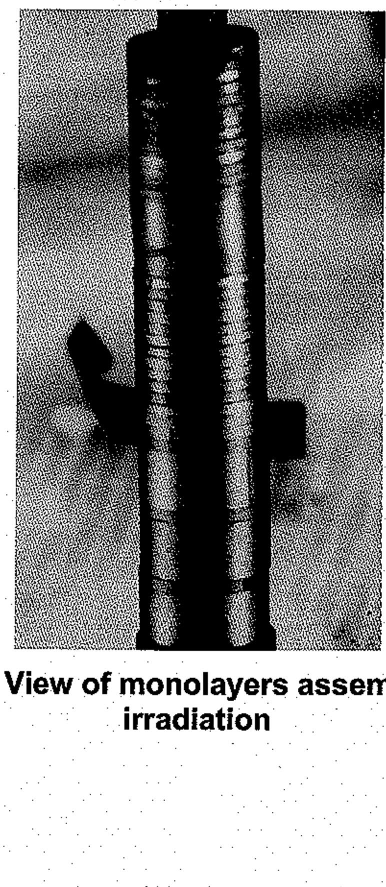


Figure 24: HFR/E96-1 - View of monolayers assembly and compacts after irradiation

3.2.5. HFR/E96-2

This experiment was decided because of the premature unloading of the E96-1 experiment. The irradiation was pursued at 900°C up to peak fast neutron dose of 5.96×10^{21} $n.cm^{-2}$ DNE and a peak burnup of about 14% for the highly enriched particles (9%U5).

Visual examination has revealed the good behaviour of the 0.4% and 3% enriched particles and a high breakage fraction of the 9% enriched particles.

A migration of the kernel was noticed in most of the particles that were observed by ceramography. However, this migration was less severe than in other higher temperature experiments (1100 and 1300°C): it never affected the inner high density PyC layer and did not seem to be responsible for any particle failure. The pyrocarbon deposit on the cold side of the kernel could be seen on many particles. Finally, in some particles, fragments of kernel were left behind this migration and were then surrounded by the pyrocarbon deposit.

The visual examinations have shown that most of the 9% enriched fuel broke by excessive fission gas pressure: radial cracks were visible in the SiC and outer layers of this type of fuel. Similar cracks were also observed in low enriched particles where excessive pressure was unlikely; this breakage was attributed to the use of a too coarse grinder for the polishing step.

The irradiation induced shrinkage of the outer PyC has led to the rupture of this layer by excessive stresses on many particles.

However, on most of the particles, fragments of the outer PyC curled outward and were lost during the polishing (Figure 25).

The propagation of the cracks in the Sic layers through the outer PyC layer was more frequent than through the inner PyC layer: this seemed to indicate that the SiC adhere strongly to the outer than to the inner PyC.

Some of the cracks have gone across the three layers.

In conclusion,

- the amoeba effect did not seem to be responsible of any particle failure;
- the breakage of the outer PyC layer by excessive irradiation induced shrinkage and the subsequent SiC layer cracking due to the too high inner gas pressure seemed to lead to the failure of most of the particles;
- the high ratio of broken 9% enriched particles was due to the high burn-up which has induced excessive fission gas pressures.

<u>*References:*</u>

The second Euratom sponsored 900°C HTR fuel irradiation experiment in the HFR Petten, Project E-96-02
H. ROTTGER, J. de BUEGER
Part 2: Post-Irradiation Examination
EURATOM Report 5463 E (1977)

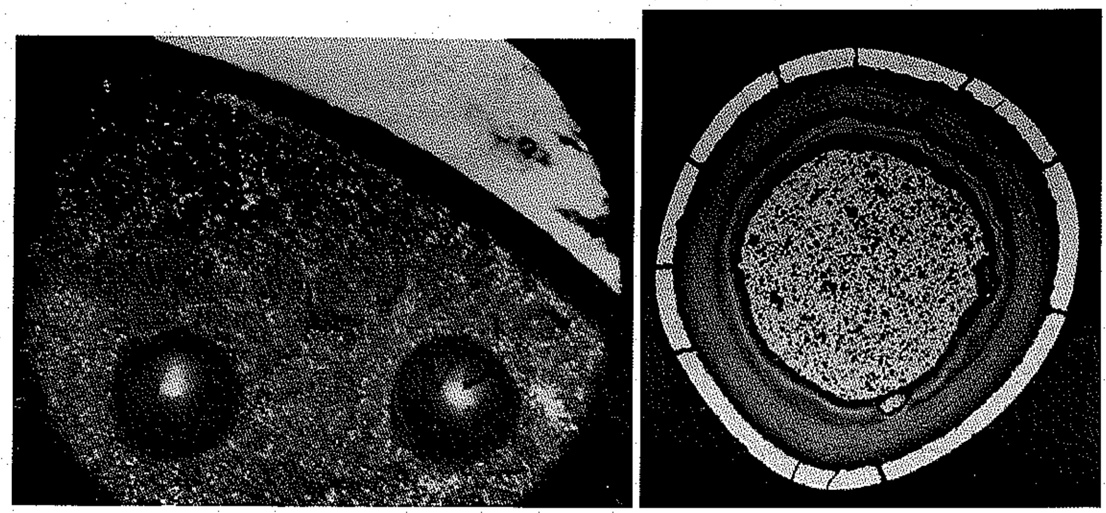


Figure 25: HFR/E96-2 experiment – 0.4% depleted particles after irradiation

3.3. DRAGON Experiments

3.3.1. DRE/MT-LET

This experiment was made of two sets of four elements known as the Matrix Test (MT) and the Low Enrichment Replacement series (LET). The two series formed a combined experiment testing of a wide range of matrix varieties over a succession of neutron doses and temperatures in two forms of coated particle compacts (cylindrical and annular).

Geometrical dimensions and deduced volume changes were measured in the temperature range of 700 to 1300°C up to a neutron dose of $3x10^{21}$ n./cm^{-2} DNE.

Annular compacts have shown a higher volume change than the cylindrical compacts (Figure 26). This fact can be explained as follows:

- The dimensional changes, deduced from the measurements performed on the compacts, consisted of permanent strains;
- These permanent strains were the combination of the densification, swelling and irradiation creep.

The enrichment and diameter of the cylindrical compacts were higher than those of annular compacts. So, it resulted that for a given temperature, the thermal gradient, and therefore the stress build-up, was higher in the cylindrical compact. The irradiation creep strains induced in the cylindrical compacts and the resulting volume increase were therefore more important and have counterbalanced to a greater extent of the compact densification.

At higher dose (longer irradiation time), when the stresses in the cylindrical compacts were sufficiently relaxed by creep, the volume change in both compact type seemed to evolve in similar way.

For each kind of compact, the combined effect of irradiation induced densification, swelling and creep was more pronounced at higher temperature.

This experiment is detailed in Raphael report n°D-FT4.3.

<u>*References:*</u>

MT and LET Series Experiment Final report
J.M.THOMSON
BN Report 7608-04, (august 1976)

Post irradiation of the DRAGON Third Matrix Test Experiment (MT3)
P.D. GOOD
P.I.D/557/1 and 3 (February 1976)

Post irradiation results from the first two of the DRAGON Matrix Test Experiments
W. KRISCHER
DPTN 573 (May 1974)

Fuel irradiation Dossier 39 Matrix Test Series MT1, MT2, MT3, MT4
Part I: Experimental Proposal (October 1971)
Part II: Design and specification (May 1972)
Part III: Pre-irradiation data (July 1972)
Part V: Interim PIE Note 2 (November 1975)

Fuel irradiation Dossier 40 Matrix Test Series LET1, LET2, LET3, LET4
Part I: Experimental Proposal (October 1971)
Part II: Design and specification (May 1972)
Part III: Pre-irradiation data (July 1972)
Part V: Interim PIE Note 1 (March 1974)
Part V: Interim PIE Note 2 (September 75)

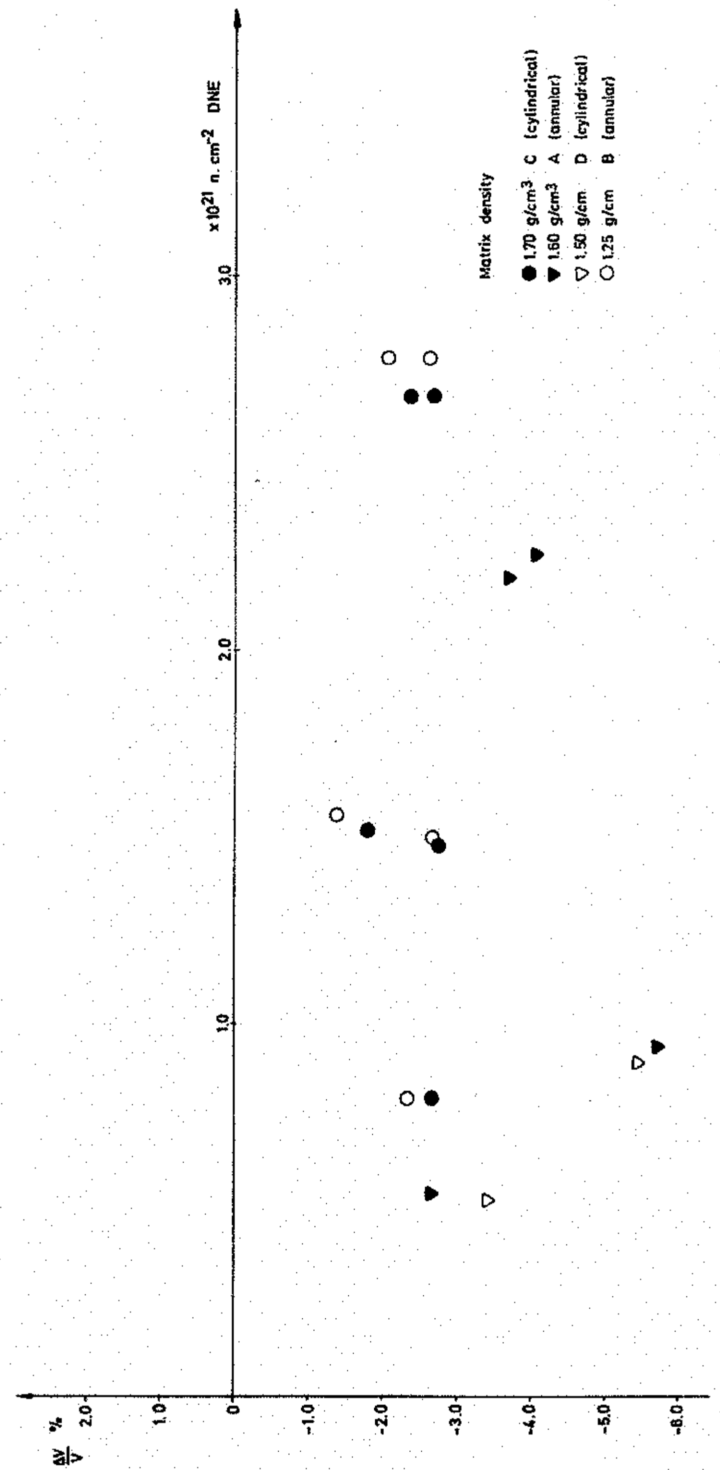


Figure 26: DRE/MT-LET - Irradiation induced volume changes of compacts (Irradiation at 1200°C)

3.3.2. DRE/Misc.Met

Coated particles with and without an external layer of overcoating have been irradiated in the Dragon reactor. The purpose was to check the behaviour of an external low density coating layer that could be utilised in conjunction with the consolidation process.

The post-irradiation examinations have shown that both types of particles behaved similarly (Figure 27), the addition of an overcoating was therefore abandoned.

This experiment is detailed in Raphael report n°D-FT4.3.

<u>*References:*</u>

Rapport final de l'irradiation de particules dans l'élément 370 du réacteur DRAGON ("Miscelleneous Metallurgical Elements")
M. GAUBE
BN Report 7308-03 (August 1973)

The loading of the Miscellaneous Metallurgical Elements
E. FORMAN, R. ROTTERDAM
DPRDD/ 1839 (June 5, 1969)

Post-irradiation examination of the BELGONUCLEAIRE particles of the fuel element 370 ("Misc.Met. Experiment)
M. GAUBE
BN Note 262.00/390/n/150 (October 1972)

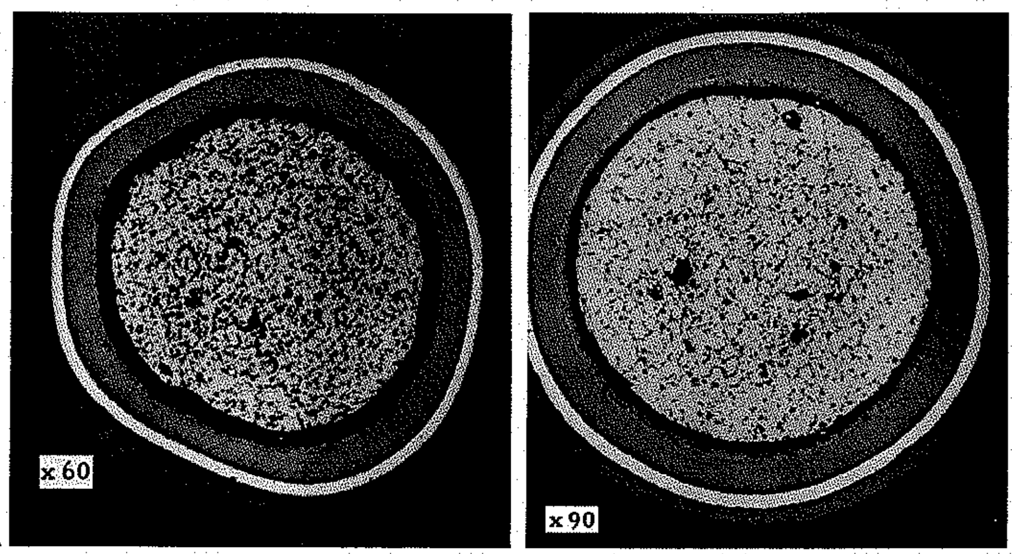
A  B

Figure 27: DRE/MiscMet - Ceramographic cross section of irradiated coated particle
- A/ with overcoating (1.5%FIMA, 1300°C)
- B/ without overcoating (1.4%FIMA, 1300°C)

3.3.3. DRE/MET V

The MET V experiment was a high temperature coated particles experiment sponsored by the Dragon project with the participation of Dragon, UKAEA, BELGONUCLEAIRE and CEA.

The BN objective was the behaviour comparison between standard uranium and plutonium loose-coated particles.

The main results are summarized below:

- Plutonium particles with diluted or concentrated kernels of low stoechiometry were irradiated at temperature ranging from 1200°C to 1500°C up to a fast neutron dose of 1.25×10^{21} n.cm^{-2} DNE and up to a burn-up of 10%FIMA. The post irradiation examinations revealed a strong effect of the irradiation temperature, 1300°C being a limit above which their performances were not anymore satisfactory. The metallography and Electron probe micro analysis have shown that the plutonium has penetrated the porous or damaged buffer to react with the inner PyC layer, modify its structure and eventually break it; the plutonium has then reacted with the SiC layer and caused the failure of the particles (Figures 28-30).

- The 3% and 9% enriched U5 particles have been irradiated in the same dose and temperature conditions but to a burnup of the order of 6.3%FIMA; they did not show any damage (Figure 31).

This experiment is detailed in Raphael report n°D-FT4.3.

<u>*References:*</u>

Metallurgical V Experiment Final Report
M. GAUBE, C. VAN LOON
BN Report 7607-04 (July 1976)

Metallurgical Experiment V – Calculation and Design
C.F. WALLROTH
DPTN 232 (Dec. 1971)

Metallurgical Experiment V – Irradiation History for charge IV, Core 5
C.F. WALLROTH
DPID 646

MET V – Irradiation History for charge IV, Core 6
C.F. WALLROTH
DPID 685

Post-irradiation examination of the BN fuel irradiated in the MET V (1) and (2) experiments
A. DRAGO
CCR ISPRA (July 1974)

Travaux effectués sur les combustibles MET V
A. SCHURENKAMPER
Technical Note C-02 EURATOM CCR ISPRA (July 1974)

Post-irradiation examination results of the BN fuel irradiated in the MET V (2) Fuel element
M. GAUBE
BN Note 262/00/100/1/187 (August 1974)

Post-irradiation Examination of MET V (3) – FE 523
P.D. GOOD
Dismantling report
PID/523/1 (Sept. 1974)

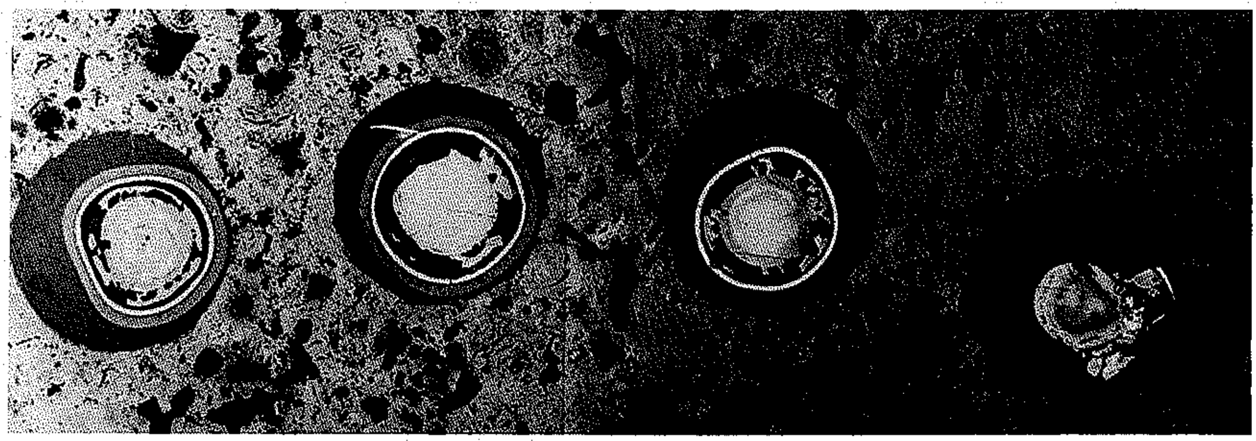
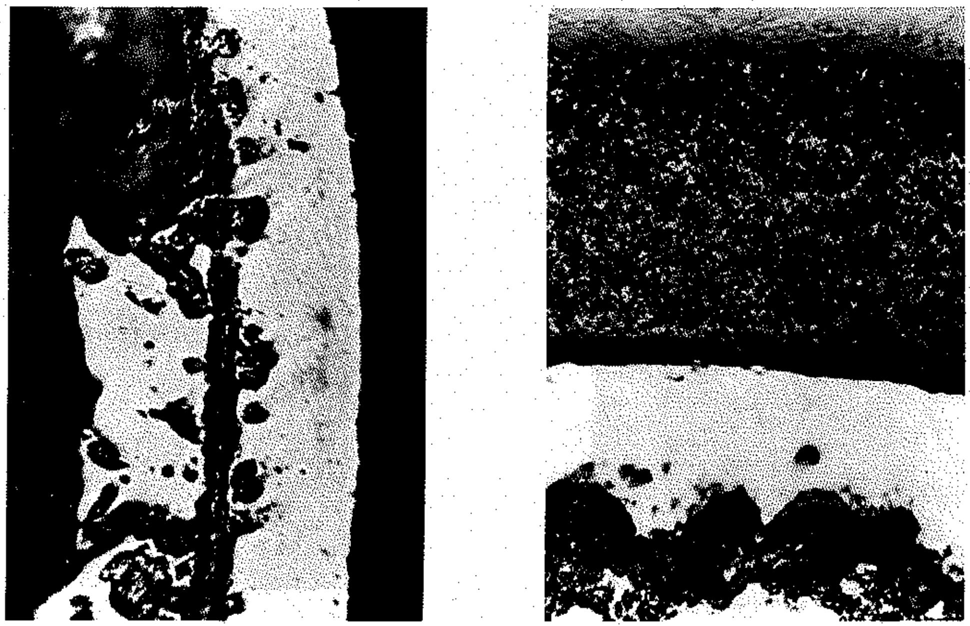


Figure 28: DRE/MET V– PuO_2 particles type T irradiated at 1400°C (58%FIMA)



PuO$_2$ type T PuO$_2$ type C

Figure 29: DRE/MET V –PuO$_2$ particles after irradiation at 1495°C (SiC attack)

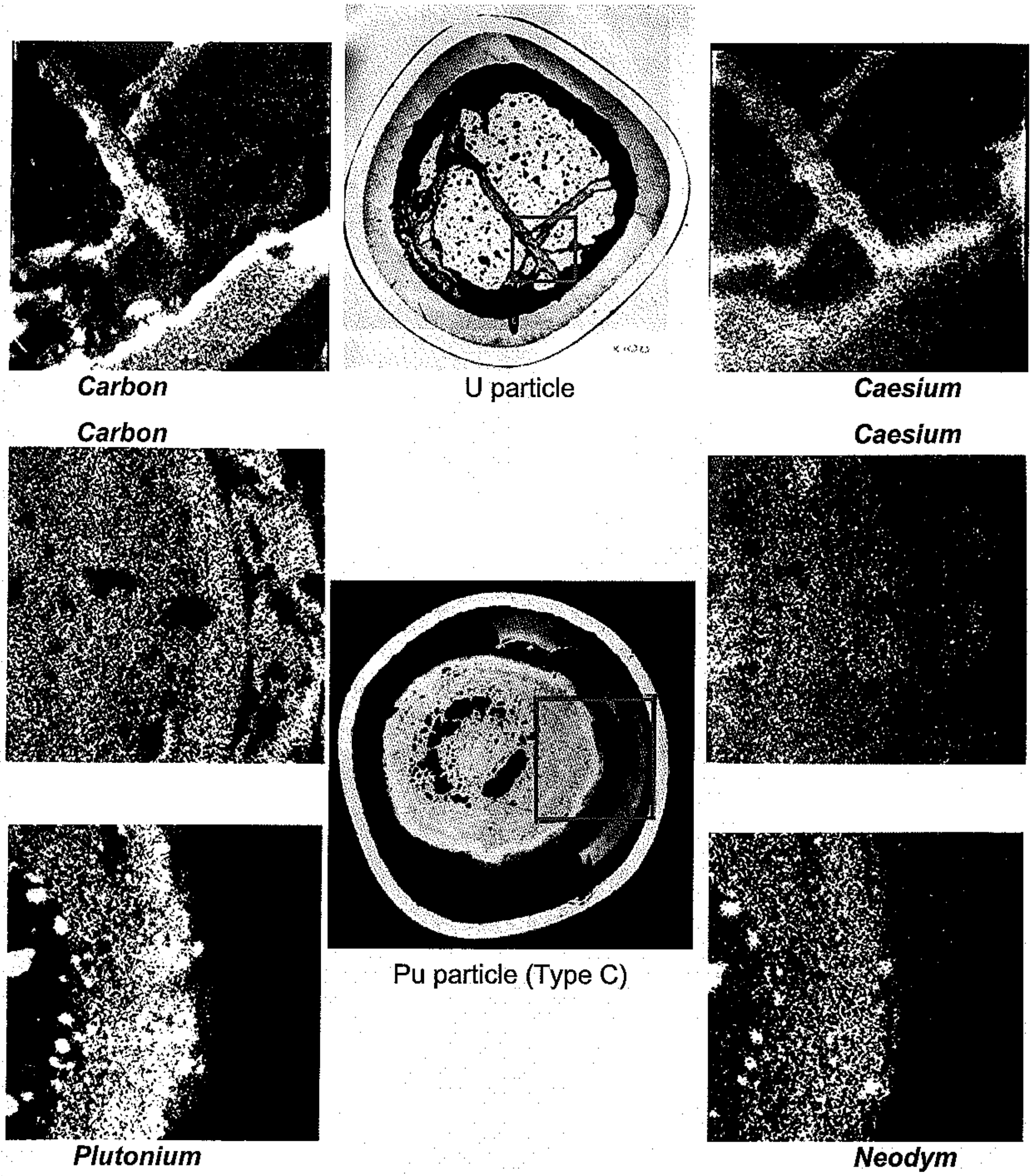


Figure 30: DRE/MET V – Microprobe analysis on irradiated U and Pu particles (irradiation at 1400 and 1300°C)

Figure 31: DRE/MET V –3% enriched Uparticle ($T_{irradiation}$=1495°C)

3.3.4. DRE/SPINES

The purpose of this experiment was to study the effect of high and low temperature irradiations on the integrity of various types of bonding material used.

54 samples covering eleven types of bonding materials (with various fractions of P514 phenolic resin, graphite powder, alcohol and solid Thor) were irradiated in three fuel elements (FE 151, 351 and 457) in the Dragon reactor at 700°C and 1050°C up to a dose of 0.66×10^{21} DNE. Particles were UO_2 kernel (757 µm diameter) with triso coating (155µm thickness).

The post irradiation examinations were performed on 47 samples and have revealed:

- a higher density of the binder at the sample periphery;
- 34 samples remained intact, 3 have lost their plug certainly during manipulation in hot cell, and 10 were broken, amongst them six have lost particles
- all the broken samples had the same binder composition:
  - P514 phenolic resin: 61%
  - Graphite powder: 30.5%
  - Alcohol: 5.5%
  - Solid Thor: -

The pre-irradiation measurements had shown that the strength of this type of matrix was poor. It was then possible that a correlation existed between the green strength and the irradiated behaviour.

- the behaviour if the particles was satisfactory (as seen in the ceramography Figure 32);
- the addition of an outer layer of porous pyrocarbon did not have any significant effect (as confirmed by the DRE/MiscMet experiment).

<u>*References:*</u>

Rapport final de l'irradiation de "bonded fuel" dans les éléments 151, 351 et 457 du réacteur Dragon ("DRE SPINES Experiment")
M. GAUBE
BN Report 7308-04 (August 1973)

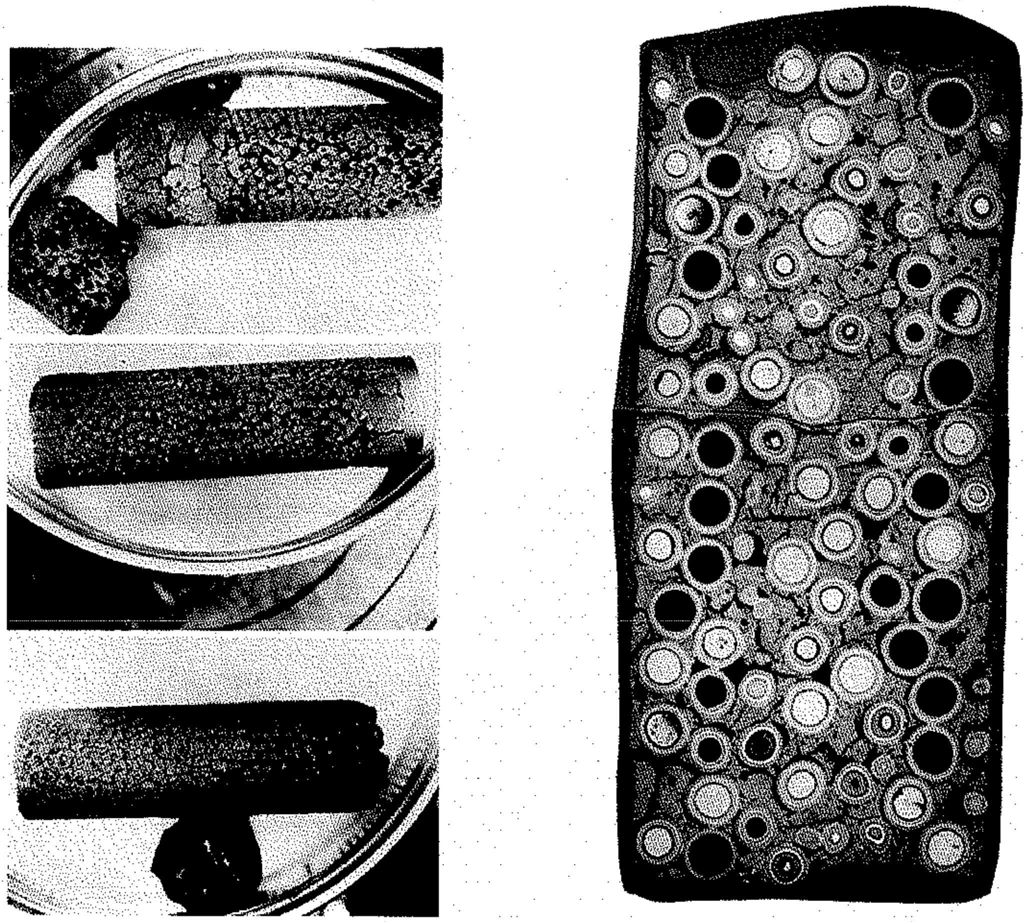
A/  B/

Figure 32: DRE-SPINES - Fuel observation
A/ Fuel bonded sticks of FE 151
B/: Cross section ceramography of sample of FE 351

3.3.5. DRE/Tubular Interacting Experiments

The Tubular Interacting (TI) design was selected in 1970 by the member companies of Inter Nuclear as the reference design following a survey assessing the thermal and mechanical performances of the different fuel element types (hollow rod, tubular interacting, teledial, and integral block, see Table 15 and Figure 33). The main advantage of the TI concept was to reduce stresses in the graphite block, since essentially no heat was transferred through them. Additional advantages were to maintain the graphite at a lower temperature, providing a larger potential heat sink in case of loss-of-coolant accident.

Figure 34 presents a view of a Tubular Interacting fuel element

Three experiments were loaded between 1970 and 1975 in the Dragon reactor. Their purpose was to study the pin behaviour in some particular circumstances, to investigate the importance of some basic parameters and to demonstrate the ability of industry to design and fabricate HTR fuel.

The annular fuel compact contained triso coated UO_2 kernels having an enrichment of approximately 9%. The fuel compacts were made using a needle coke graphite matrix powder.

Fuel characteristics and design data are described in Table 16. Kernels and coated particles are shown in Figures 35-36.

The first experiment, **INTER 1**, was a long-term experiment designed to test the TI fuel concept at the peak operating conditions expected in a commercial HTR.

The experimental channel consisted of four pins (3 of 500mm and 1 of 159 mm long), that were irradiated, in the centre channel of a D16/1 fuel carrier.

At the end of the irradiation (753 days), the peak fast neutron and the peak burn-up accumulated were respectively $3.25x10^{21}$ n./cm^{-2} DNE and 9.75% FIMA and the peak temperature exceeded 1400°C.

The post-irradiation examination are summarized below:

- The D16 carrier was in good condition;
- The surface of the graphite pins were in excellent condition although some pitting could be seen in some regions (Figure 37);
- For two pins, the fuel recovery was complicated by a strong interaction between the compacts and the outer graphite sleeve; the solid end had been cut off and the graphite sleeves had to be sacrificed.

The second experiment, **INTER 2**, was unloaded after having reached a peak fast neutron dose of $3.18x10^{21}$ n.cm^{-2} DNE and a estimated peak burn-up of 7.2%FIMA. The results of the post-irradiation examination are summarized below:

- none of the graphite pins was damaged;
- all the compacts were in excellent condition;
- no effect of the pin design (trepanned or separated inner sleeve) on the corrosion of the compacts could be identified;
- the interaction of the compacts with the outer sleeve existed but was less marked than for the INTER 1 experiment.

The four tubular interacting pins of the **INTER 3** experiment were irradiated up to a fast neutron dose of 1.38×10^{21} n./cm^{-2} DNE, a peak burn-up of 5% FIMA and at a peak temperature which exceeded 1400°C.

No significant fission products release was found during the irradiation, as confirmed by annealing test and γ-spectrometry.

The low shrinkage of the natural graphite compacts (Figure 38) reduced very much their interaction with the inner sleeve. All the components were found intact. No damage of the graphite pins by excessive stresses of the end caps or sleeves was detected and no apparent difference of behaviour between the two designs was noticed.

The destructive examinations have also shown that the behaviour of the coated particles was satisfactory.

<u>*References:*</u>

HTR fuel element design
E. SMITH
DPR 672 (October 1969)

Pre irradiation data for the first INTERNUCLEAR Tubular Interacting Experiment TI35
L. AERTS, M. GAUBE
BN report 7201-01 (January 1972)

Design of the INTERNUCLEAR Tubular Interacting Experiment TI35 in the Dragon reactor
M. GAUBE
BN Report 7012-02 (December 1970)

Design of the INTERNUCLEAR Tubular Interacting Experiment TI43
M. GAUBE
BN Report 7017-04 (July 1971)

INTERNUCLEAR TI43 Experiment 2
L. AERTS
BN Report 7107-02 (July 1971)

INTERNUCLEAR 3 (TI51) Tubular Interacting Experiment Final Report
M. GAUBE
BN Report 7408-02 (August 1974)

Type	Symbol	Characteristics
Hollow rod	HR	Rod cooled externally only – annular fuel region
Tubular	TI	Annular rods cooled from both sides
Teledial	Te	Cylindrical rods with plain fuel sticks
Integral block	BI	Plain fuel sticks, incorporated directly in the block

Table 15: Type of HTR fuel elements

Kernel	Chemical form	UO2	
	Diameter	800 µm	
	Fuel enrichment	9%	
	Density	10 g/cc	
	Porosity	8.5%	
Coating		Thickness (µm)	Density (g/cc)
	Porous Pyc	35	1
	Sealing PyC	30	1.4
	Inner HDI	30	1.8
	SiC	35	3.18
	Outer HDI	40	1.8
Compact	Graphite matrix powder	Needle coke	
	Resin	TPR 7	
	Volume loading	35%	
	Heavy Metal Loading	1 g/cc	
	Matrix density	1.5 g/cc – 1.8 g/cc	
	Outer diameter	49.mm	
	Inner diameter	35 mm	
	length	23 – 29 mm	

Table 16: DRE/TI – Particles and compacts characteristics

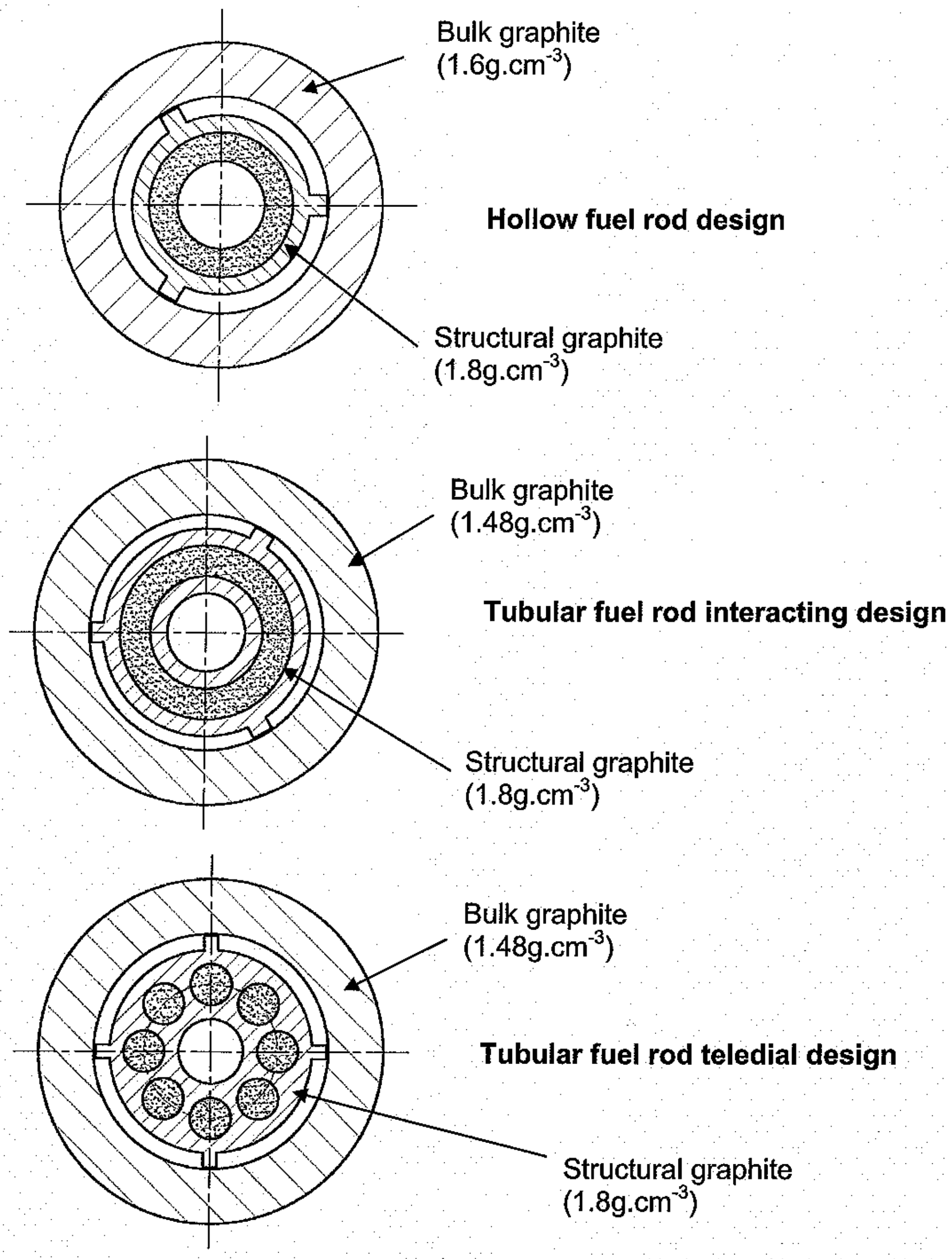


Figure 33: Design of HTR fuel elements

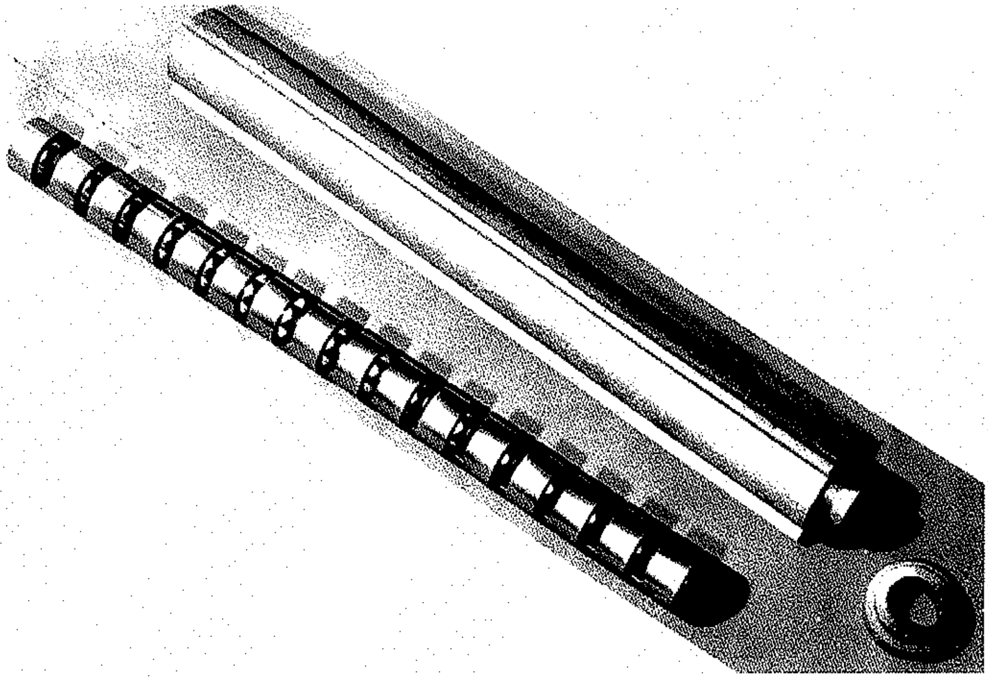


Figure 34: DRE/TI – Components of a fuel rod

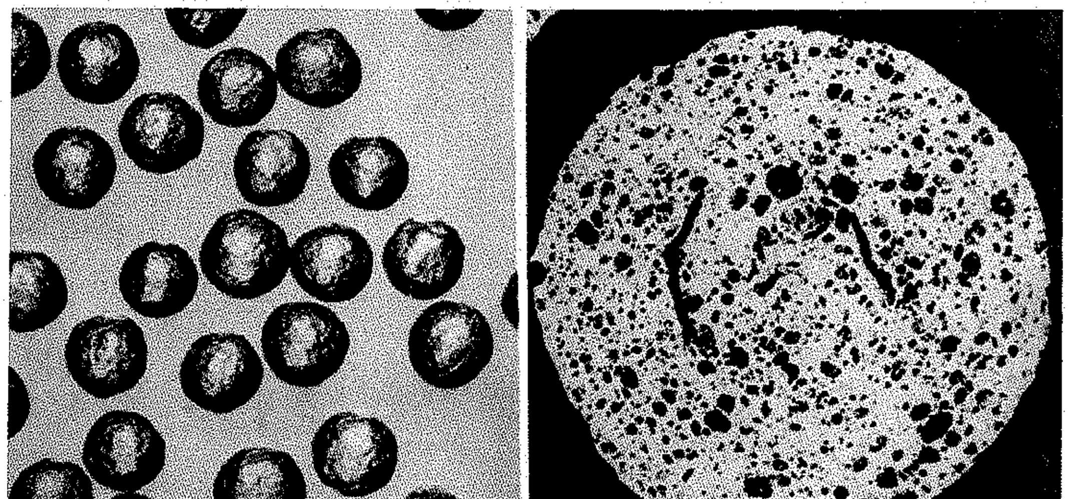


Figure 35: DRE/TI – Macroview and ceramographic cross-section of kernels

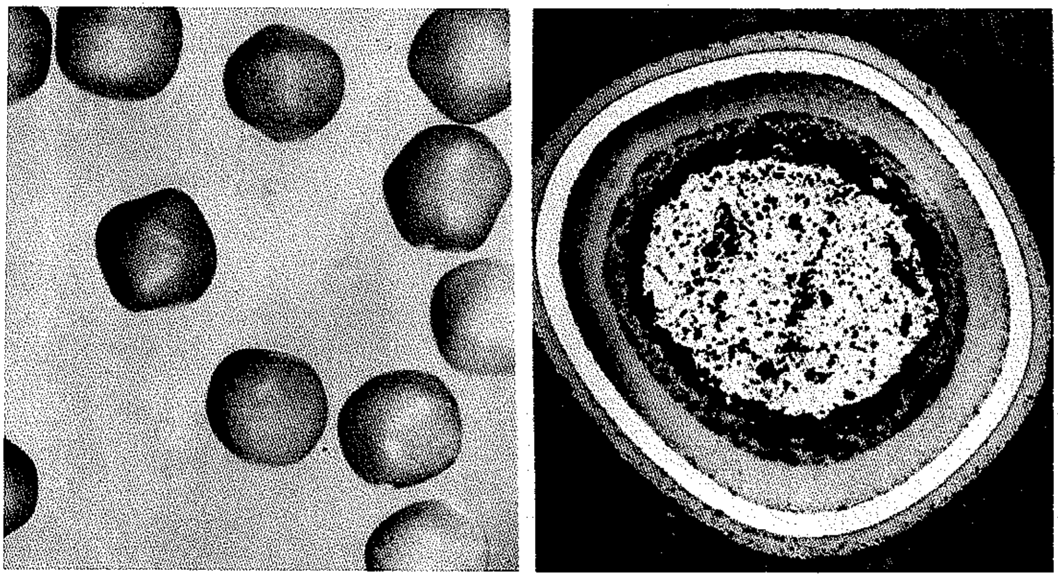


Figure 36: DRE/TI– Macroview and ceramographic cross section of coated particles

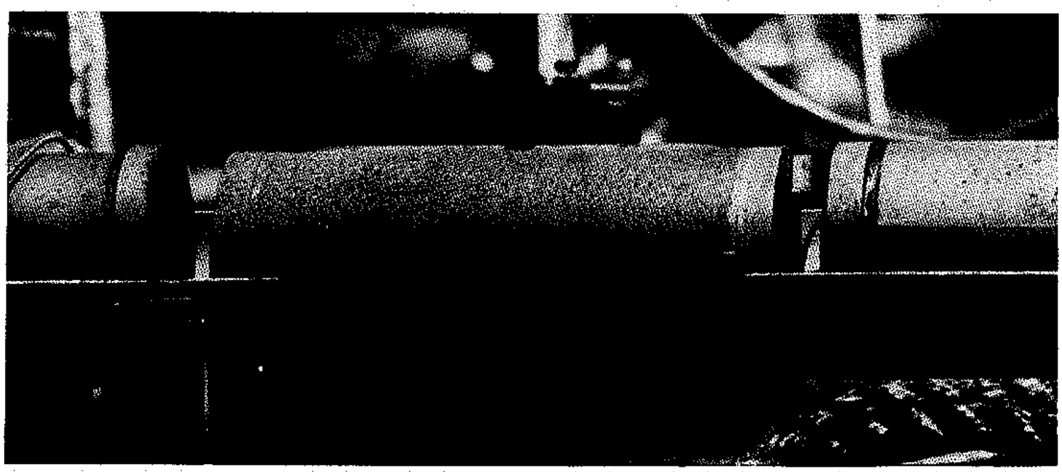


Figure 37: DRE/TI-1 – Graphite pin TI37 after irradiation

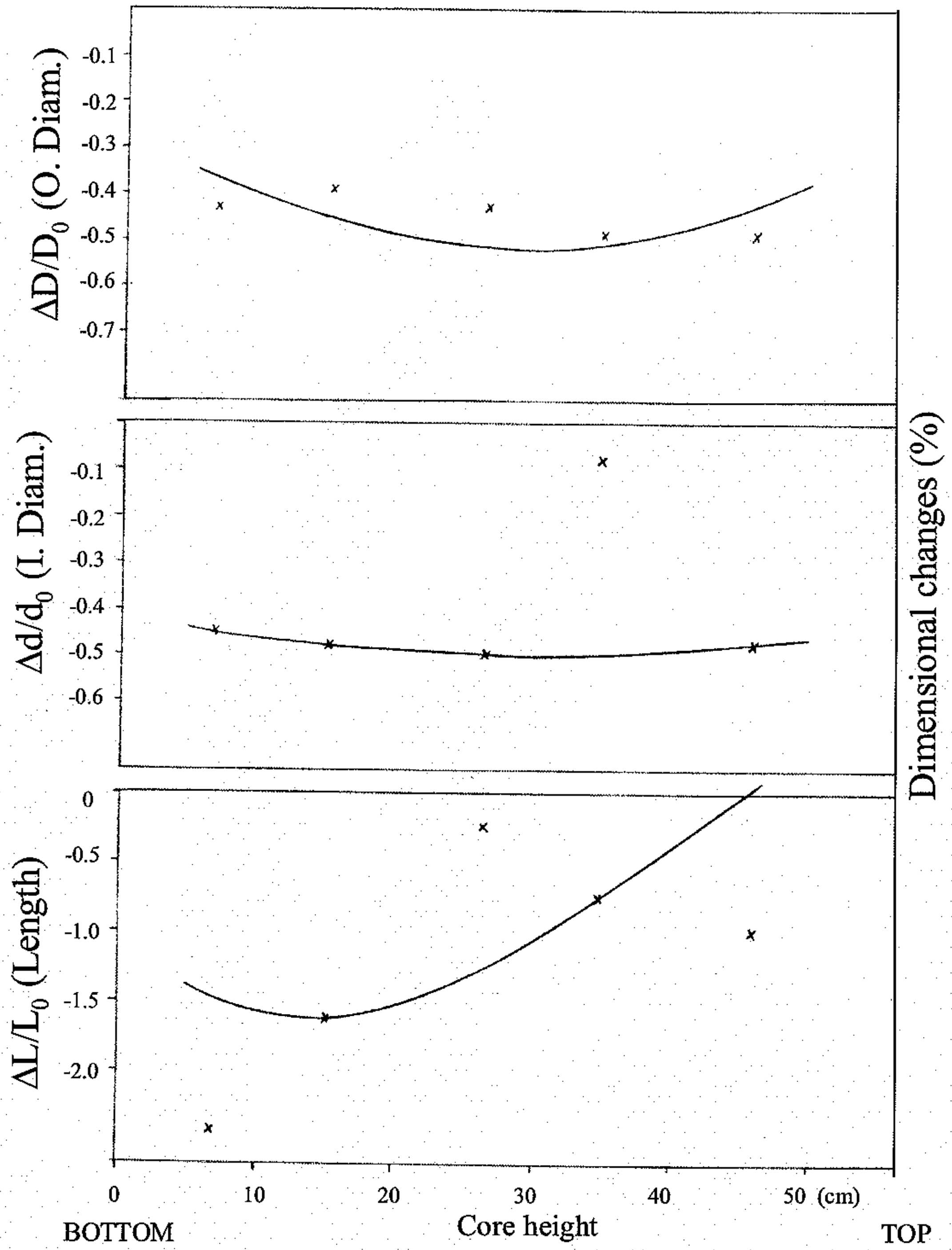


Figure 38: DRE/TI-3 – Irradiation induced dimensional changes of compact

3.3.6. DRE/MHGB

The Multi-Hole Graphite Block experiments were irradiated in the Dragon Reactor and sponsored by CEA and BELGONUCLEAIRE. A photograph of a block is shown in Figure 39.

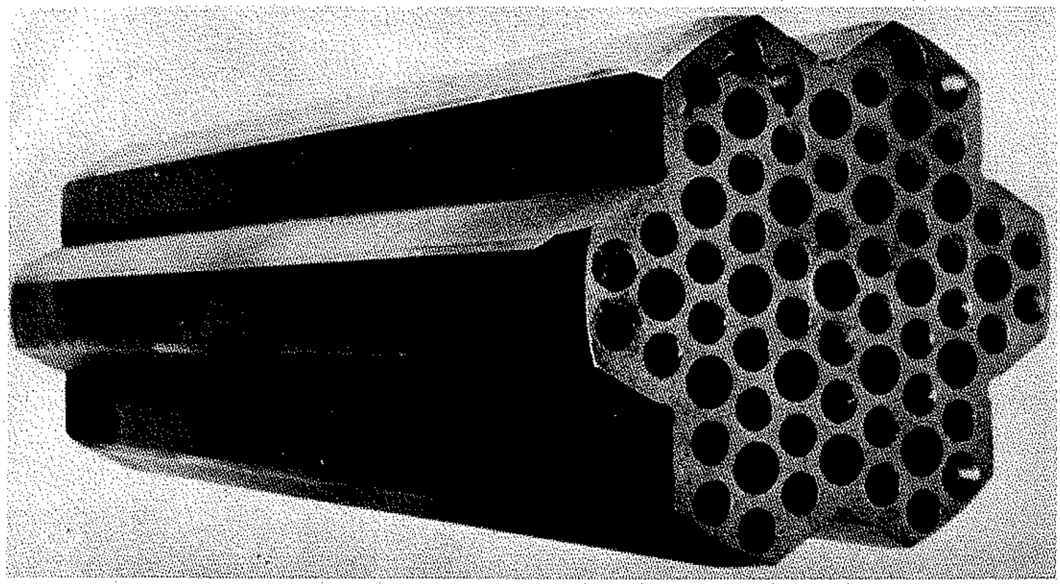


Figure 39: DRE/ view of a Multi-Hole Graphite Block

DRE/MHGB-1A

Two kinds of UO_2 coated particles were used for the fabrication of the BN compacts that were loaded in the first MHGB: triso particles with 8% U5-enriched kernels manufactured according a sol-gel process and triso particles with 7%-enriched kernels obtained by powder agglomeration. The fuel characteristics are detailed in Table 17.

The fuel element was discharged in September 1975 after having accumulated an estimated peak fast neutron dose and peak burn-up of respectively 2.4×10^{21} n.cm^{-2} DNE and 5% FIMA.

All compacts were in good conditions except the bottom compacts.

The length and outside diameter of each compact were measured; the variation are given in Table 18:

The burn-ups were calculated from the fission products inventory: mean burn-ups of 4.4% FIMA were found for channels 8B1 and 8B15 and 6.2% for channel 7D1.

DRE/MHGB-2A

For the second MHGB experiment, three fuel blocks were irradiated in the Dragon reactor, two large (740 mm) and one small (167 mm), containing each 48 fuel holes and 19 cooling channels.

The fuel, manufactured by BN, was standard driver fuel: compacts with UO_2-10C-93% enriched coated particle. Fuel characteristics are detailed in Table 19.

The experiment was irradiated in the Dragon reactor during 236 power days; the accumulated peak fast neutron dose has been estimated at 1.2×10^{21} n./cm^{-2} DNE.

The main results of the post-irradiation examination are summarized below:

- The element was in good condition, no bowing was noticed;
- Rod pattern were observed on the bottom face of the top (unfuelled) end block, on the bottom face of the top fuel block (Figure 40) and of the top face of the middle fuel block;
- The discharge of the fuel compacts from the blocks went out without difficulty;
- The surface of all the compacts was excellent (Figure 41);
- The metrology of the block did not reveal any significant dimensional changes.

<u>References:</u>

High temperature gas cooled reactor fuel
Annual Report
BN 7702-04 (February 1977)

High temperature gas cooled reactor fuel
Annual Report
BN 7602-07 (February 1976)

Multihole Graphite Block experiment MHGB-2A
M. Gaube
Fuel loading Report 260.00/390/n/54 (December 1973)

Kernels		
Enrichment	7.053%	8.116%
Process	powder agglomeration	sol-gel
Diameter (mm)	790 ±41	793 ±25
Density	87.1 %TD	88.2 %TD
O/U	2.00	2.01
Mean weight of UO_2 (mg)	2.464	2.534
Mean weight of U (mg)	2.166	2.230
Coated particles		
Overall density (Hg) (g/cc)	4.51	4.50
Overall size (µm)	1147 ±42	1149 ±21
Kernel diameter (µm)	793 ±41	782 ±25
Buffer Thickness (µm)	30 - 35	30 - 35
Density (g/cc)	1.00 – 1.10	1.00 – 1.10
Sealing Thickness (µm)	25 - 30	25 - 30
Density (g/cc)	1.4 – 1.6	1.4 – 1.6
Inner PyC Thickness (µm)	30 - 35	30 - 35
Density (g/cc)	1.75	1.75
SiC Thickness (µm)	36 ±2	33 ±3
Density (g/cc)	3.19	3.19
Crack barrier (µm)	2	2
Outer PyC Thickness (µm)	35 ±3	38 ±3
Density (g/cc)	1.76	1.75
Total coating thickness (µm)	177 ±14	185 ±9
Compacts		
Weight of individual compact (g)	5.72	9.37
Outer diameter (mm)	12.51	12.52
Overall length (mm)	27.84	33.7
Matrix* density (g/cc)	1.54 ±2	1.54 ±2
Weight of U / compact (g)	0.4099	2.767
Weight of U235 / compact (g)	0.0289	0.2214
Number of CP / compact	192	1241
Volume loading (%)	4.18	23.6

*Carbon Lorraine natural graphite powder (14%resin 1% stearic acid)

Table 17: DRE/MHGB-1A - BN Fuel characteristics

	7% enriched fuel	8% enriched fuel	
	Channel 7D1	Channel 8B1	Channel 8B15
ΔL/L	-0.64%	-1.14%	-1.04%
ΔD/D	-1.42%	-1.12%	-1.16%

Table 18: DRE/MHGB-1A– Compact dimensional change

Kernels	
Composition	UO_2-10C
C/U	9.86 ±0.2
Enrichment U5	93.0 ±0.9 %
O/U	2.04
Diameter (mm)	583 ±411
Density (Hg)	3.24 ±0.19
Porosity	28.3 ±4.3
Mean weight (µg)	334 ±39
Coated particles	
Overall size (µm)	910 ±40
Kernel diameter (µm)	793 ±41
Buffer Thickness (µm)	25
Density (g/cc)	1.0 ± 0.1
Sealing Thickness (µm)	20
Inner PyC Thickness (µm)	30
Density (g/cc)	1.7
SiC Thickness (µm)	32 ±3
Density (g/cc)	3.2
Outer PyC Thickness (µm)	42 ±6
Density (g/cc)	1.7
Total coating thickness (µm)	155 ±20
Compacts	
Outer diameter (mm)	12.48
Overall length (mm)	28.3
Matrix* density (g/cc)	1.55 – 1.65
Weight of U235 / compact (g)	0.039

*: natural graphite, degassed at 1800°C

Table 19: DRE/MHGB-2A Experiment – BN Fuel characteristics

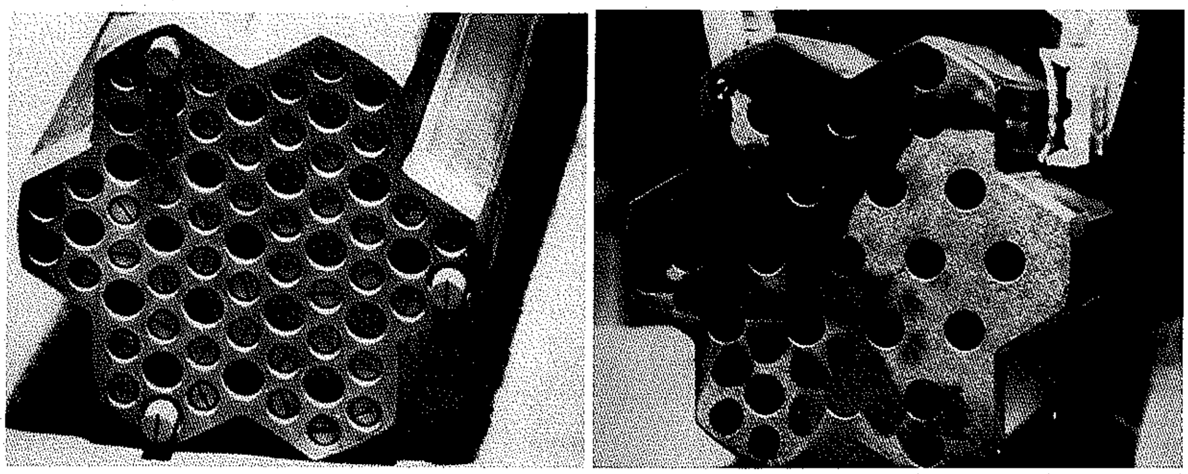


Figure 40: DRE/MHGB-2A – View of fuel blocks end faces

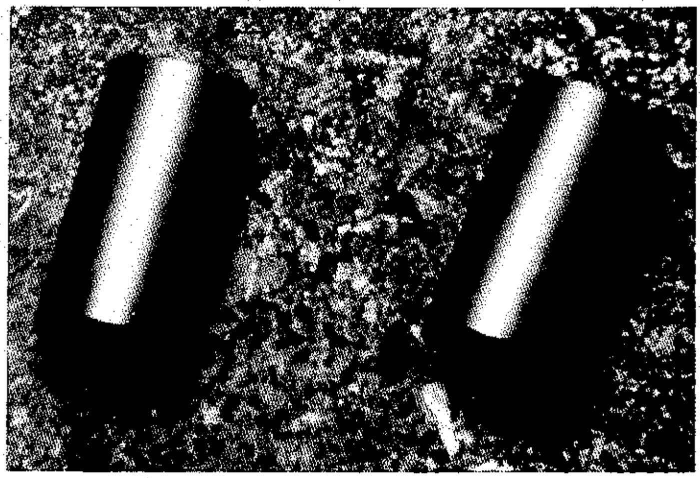


Figure 41: DRE/MHGB-2A – Compacts after irradiation

3.4. BR3 Experiment

BR3/2bis

The purpose of this experiment was the study of the behaviour under irradiation of different matrix material with or without UO_2 coated particles. The compacts were loaded in the BR3 reactor in the temperature range 300-1200°C up to a dose of $2x10^{21}$ $n.cm^{-2}$ DNE and about 7%FIMA (for the 12% enriched particles).

The behaviour of matrix materials and particles was satisfactory; no amoeba effect was noticed. The cracks that were observed along the coatings layers were attributed to the fabrication process (Figure 42).

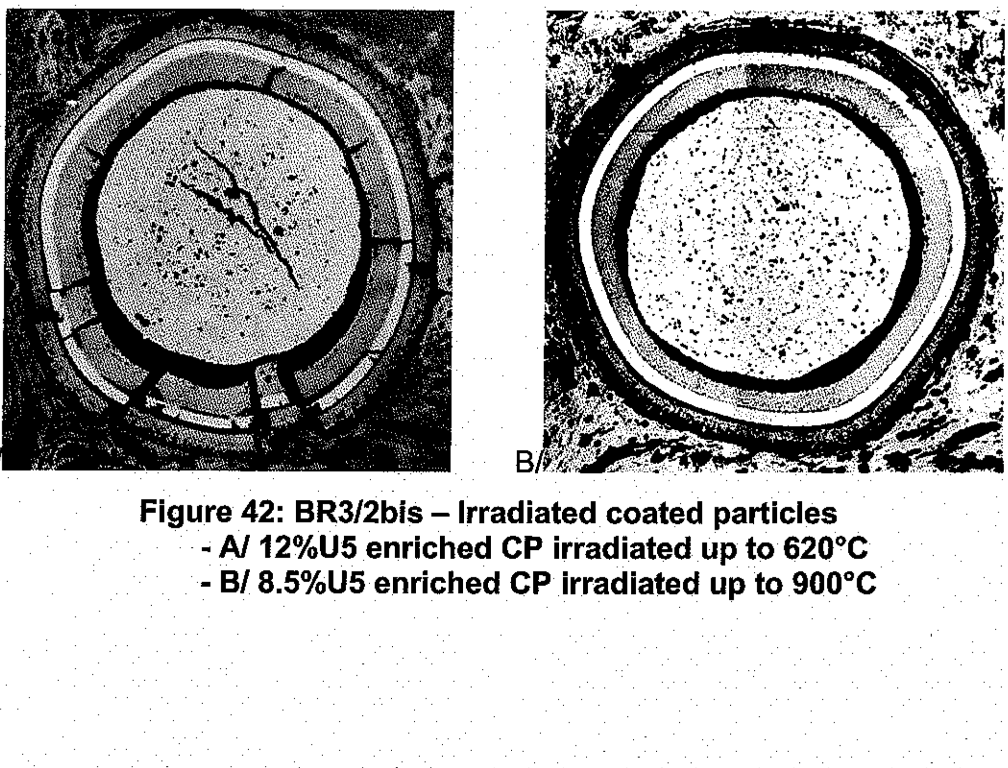
This experiment is detailed in Raphael report n°D-FT4.3.

<u>References:</u>

Rapport final de l'irradiation de combustible HTR dans la cellule n°80 du coeur BR3/2bis
M.GAUBE, J.M. THOMSON
BN Report 7312-05 (December 1973)

Examens des particules recouvertes irradiées dans le tube modérateur de la cellule 80 du BR3/2bis
C. VAN LOON
Note CEN, TEC/43.8790/17/CVL/cc

Examens postirradiatoires de la cellule HTR 80 du réacteur BR3/2bis
R. NAUCHE, A. TERGAZHI
Note CEN, Dossier GEX n° 8790, TEC/43.8790/28/RN/FD



A/ B/

Figure 42: BR3/2bis – Irradiated coated particles
- A/ 12%U5 enriched CP irradiated up to 620°C
- B/ 8.5%U5 enriched CP irradiated up to 900°C

3.5. ISPRA Experiment

ISPRA-1

The purpose of this experiment was the investigation of the fission products release from annular compacts with given fraction of bare kernels.

By assuming that the release derives exclusively from defective particle, the R/B per particle (*$(R/B)_p$*) was calculated by dividing the R/B values by the fraction of defective particles (i.e. bare kernel).

The results can be summarized as follows:

- the release rate per particle is of order of 1% at 1130°C for Kr85 and slightly higher for Xe133 (6.8%)
- the *$(R/B)_p$* is temperature dependent according to the following equation

$$(R/B)_p = (R/B)_p^0 \exp^{-\frac{Q}{RT}},$$

an activation energy Q of 25 kcal/mole.°C was determined (Figure 43).

References:

Irradiation dans le réacteur ISPRA1 de compacts avec des fractions connues de particules non recouvertes
M. GAUBE
BN Report 7401-03 (January 1974)

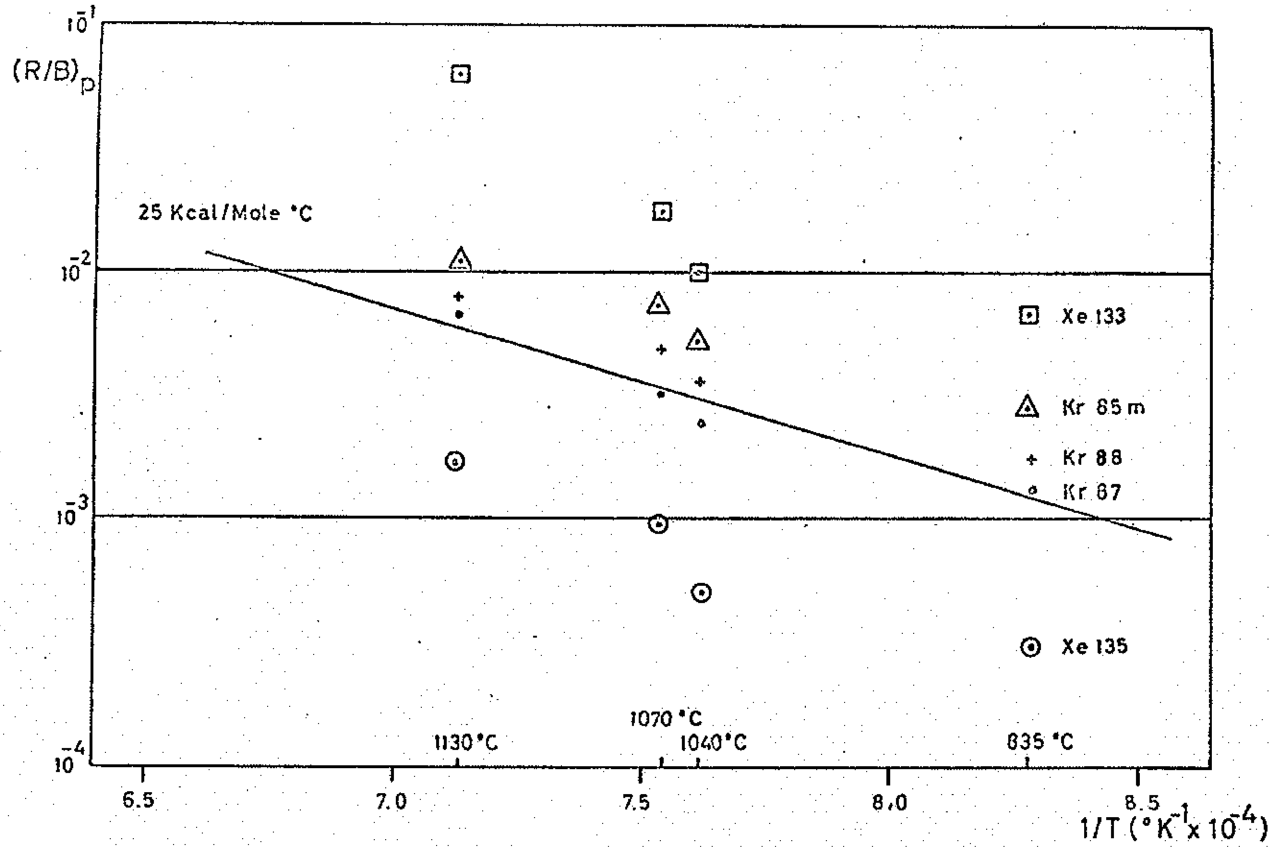


Figure 43: ISPRA-1– Temperature dependence of $(R/B)_{particle}$