European Appl. Res. Rept.—Nucl. Sci. Technol.
Vol. 2, No. 1 (1980), pp. 129–177
0379-4229/80/0201-0129$9.40/0
Printed in France

POST IRRADIATION ANALYSIS OF THE OBRIGHEIM PWR SPENT FUEL

P. BARBERO[1], G. BIDOGLIO[1], A. CALDIROLI[2], F. DANIELE[2], R. DE MEESTER[3], R. ERNSTBERGER[3], S. FACCHETTI[1], A. FRIGO[5], S. GUARDINI[4], G. GUZZI[1], P. HANSEN[2], L. LEZZOLI[1], L. KOCH[3], W. KONRAD[2], L. MAMMARELLA[1], F. MANNONE[1], A. MARELL[1], A. SCHURENKAMPER[5], P.R. TRINCHERINI[1], and R. WELLUM[3]

Commission of the European Communities, Joint Research Centre Ispra Establishment, Italy

[1]*Chemistry Division,* [2]*ESSOR Division,* [3]*Transuranium Institute (Karlsruhe),* [4]*Physics Division,* [5]*Materials Division*

EUR 6589 EN

POST-IRRADIATION ANALYSIS OF THE OBRIGHEIM PWR SPENT FUEL

1. INTRODUCTION AND SUMMARY

The cooperation Agreement N. 139-75 PIPG D among the Kernkraftwerk Obrigheim GmbH (KWO), the Kraftwerk Union AG Erlangen (KWU) and the Commission of the European Communities was set up with the aim of performing post-irradiation analyses of fuel rods taken from the fuel assemblies BE 124 and BE 210, unloaded from the Obrigheim reactor at the end of the fifth irradiation cycle. The programme was carried out at the Joint Research Centre, Ispra and Karlsruhe Establishments, within the framework of the "Bench Mark Experiment" activity.
The objective was the measurement of the burnup and isotopic composition of selected fuel samples in order to obtain a set of reference data to be used for checking the accuracy of nuclear code calculations.

The fuel rods were subjected to the following procedures

- gamma scanning examinations carried out at the ADECO laboratory of the Ispra Establishment, to determine the proper cutting positions,
- fuel samples chosen for radiochemical analyses were cut from the rods at different levels of the fuel stack,
- gamma spectrometry measurements on the selected samples performed at the LMA Laboratory to determine the fission product activity.

The samples were then dissolved in the Laboratories of Ispra and Karlsruhe and aliquots of the solutions were subjected to radiochemical processes and to gamma, mass and alpha spectrometry determinations.

Gamma spectrometry was mainly used to determine the ^{137}Cs activity from which the burnup was derived.

Mass spectrometry, combined with isotope dilution technique, was used to determine the concentrations and/or the isotopic compositions of uranium, plutonium, americium, neodymium and of the krypton and xenon fission gases. The concentration of ^{148}Nd was used for the evaluation of the burnup.
Alpha spectrometry was carried out to determine the concentrations of some nuclides of plutonium, americium and curium.
In section 3.1. all the original data of the gamma, mass and alpha determinations are reported. These data were processed in order to derive values of burnup and of isotopic composition, and of build up and depletion of heavy isotopes, which are reported in section 3.2. The nuclear data used in the processing of experimental values are also listed in this section.
The availability of the original experimental data, reported in section 3.1., also allows application of data processing based on different nuclear data or different assumptions.

In order to check the accuracy of the values of burnup and isotopic compositions measured in our experiments three procedures were applied :

- use of different methods in the measurement of the same quantity : this procedure was applied for the burnup values which were determined both from ^{137}Cs and ^{148}Nd
- comparison between the results of different laboratories : this procedure was applied by analysing 4 pairs of adjacent pellets in the Laboratories of Ispra and Karlsruhe
- use of the isotopic correlation technique.

Similar programmes of post-irradiation examinations had been carried out on the fuel from Garigliano and Trino Vercellese reactors, operated by Ente Nazionale per la

Energia Elettrica (ENEL) [1,2,3] and on the fuel from the Gundremmingen reactor, operated by Kernkraftwerk RWE-Bayernwerk GmbH (KRB) [4].

2. FUEL CHARACTERISTICS AND SELECTION OF THE FUEL SAMPLES

The Obrigheim Nuclear Power Plant, operated by KWO, is equipped with a pressurised water reactor rated at 350 MW (e). Six fuel rods were chosen for examination, selected from :

- fuel element BE 124 whose irradiation started on the 30th September 1970 and finished on the 16th August 1974,
- fuel element BE 210 whose irradiation started on the 30th September 1971 and finished on the 16th August 1974.

In Table IA the irradiation history of these two fuel elements is given.
The fuel assemblies BE 124 (average burnup of 29,000 MWD/MTU) and BE 210 (average burnup of 30,000 MWD/MTU) selected for post-irradiation analyses were composed of 180 rods with initial enrichment of 3.00 wt% in ^{235}U and 2.83 wt% in ^{235}U, respectively.
In Table IB the main characteristics of the fuel assembly are presented.
Table II indicates the locations of the fuel pellets analysed, the Laboratories (Ispra and Karlsruhe) where the analyses were carried out, the neutron spectrum in which the fuel rods were irradiated and the measured burnup values for each fuel section.
Schematic maps of the reactor core indicating the positions of the selected fuel assemblies during the various irradiation cycles are given in Figure 1.

CYCLE OF OPERATION	PERIODS	DAYS (a)	BURNUP (MWD/MTU)			
			POSITION	BE 124	POSITION	BE 210
SECOND	30.09.70 12.08.71	258	G-1	6,600		
Shut-down	13.08.71 29.09.71	48				
THIRD	30.09.71 07.09.72	295			D-11	9,900
Shut-down	08.09.72 04.10.72	27				
FOURTH	05.10.72 01.09.73	283	D-7	18,600	J-5	21,300
Shut-down	02.09.73 24.09.73	23				
FIFTH	25.09.73 16.08.74	229	D-4	29,000	G-3	30,100

(a) Full power days only.

TABLE IA - Irradiation History of the Obrigheim Fuel Elements BE 124 and BE 210

CORE	
Number of fuel assemblies	121
First charge enrichment (wt% 235U)	2.5-2.8-3.1
Number of control rod clusters	32
UO_2 in square fuel assemblies (Kg)	39,930
Total U weight (Kg)	35,200
SQUARE FUEL ASSEMBLY	
Number of fuel rods	180 (14x14-16)
Rod pitch (cm)	1.43
Side of square fuel section (cm)	20.0
Total lenght (cm)	317.0
Channel material	Zircaloy-4
FUEL PELLET	
UO_2 linear density (g/cm):	
Fuel element BE 124	6.68
Fuel element BE 210	6.52
Diameter (cm)	0.904
Lenght of pellet stack in fuel (cm)	295.6

FUEL CLAD	
Outside diameter (cm)	1.076
Inside diameter (cm)	0.9318
Wall thickness (cm)	0.0721
Material	Zircaloy-4
CONTROL ROD	
Absorbing material	Ag-In-Cd
Canning material	stainless steel
Volume of guide tube positions and water gaps per fuel rod cell (cm^2)	0.20173
MODERATION AND COOLING	
Inlet temperature (°C)	283
Outlet temperature (°C)	313
Coolant pressure (bar)	145
Coolant flow rate (t/h)	22,000

TABLE IB - Obrigheim Reactor Characteristics

TABLE II : SELECTED FUEL SAMPLES

AXIAL LOCATION	FUEL ELEMENT BE 124								FUEL ELEMENT BE 210			
	I	K	I	K	I	K	I	K	I	K	I	K
TOP												
P5			22,800			25,800						
P5(2)									24,200			
P5(1)									30,100			
P4			30,900		27,700			24,900				
P4(1)									35,600			32,900
P3	33,700		36,200		31,300		29,400					
P3(1)									37,500		36,600	
P2	28,000 (*)			35,100	25,800							
P1		21,200	20,200		17,100			15,600			25,500	
BOTTOM												
RODS	D1		E3		G7		M14		G14		K14	
NEUTRON SPECTRUM	PERTURBED (CLOSE TO THE ASSEMBLY'S EDGE)		INTERMEDIATE (NEAR WATER HOLE)		ASYMPTOTIC		PERTURBED (CLOSE TO THE ASSEMBLY'S EDGE)		PERTURBED (CLOSE TO THE ASSEMBLY'S EDGE)			

I. samples analyzed at Ispra K. samples analyzed at Karlsruhe

Experimental burnup value is indicated for each sample in MWD/MTU

(*) Value obtained by ^{137}Cs non-destructive determination.

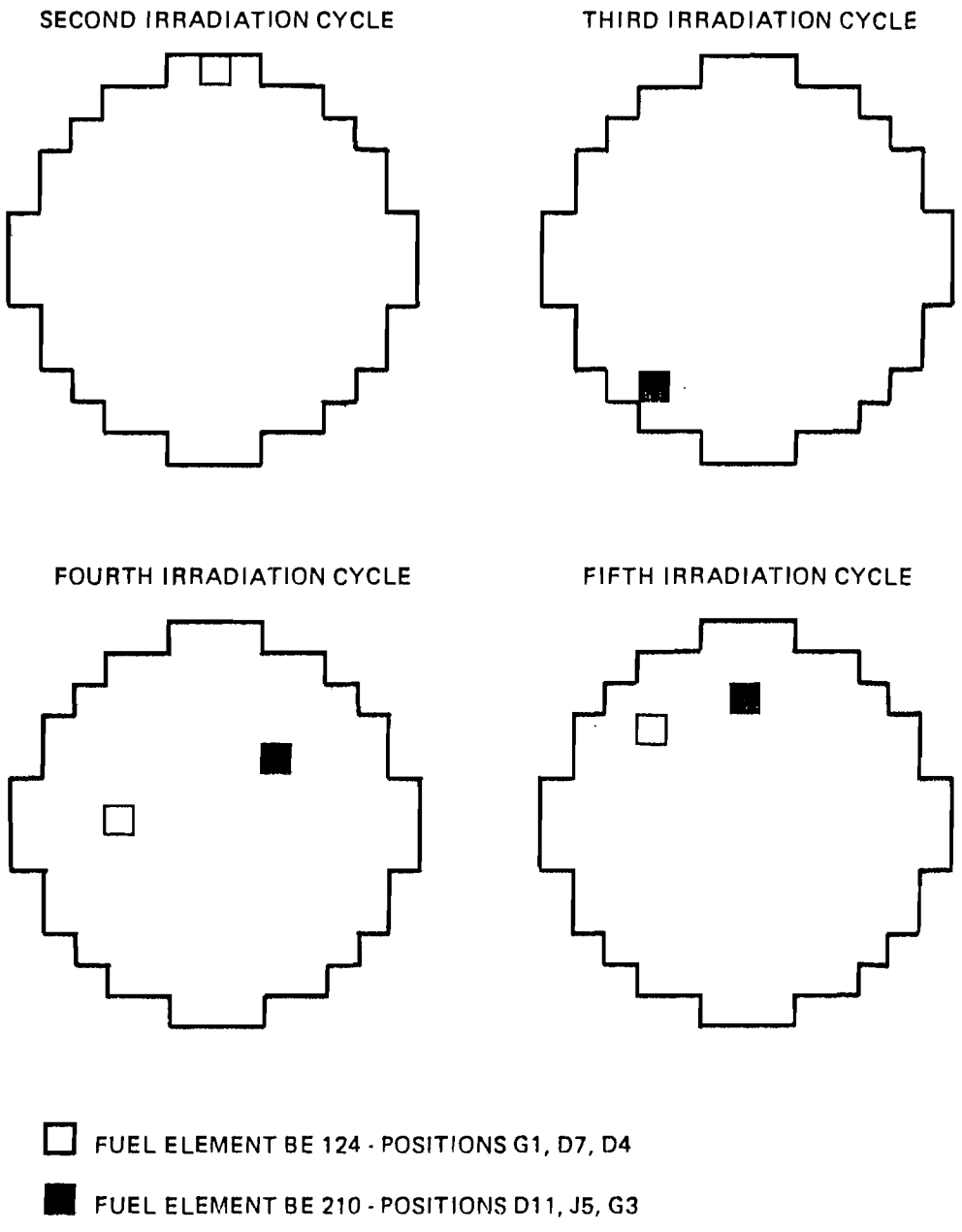


FIG. 1 : SCHEMATIC CORE MAPS OF THE OBRIGHEIM REACTOR DURING DIFFERENT IRRADIATION CYCLES.
THE POSITION OF THE ELEMENTS BE 124 AND BE 210 ARE INDICATED.

Figure 2 shows the locations in the assemblies of the fuel rods selected for the measurements, and their cutting positions.

3. DETERMINATION OF BURNUP AND ISOTOPIC COMPOSITIONS

3.1. Experimental Procedures

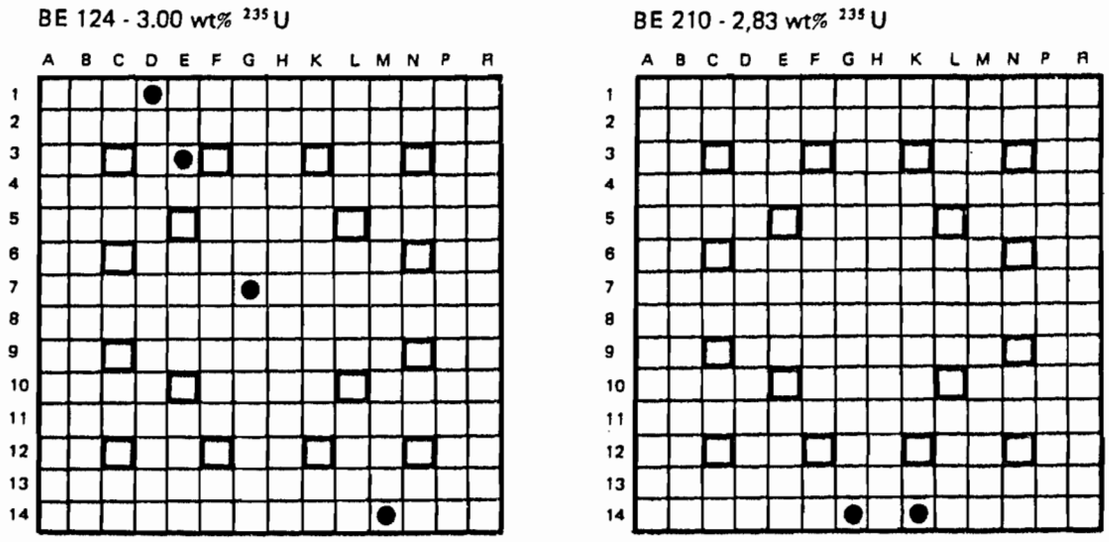
A total of six fuel rods belonging to both BE 124 and BE 210 assemblies were subjected to non-destructive gamma spectrometry measurements in the LMA laboratory to determine the ^{137}Cs activity.

In the selected positions (see Fig. 2) four rods of the fuel assembly BE 124 and two rods of the fuel assembly BE 210 were cut and samples, 10 mm thick, were prepared for radio chemical analysis. The cutting positions were determined with an uncertainty of $\pm$ 3 mm.
In the Karlsruhe Laboratory fission gases were collected during fuel sample dissolution.
The analyses were performed at Ispra (17 samples) and at Karlsruhe (6 samples plus 4 for cross-checking).
Radioactive fission products were determined by gamma spectrometry. The isotopic compositions and concentrations of U, Pu, Nd and Am were determined by isotopic dilution and mass spectrometry as well as the isotopic composition of the fission gases krypton and xenon. ^{238}Pu, ^{241}Am, ^{242}Cm and ^{244}Cm were determined by alpha spectrometry.
The experimental procedures adopted at Ispra and Karlsruhe are described in detail in Ref. 1, 2, 3, 4 and 5.

3.1.1. Gamma scanning

A gross total gamma scanning of the fuel rods was carried out in the ADECO Laboratory before they were cut.

OBRIGHEIM REACTOR FUEL ASSEMBLIES



CUTTING POSITIONS (mm)

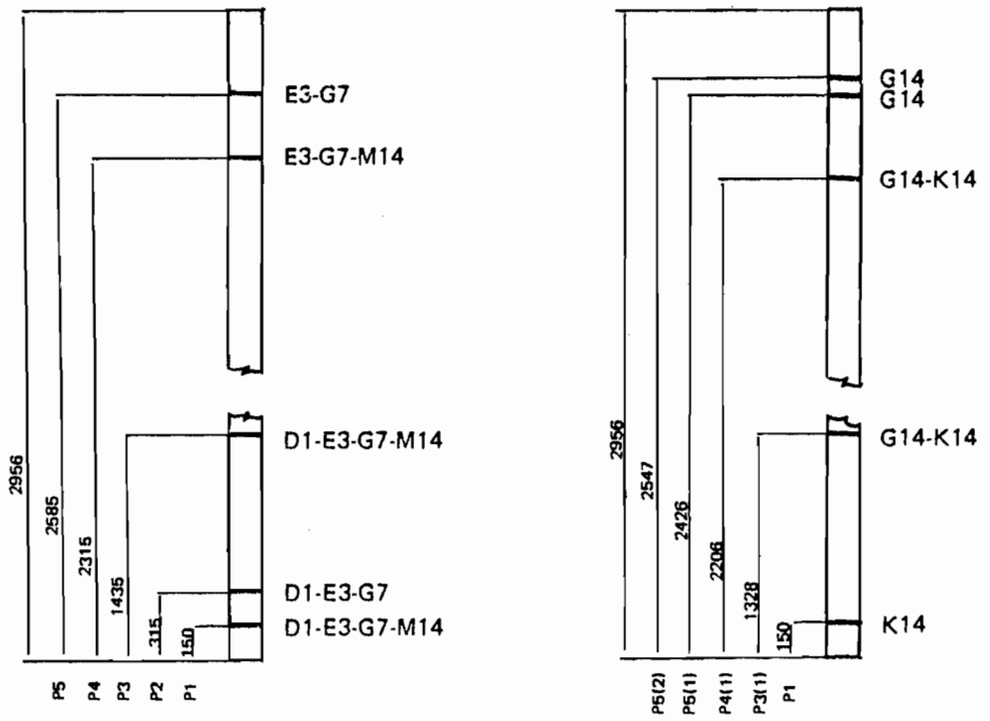


FIG. 2: CUTTING POSITIONS OF THE OBRIGHEIM REACTOR FUEL SAMPLES SELECTED FOR ANALYSIS

◉ SELECTED ROD

The equipment consisted of a Ge(Li) detector having a FWHM of 2.3 kev at 1332 kev of ^{60}Co, connected to an Intertechnique Plurimat Multi 8 processor.
A lead collimator with an aperture of 0.5x10x284 mm was used. The rods are moved axially by a stepping motor. Steps of 0.2 mm each, are set up but measurements have been done every 2 steps (0.4 mm).
From the distribution curve of gamma activity obtained, it was possible to determine the length of the rods and consequently the cutting levels, with a precision given by the scanning step i.e. 0.4 mm.
Figure 3 shows the results for rod G7 of the BE 124 fuel element. In the same figure the cutting positions are also indicated by arrows.

3.1.2. Gamma spectrometry

Gamma spectrometry measurements were carried out on the solid fuel pellets to determine ^{137}Cs activity and on the fuel solutions to determine the activities of ^{137}Cs, ^{134}Cs and ^{154}Eu.
Non-destructive and destructive measurements of ^{137}Cs were used to evaluate the burnup of the fuel samples. The measurements of ^{134}Cs and ^{154}Eu were used to check the other experimental determinations (burnup and isotopic compositions) by means of the isotope correlation technique.

a) The measurements on the solid fuel pellets were carried out with a coaxial type Ge(Li) detector having a FWHM of 2.6 kev at 1332 kev of ^{60}Co, connected to an Intertechnique PLURIMAT 20 processor. They were performed on 25 fuel pellets cut adjacent to the ones used for destructive determinations. The standard deviation of the measurements was 2.0%.

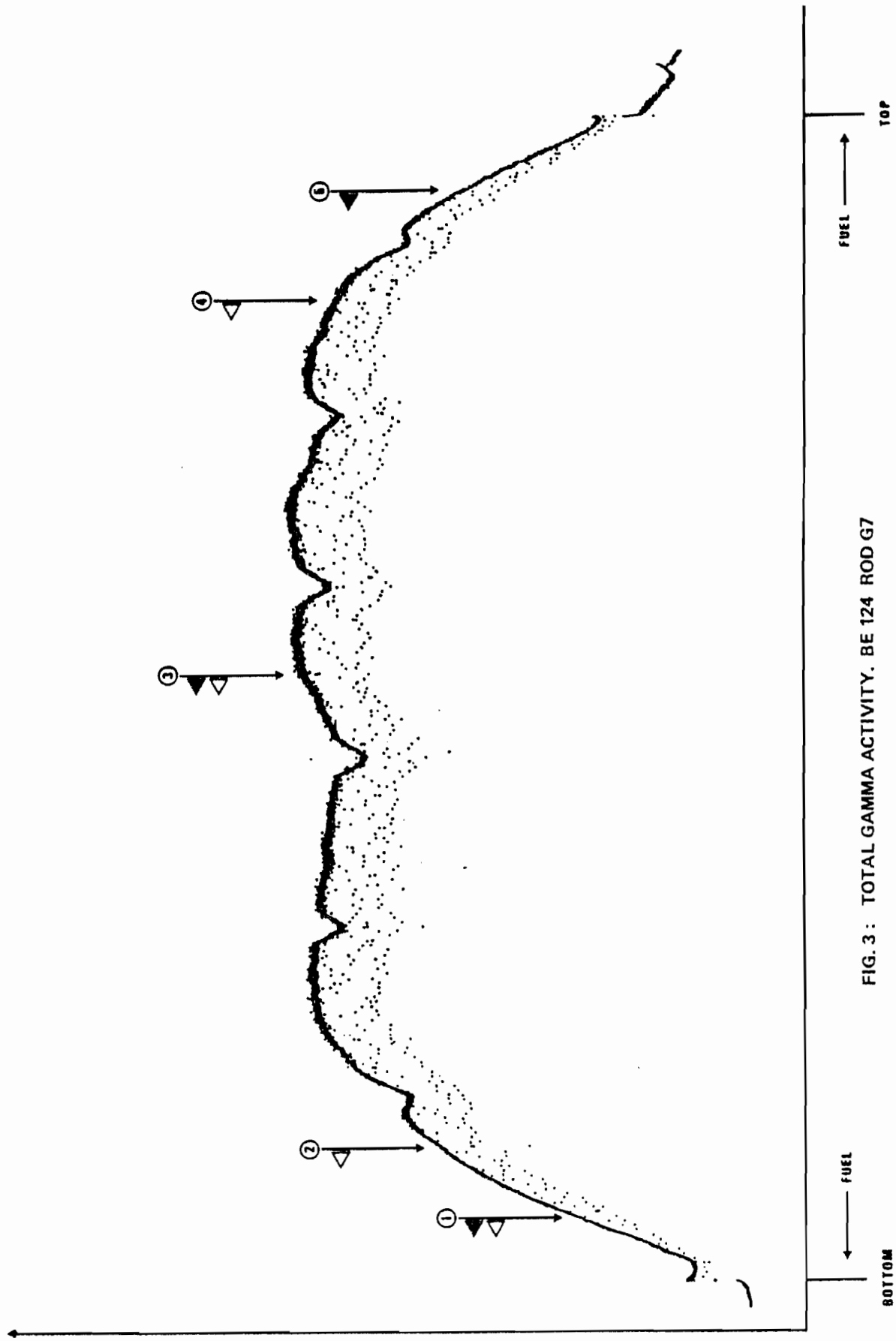


FIG. 3 : TOTAL GAMMA ACTIVITY. BE 124 ROD G7

▽ SAMPLES ANALYSED AT ISPRA

▼ SAMPLES ANALYSED AT KARLSRUHE

b) The measurements on fuel solutions were carried out with a coaxial type Ge(Li) detector, having a FWHM of 2.3 kev at 1332 kev of ^{60}Co, connected through a preamplifier and amplifier to a minicomputer LABEN 701.

The standard deviation for ^{134}Cs and ^{137}Cs was 1.5%, while the standard deviation for ^{154}Eu was 5.0%. Further details of on the measurement techniquesare given in Ref. 1, 2 and 3.
The results obtained at Ispra for ^{137}Cs, ^{134}Cs and ^{154}Eu expressed as dis/sec/g of final uranium at reactor shut down are reported in Table III.

3.1.3. Radiochemical procedures

The purpose of the radiochemical processes is to obtain from the original fuel solution purified samples of uranium, plutonium, americium and neodymium to be analysed by mass and alpha spectrometry.
At Ispra uranium and plutonium were both purified in the same step by means of solvent extraction techniques [4].
At Karlsruhe an ion-exchange technique was employed for the radiochemical purification of uranium, plutonium, neodymium and americium. The procedures are briefly described.

Uranium and plutonium purification

Weighed amounts of sample (containing about 1 µg Pu) and spikes (^{233}U and ^{242}Pu) were mixed and dried. A redox step consisting of reduction with hydroxylamine and oxidation with nitric acid was carried out on the redissolved residue to promote isotopic exchange. The solution in 8M HNO_3 was then passed through a DOWEX-1 ion-exchange column (0.5 g, x 8, 200-400 mesh). The first fraction

TABLE III - Specific Activities of Fission Products and Activity Ratios (dps/g of final Uranium) at Reactor Shut Down

FUEL ELEMENT	SAMPLES	LABORATORY	^{137}Cs ($\times 10^{9}$)	^{134}Cs ($\times 10^{9}$)	^{154}Eu ($\times 10^{8}$)	$\frac{^{134}Cs}{^{137}Cs}$	$\frac{^{154}Eu}{^{137}Cs}$ ($\times 10^{-2}$)
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe					
	P2	Ispra	3.308	4.287	1.916	1.296	5.790
	P3	Ispra	4.097	6.512	2.652	1.590	6.470
	E3 P1	Ispra	2.372	2.115	0.779	0.892	3.284
	P2	Karlsruhe					
	P3	Ispra	4.361	6.926	2.555	1.588	5.859
	P4	Ispra	3.774	5.568	2.147	1.475	5.689
	P4	Karlsruhe					
	P5	Ispra	2.751	3.084	1.186	1.121	4.311
	G7 P1	Ispra	2.053	1.714	0.547	0.835	2.664
	P1	Karlsruhe					
	P2	Ispra	3.041	3.749	1.653	1.233	5.436
	P3	Ispra	3.871	5.940	2.481	1.534	6.411
	P3	Karlsruhe					
	P4	Ispra	3.356	4.752	1.999	1.412	5.941
	P5	Karlsruhe					
	M14 P1	Karlsruhe					
	P3	Ispra	3.539	5.068	2.102	1.432	5.940
	P4	Karlsruhe					
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	4.665	7.597	2.743	1.628	5.880
	P3(1)	Karlsruhe					
	P4(1)	Ispra	4.376	6.842	2.609	1.564	5.962
	P5(1)	Ispra	3.758	5.091	2.079	1.355	5.532
	P5(2)	Ispra	2.968	3.244	1.338	1.093	4.508
	K14 P1	Ispra	2.782	3.236	1.364	1.163	4.900
	P3(1)	Ispra	4.438	7.353	2.909	1.657	6.554
	P4(1)	Karlsruhe					

containing fission products was discarded; most of the uranium was removed with 8M HNO_3 and a mixed uranium-plutonium fraction washed off with 0.35 M HNO_3. After drying, the residue was dissolved in 1M HNO_3 for mass spectrometry.

Neodymium and americium purification

After the addition of ^{150}Nd spike the solutions were conditioned as for the uranium/plutonium purification and passed through a DOWEX-1 column in 8M HNO_3. The first fraction, containing fission products as well as neodymium and americium, was collected and dried. The residue was dissolved in 0.05 M HCl and the rare-earths and americium absorbed on a DOWEX-50 cation column (0.3 g, x8, 200-400 mesh). After washing with 0.05 M HCl, the neodymium was eluted with 0.05 M α-isobutyric acid at pH 4.6. The elution was monitored by an alpha counter, by which means the fraction containing americium could be observed and kept and the next fraction, containing neodymium, could also be preserved.
The Am and Nd fractions were evaporated in the presence of 14M HNO_3 to eliminate traces of organic material and the residue taken up in 1M HNO_3 for subsequent mass spectrometry.
All analyses were made in triplicate, the third being held in reserve if lack of agreement was found in the results from the mass spectrometry of the first two.

3.1.4. Mass spectrometry

The instruments employed at Karlsruhe for the measurement of the isotopic ratios were of model CH-5 (Varian, Bremen). One was equipped with an automatic sample changer and has been described previously [6]; the other was a standard model. The data were handled by an automatic

system comprising a PDP-11 as data collector connected to an IBM-370 for data storage and for calculating results.

Uranium and plutonium were measured as metal ions using the two-filament technique. An amount containing as maximum 100 µg Pu as determined by α counting was dried on the evaporation filament for the purpose of mass spectrometry. The total error in the determination of the concentrations of uranium and plutonium is 0.5%. This error includes the uncertainties due to the isotope dilution procedures.

Neodymium was measured as MeO^+ ion from a single filament on the CH-5 mass spectrometer. Corrections in this case were made for contributions due to ^{18}O and also from background neodymium which was estimated from the level of ^{142}Nd found. During the analysis checks were made for Sm and Ce which interfere in the Nd measurement. The total error in the determination of ^{148}Nd, inclusive of the uncertainties due to the isotope dilution procedures, is about 1%.

The ^{150}Nd, ^{233}U, ^{242}Pu spikes were calibrated against NBS and CBNM Geel standards. The mass discrimination factors for these elements were determined for each machine also using NBS standards.
At the Ispra Laboratory a Micromass VG 30 B mass spectrometer was used, equipped with thermal ionisation triple-filament sources and connected to a PDP-8 for data treatment. The isotope composition and the concentration of americium was also determined in a few cases at Ispra by mass spectrometry and isotopic dilution using ^{243}Am as a spike. Further details of the methods employed are given in reference [1].
The atom ratios for uranium and plutonium are reported in Table IV with the date of the measurements.
The average standard deviations of the measurements

TABLE IV - Atom Ratios of U and Pu from Mass Spectrometry

FUEL ELEMENT	SAMPLES	LABORATORY	U (x 10^{-2})		Pu (x 10^{-1})			
			$^{235}/_{238}$	$^{236}/_{238}$	Date of measurement	$^{240}/_{239}$	$^{241}/_{239}$	$^{242}/_{239}$
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	1.438	0.313	17.07.78	3.021	1.183	0.291
	P2	Ispra	0.998	0.396	05.04.78	4.046	1.776	0.685
	P3	Ispra	0.742	0.431	12.04.78	4.686	2.133	1.111
	E3 P1	Ispra	1.337	0.297	08.07.77	3.165	1.259	0.321
	P2	Karlsruhe	0.897	0.400	17.07.78	4.298	1.818	0.814
	P3	Ispra	0.653	0.441	22.07.77	5.041	2.287	1.304
	P4	Ispra	0.798	0.408	16.07.77	4.481	2.059	0.928
	P4	Karlsruhe	0.830	0.412	17.07.78	4.548	2.007	0.982
	P5	Ispra	1.258	0.383	04.10.77	3.475	1.549	0.448
	G7 P1	Ispra	1.607	0.308	13.12.77	2.722	1.096	0.231
	P1	Karlsruhe	1.516	0.306	17.07.78	2.800	1.102	0.251
	P2	Ispra	1.146	0.408	19.12.77	3.778	1.665	0.576
	P3	Ispra	0.813	0.420	30.03.78	4.539	2.010	0.947
	P3	Karlsruhe	0.794	0.420	17.07.78	4.524	1.991	0.978
	P4	Ispra	1.077	0.430	19.12.77	4.005	1.844	0.713
	P5	Karlsruhe	1.064	0.378	17.07.78	3.813	1.703	0.624
	M14 P1	Karlsruhe	1.639	0.283	17.07.78	2.606	0.958	0.187
	P3	Ispra	0.936	0.405	18.04.78	4.138	1.865	0.756
	P4	Karlsruhe	1.088	0.377	17.07.78	3.737	1.673	0.596
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	0.542	0.416	11.11.77	5.455	2.452	1.692
	P3(1)	Karlsruhe	0.470	0.412	17.07.78	5.451	2.363	1.689
	P4(1)	Ispra	0.529	0.422	11.11.77	5.363	2.326	1.521
	P5(1)	Ispra	0.667	0.388	13.12.77	4.810	2.122	1.120
	P5(2)	Ispra	0.947	0.353	30.10.77	4.009	1.721	0.658
	K14 P1	Ispra	1.066	0.341	06.04.78	3.759	1.636	0.567
	P3(1)	Ispra	0.521	0.413	20.04.78	5.248	2.312	1.550
	P4(1)	Karlsruhe	0.541	0.408	17.07.78	5.234	2.225	1.441

of the different isotopic ratios are given in Table V. The $^{148}Nd/^{238}U$, the $^{137}Cs/^{238}U$ and $^{239}Pu/^{238}U$ atom ratios and the total Pu/U mass ratio are reported in Table VI. The Karlsruhe Laboratory also performed isotopic composition measurements by mass spectrometry on the krypton and xenon fission gases.
The results of these measurements are reported in Table VII. Their average standard deviations are reported in Table V. A detailed comparison between the results obtained at Ispra and Karlsruhe is presented in section 3.3.2.

3.1.5. Alpha-spectrometry

α spectra were obtained at Karlsruhe using an ORTEC silicon semiconductor detector connected to a multichannel analyser (FRIESIKE & HOEPFNER) which in turn was connected to the PDP-11 computer for spectra processing and data transmission. Samples of 5 - 10 μl were dried on stainless-steel planchets and then heated to dull red heat to remove volatile salts and organic material.

Total alpha counts were also made using a gas-flow proportional counter with an efficiency determined relative to a standard $^{239}Pu+^{240}Pu$ source.
Alpha spectrometry was carried out at Ispra by means of a silicon semiconductor detector connected to a LABEN 701 computer for data storage and processing.
Samples, countaining about 0.01 mg of uranium were dropped onto tantalum counting-plates and dried before counting.

α spectra were taken directly from the original solution and also on the plutonium/uranium fraction after separation.
Activity ratios

$$\frac{^{238}Pu + ^{241}Am}{^{239}Pu + ^{240}Pu}, \quad \frac{^{242}Cm}{^{239}Pu + ^{240}Pu}, \quad \frac{^{244}Cm}{^{239}Pu + ^{240}Pu}$$

TABLE V - Average Standard Deviations of the Measurements

ISOTOPIC RATIOS OF U AND Pu

$^{235}U/^{238}U$	0.05 %
$^{236}U/^{238}U$	0.08 %
$^{240}Pu/^{239}Pu$	0.08 %
$^{241}Pu/^{239}Pu$	0.1 %
$^{242}Pu/^{239}Pu$	0.3 %

ISOTOPIC RATIOS OF FISSION GASES

$^{83}Kr/^{86}Kr$	0.3 %	$^{130}Xe/^{134}Xe$	0.5 %
$^{84}Kr/^{86}Kr$	0.2 %	$^{131}Xe/^{134}Xe$	0.2 %
$^{85}Kr/^{86}Kr$	0.3 %	$^{132}Xe/^{134}Xe$	0.2 %
		$^{136}Xe/^{131}Xe$	0.2 %

BURNUP DETERMINATION

^{148}Nd	1.5 %	
^{137}Cs	2.5 %	(destructive)
^{137}Cs	5.0 %	(non-destructive)

TABLE VI – Atom Ratios of Nd, Cs and Pu Referred to Final U

FUEL ELEMENT	SAMPLES	LABORATORY	$^{148}Nd/^{238}U$ ($\times 10^{-4}$)	$^{137}Cs/^{238}U$ ($\times 10^{-3}$)	$^{239}Pu/^{238}U$ ($\times 10^{-3}$)	Total Pu/U Mass Ratio ($\times 10^{-3}$)
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	4.01		4.54	6.65
	P2	Ispra		1.88		
	P3	Ispra	6.46	2.33	5.29	9.82
	E3 P1	Ispra	3.81	1.35	4.45	6.62
	P2	Karlsruhe	6.73		4.88	8.50
	P3	Ispra	6.96	2.48	5.04	9.69
	P4	Ispra	5.89	2.15	5.19	9.30
	P4	Karlsruhe	5.91		5.17	9.31
	P5	Ispra	4.34	2.36	4.86	7.63
	G7 P1	Ispra	3.23	1.77	4.42	6.25
	P1	Karlsruhe	4.31		4.59	6.56
	P2	Ispra	4.91	2.59	4.92	8.09
	P3	Ispra	6.02	3.28	5.25	9.47
	P3	Karlsruhe	5.95		5.31	9.60
	P4	Ispra	5.29	2.86	5.33	9.03
	P5	Karlsruhe	4.91		5.33	8.80
	M14 P1	Karlsruhe	2.94		4.13	5.17
	P3	Ispra	5.60	3.01	5.27	9.07
	P4	Karlsruhe	4.73		5.19	8.49
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	7.31	3.94	4.77	9.73
	P3(1)	Karlsruhe	7.09		4.86	9.85
	P4(1)	Ispra	6.82	3.70	4.65	8.84
	P5(1)	Ispra	5.74	3.20	4.57	8.17
	P5(2)	Ispra	4.59	2.55	4.46	7.21
	K14 P1	Ispra	4.83	2.39	4.81	7.57
	P3(1)	Ispra	7.03	3.75	4.94	9.36
	P4(1)	Karlsruhe	6.28		4.74	9.28

TABLE VII - Isotopic Ratios of Fission Gases

FUEL ELEMENT	SAMPLES	LABORATORY	Krypton			Xenon			
			83/86	84/86	85/86	130/134	131/134	132/134	136/134
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	0.134	0.305	0.055	0.0024	0.335	0.685	1.412
	P2	Ispra							
	P3	Ispra							
	E3 P1	Ispra							
	P2	Karlsruhe	0.233	0.613	0.100	0.0046	0.306	0.734	1.520
	P3	Ispra							
	P4	Ispra							
	P4	Karlsruhe	0.231	0.611	0.105	0.0049	0.296	0.625	1.518
	P5	Ispra							
	G7 P1	Ispra							
	P1	Karlsruhe	0.255	0.573	0.104	0.0024	0.333	0.656	1.400
	P2	Ispra							
	P3	Ispra							
	P3	Karlsruhe	0.229	0.613	0.104	0.0049	0.294	0.757	1.534
	P4	Ispra							
	P5	Karlsruhe	0.245	0.602	0.107	0.0036	0.304	0.723	1.487
	M14 P1	Karlsruhe	0.257	0.567	0.106	0.0020	0.339	0.649	1.375
	P3	Ispra							
	P4	Karlsruhe	0.241	0.594	0.105	0.0036	0.303	0.700	1.491
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra							
	P3(1)	Karlsruhe	0.212	0.631	0.104	0.0065	0.275	0.761	1.585
	P4(1)	Ispra							
	P5(1)	Ispra							
	P5(2)	Ispra							
	K14 P1	Ispra							
	P3(1)	Ispra							
	P4(1)	Karlsruhe	0.241	0.594	0.105	0.0036	0.303	0.700	1.491

were determined from the untreated solution with average relative standard deviations of 1.6%, 3.6% and 2.7% respectively, and

$$\frac{^{238}Pu}{^{239}Pu + ^{240}Pu}$$

from the uranium/plutonium fraction with an average relative standard deviation of 0.9%.
Alpha spectrometry results obtained at Ispra and Karlsruhe Laboratories are reported in Table VIII with the date of the measurements.

3.2. Processing of the Experimental Data

3.2.1. Burnup determination by the ^{148}Nd and ^{137}Cs methods

The processing of the rough analytical data in order to obtain the burnup from ^{148}Nd or ^{137}Cs atom content of the samples, has been extensively illustrated in previous reports [1, 2, 3, 4, 5].

The ^{148}Nd amount measured has been corrected to take into account :

a) the ^{148}Nd burn-out by neutron capture. Correction factors not higher than 1% (0.5 + 1.0% for different burnup levels) have been applied

b) the ^{148}Nd build up from ^{147}Nd capture. The amount of the correction (1.0 + 1.5% according to the different burnup levels) has been evaluated on the basis of a work by Maek et al. [7] in which the value of the activation cross section of ^{147}Nd is suggested as

TABLE VIII - Alpha Activity Ratios of Pu, Am and Cm Isotopes

FUEL ELEMENT	SAMPLES	LABORATORY	Before Separation				After Separation	
			Date of measurement	$\frac{^{238}Pu+^{241}Am}{^{239}Pu+^{240}Pu}$	$\frac{^{242}Cm}{^{239}Pu+^{240}Pu}$	$\frac{^{244}Cm}{^{239}Pu+^{240}Pu}$	Date of measurement	$\frac{^{238}Pu}{^{239}Pu+^{240}Pu}$
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	19.07.78	2.07	0.098	0.32	19.07.78	1.32
	P2	Ispra	26.02.78	3.54	0.21	1.24	10.01.78	2.46
	P3	Ispra	07.03.78	4.54	0.25	2.87	15.03.78	3.48
	E3 P1	Ispra	10.08.77	2.04	0.26	0.26	09.08.77	1.37
	P2	Karlsruhe	19.07.78	3.21	0.23	1.42	19.07.78	2.47
	P3	Ispra	28.08.77	4.69	0.60	3.62	30.08.77	3.65
	P4	Ispra	28.08.77	3.96	0.56	2.24	30.08.77	2.97
	P4	Karlsruhe	19.07.78	3.99	0.31	2.26	19.07.78	2.92
	P5	Ispra	10.08.77	2.52	0.35	0.58	09.08.77	1.80
	G7 P1	Ispra	21.12.77	1.86	0.10	0.14	10.01.78	1.17
	P1	Karlsruhe	19.07.78	1.98	0.06	0.29	19.07.78	1.24
	P2	Ispra	21.12.77	3.28	0.23	0.98	10.01.78	2.14
	P3	Ispra	31.12.77	4.67	0.32	2.60	10.01.78	3.19
	P3	Karlsruhe	19.07.78	3.53	0.24	2.17	19.07.78	3.05
	P4	Ispra	03.01.78	3.95	0.27	1.54	10.01.78	2.60
	P5	Karlsruhe	19.07.78	2.95	0.19	1.17	19.07.78	2.28
	M14 P1	Karlsruhe	19.07.78	1.55	0.04	0.18	19.07.78	0.96
	P3	Ispra	21.02.78	3.75	0.21	1.65	15.03.78	2.58
	P4	Karlsruhe	19.07.78	2.95	0.16	1.01	19.07.78	2.17
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	13.10.77	5.15	0.45	5.16	14.11.77	4.17
	P3(1)	Karlsruhe	19.07.78	4.48	0.41	4.45	19.07.78	3.79
	P4(1)	Ispra	13.10.77	4.74	0.44	4.21	14.11.77	3.66
	P5(1)	Ispra	13.10.77	3.83	0.39	2.14	14.11.77	2.87
	P5(2)	Ispra	13.10.77	2.79	0.29	0.77	14.11.77	1.91
	K14 P1	Ispra	02.03.78	2.70	0.16	0.69	15.03.78	1.86
	P3(1)	Ispra	21.03.78	4.89	0.26	4.39	15.03.78	3.76
	P4(1)	Karlsruhe	19.07.78	3.73	0.35	3.22	19.07.78	3.39

$$\sigma_c^{147} = 440 \pm 150 \text{ b}$$

The measured ^{137}Cs has also been corrected for in-pile decay, applying correction factors of the order of 4.0% (3.7 + 4.1% according to the irradiation histories).

The burnup has also been evaluated from the non-destructive ^{137}Cs measurements already referred to (see 3.1.2.). The ^{137}Cs activity values measured have been converted to burnup by means of a calibration curve obtained by using the burnup values derived from ^{148}Nd measurements carried out at the Ispra Laboratory, as indicated in Table IX.

The following correlation has been established :

$$Y = 108.19\ X + 3957.4$$

where : Y = MWD/MTU

X = counts/min ^{137}Cs measured at the fuel rod.

The correlation coefficient of the regression equation was 0.977.
This equation has been considered as a calibration line for the determination of other burnup values.

The burnup values determined at Ispra and Karlsruhe from ^{148}Nd and ^{137}Cs are reported in Table IX.

3.2.2. Nuclear data

The average fission yield evaluated for ^{148}Nd and ^{137}Cs at different burnup levels, using the nuclear data of Table X [8, 9], and the fission fractions of Table XI [10], are the following :

TABLE IX – Burnup Values Obtained by Means of Three Independent Experimental Methods (MWD/MTU)

FUEL ELEMENT	SAMPLES	LABORATORY	^{148}Nd	^{137}Cs Destructive	^{137}Cs Non-destruc.(a)
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	21,170		19,520
	P2	Ispra			27,910
	P3	Ispra	33,750	33,160	33,760
	E3 P1	Ispra	20,180	19,540	19,420
	P2	Karlsruhe	35,100		29,350
	P3	Ispra	36,260	35,220	35,510
	P4	Ispra	30,890	30,640	28,140
	P4	Karlsruhe	30,940		
	P5	Ispra	22,860	22,570	22,920
	G7 P1	Ispra	17,130	16,970	17,490
	P1	Karlsruhe	22,700		
	P2	Ispra	25,830	24,880	26,240
	P3	Ispra	31,500	31,400	31,920
	P3	Karlsruhe	31,140		
	P4	Ispra	27,710	27,420	29,460
	P5	Karlsruhe	25,810		28,830
	M14 P1	Karlsruhe	15,600		15,790
	P3	Ispra	29,360	28,800	27,200
	P4	Karlsruhe	24,900		27,460
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	38,100	37,720	36,290
	P3(1)	Karlsruhe	36,880		
	P4(1)	Ispra	35,640	35,480	36,070
	P5(1)	Ispra	30,160	30,660	31,870
	P5(2)	Ispra	24,220	24,400	26,060
	K14 P1	Ispra	25,450	22,900	22,460
	P3(1)	Ispra	36,670	35,990	35,120
	P4(1)	Karlsruhe	32,900		34,630

(a) Measurement performed at the Ispra Establishment LMA Laboratory

TABLE X – Nuclear Data Used in the Determination of the Burnup Values

ENERGY RELEASE PER FISSION			
^{235}U	201.7 MeV	^{239}Pu	210.0 MeV
^{238}U	205.0 MeV	^{241}Pu	212.4 MeV
FISSION YIELDS OF ^{137}Cs			
^{235}U	6.23 %	^{239}Pu	6.69 %
^{238}U	6.28 %	^{241}Pu	6.60 %
FISSION YIELDS OF ^{148}Nd			
^{235}U	1.69 %	^{239}Pu	1.69 %
^{238}U	2.13 %	^{241}Pu	1.91 %

TABLE XI – Fission Fractions for the Various Nuclides at Different Burnup Levels Used in the Determination of the Burnup Values

MWD/MTU	P E R C E N T A G E			
	^{235}U	^{239}Pu	^{241}Pu	^{238}U
15,000	68.5	22.0	1.5	8.0
20,000	64.5	25.0	2.5	8.0
25,000	60.0	28.0	4.0	8.0
30,000	56.0	30.5	5.5	8.0
35,000	52.0	33.0	7.0	8.0

MWD/MTU	^{148}Nd Average fission yield	^{137}Cs Average fission yield
15,000	$1.729 \cdot 10^{-2}$	$6.341 \cdot 10^{-2}$
20,000	$1.731 \cdot 10^{-2}$	$6.358 \cdot 10^{-2}$
25,000	$1.734 \cdot 10^{-2}$	$6.377 \cdot 10^{-2}$
30,000	$1.737 \cdot 10^{-2}$	$6.394 \cdot 10^{-2}$
35,000	$1.741 \cdot 10^{-2}$	$6.412 \cdot 10^{-2}$

Due to the small differences among the average fission yields at different burnup levels, the values

$$Y(^{148}Nd) = 1.734 \cdot 10^{-2}$$

$$Y(^{137}Cs) = 6.376 \cdot 10^{-2}$$

have been adopted for all the fuel samples.

Since : 1 MWD = $\frac{5.387 \times 10^{23}}{E}$ fissions,

where E is the average energy released per fission in MeV, once F_T is known, we have :

$$B(MWD/MTU) = \frac{F_T \cdot 10^4 \cdot N/A}{(5.387 \cdot 10^{23})/E}$$

where :

N is Avogadro's number

A is the average atomic weight of a heavy atom (A = 238).

Using the energy release per fission and the fission fraction of the Tables X and XI, the following average values of MeV/fission have been calculated at different burnup levels :

MWD/MTU.	MeV/fission (average values)
15,000	203.95
20,000	204.31
25,000	204.72
30,000	205.08
35,000	205.45

Due to the small differences among the average values at different burnup levels, the value of 204.7 MeV/fission has been used for all the samples.
The burnup value is then given by the equation :

$$B = 9.617 \cdot 10^3 \cdot F_T \ (MWD/MTU)$$

The half lives of Pu, Am and Cm isotopes utilised in this work are the following [11] in years :

^{238}Pu	87.8 ± 0.8	^{241}Am	432 ± 4
^{239}Pu	$(2.41 \pm 0.01) \times 10^4$	^{242}Am	152 ± 7
^{240}Pu	$(6.55 \pm 0.07) \times 10^3$	^{243}Am	7370 ± 40
^{241}Pu	14.7 ± 0.4	^{242}Cm	0.446 ± 0.0003
^{242}Pu	$(3.87 \pm 0.05) \times 10^5$	^{244}Cm	18.11 ± 0.01

3.3.1. Comparison between burnup values determined by different experimental techniques

The total error in the determination of the burnup values can be evaluated by combining the statistical errors of the measurements and the nuclear data uncertainties. Errors are reported in Table V. A comparison between the burnup values obtained from ^{148}Nd and ^{137}Cs is presented in Table IX.
The values derived from destructive ^{137}Cs determinations are generally lower than those derived from ^{148}Nd.
The average difference between the values obtained at Ispra from ^{137}Cs and ^{148}Nd is 1.2%.

As the sample K14 P1 presents a difference of about 11.0%, a migration of fission products could have occurred in the cutting area. This value was not considered in the comparison.

The comparison between burnup values determined from destructive and non-destructive measurements of ^{137}Cs is also presented in Table IX.
The average difference between the two sets of data is about 0.7%.

3.3.2. Comparison between the results obtained at Ispra and Karlsruhe

The statistical errors in the determination of atom ratios by mass spectrometry and of activity ratios by alpha spectrometry, expressed as standard deviations of the measurements, were reported in section 3.1.

The systematic errors in these determinations are much more difficult to evaluate.
However, we feel that comparison between the results

3.2.3. Determination of isotopic composition, build up and depletion of heavy isotopes

From the mass spectrometry data (Table IV) the isotopic composition of uranium was calculated and presented in Table XII.

The isotopic composition of plutonium presented in Table XIII was calculated from the mass spectrometry data (Table IV) and alpha spectrometry data (Table VIII) which were combined using simple equations.

The heavy isotope data were also processed in order to obtain the build up and depletion of the different isotopes.

The build up and depletion of U, Pu, Cm and Am isotopes are presented in Tables XIV, XV and XVI.

3.3. Analysis of the Accuracy of the Values of Burnup and Isotopic Composition

The evaluation of the accuracy of the experimental values of burnup and isotopic composition was carried out following three different procedures :

- The burnup values were determined by means of two different techniques.
- Pairs of adjacent fuel sections were analysed at Ispra and Karslruhe.
- Isotope Correlations Technique has been extensively used [1, 2, 3, 4, 12, 13, 14, 15, 16, 17].

TABLE XII - Uranium Isotopic Composition (Atoms)

FUEL ELEMENT	SAMPLES	LABORATORY	^{235}U	^{236}U	^{238}U
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	1.413	0.308	98.279
	P2	Ispra	0.985	0.391	98.624
	P3	Ispra	0.733	0.429	98.838
	E3 P1	Ispra	1.316	0.292	98.392
	P2	Karlsruhe	0.886	0.395	98.719
	P3	Ispra	0.646	0.436	98.918
	P4	Ispra	0.789	0.403	98.808
	P4	Karlsruhe	0.820	0.407	98.773
	P5	Ispra	1.238	0.377	98.385
	G7 P1	Ispra	1.577	0.302	98.121
	P1	Karlsruhe	1.489	0.301	98.210
	P2	Ispra	1.128	0.402	98.470
	P3	Ispra	0.803	0.415	98.780
	P3	Karlsruhe	0.784	0.415	98.801
	P4	Ispra	1.061	0.424	98.515
	P5	Karlsruhe	1.049	0.373	98.578
	M14 P1	Karlsruhe	1.608	0.278	98.114
	P3	Ispra	0.924	0.400	98.676
	P4	Karlsruhe	1.072	0.372	98.556
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	0.537	0.412	99.051
	P3(1)	Karlsruhe	0.466	0.408	99.126
	P4(1)	Ispra	0.524	0.418	99.058
	P5(1)	Ispra	0.660	0.384	98.956
	P5(2)	Ispra	0.935	0.348	98.717
	K14 P1	Ispra	1.051	0.336	98.613
	P3(1)	Ispra	0.516	0.409	99.075
	P4(1)	Karlsruhe	0.531	0.400	98.069

TABLE XIII - Plutonium Isotopic Composition (Atoms) at Reactor Shut Down

FUEL ELEMENT	SAMPLES	LABORATORY	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	0.55	67.31	20.42	9.70	2.02
	P2	Ispra	1.16	58.43	23.64	12.76	4.01
	P3	Ispra	1.68	53.62	25.13	13.62	5.95
	E3 P1	Ispra	0.65	66.54	21.06	9.62	2.13
	P2	Karlsruhe	1.29	57.02	24.57	12.44	4.68
	P3	Ispra	1.77	51.77	26.09	13.62	6.75
	P4	Ispra	1.41	55.46	24.85	13.13	5.15
	P4	Karlsruhe	1.16	54.85	25.04	13.41	5.53
	P5	Ispra	0.84	63.07	21.92	11.35	2.82
	G7 P1	Ispra	0.52	69.87	19.02	8.98	1.61
	P1	Karlsruhe	0.62	68.97	19.39	9.25	1.78
	P2	Ispra	1.02	60.32	23.16	11.97	3.53
	P3	Ispra	1.54	55.08	25.01	13.16	5.21
	P3	Karlsruhe	1.58	54.76	24.87	13.28	5.51
	P4	Ispra	1.23	58.52	23.44	12.64	4.17
	P5	Karlsruhe	0.94	59.88	22.92	12.42	3.84
	M14 P1	Karlsruhe	0.47	71.22	18.63	8.31	1.37
	P3	Ispra	1.23	57.71	23.88	12.82	4.36
	P4	Karlsruhe	0.94	60.39	22.66	12.31	3.70
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	2.07	48.95	26.70	14.00	8.28
	P3(1)	Karlsruhe	1.49	49.04	26.84	14.11	8.51
	P4(1)	Ispra	1.81	50.10	26.87	13.60	7.62
	P5(1)	Ispra	1.40	53.54	25.75	13.32	5.99
	P5(2)	Ispra	0.91	59.43	23.83	11.92	3.91
	K14 P1	Ispra	0.87	60.92	22.90	11.86	3.45
	P3(1)	Ispra	1.82	50.21	26.35	13.84	7.78
	P4(1)	Karlsruhe	1.42	50.70	26.64	13.74	7.51

TABLE XIV - Buildup and Depletion of Uranium Isotopes ($kg/MTU_{initial}$)

FUEL ELEMENT	SAMPLES	LABORATORY	^{235}U Depletion	^{236}U Build up	^{238}U Depletion
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	16.31	2.99	15.95
	P2	Ispra			
	P3	Ispra	23.07	4.06	25.32
	E3 P1	Ispra	17.25	2.82	12.88
	P2	Karlsruhe	21.56	3.77	27.85
	P3	Ispra	23.91	4.12	27.01
	P4	Ispra	22.52	3.83	22.22
	P4	Karlsruhe	22.16	3.91	23.83
	P5	Ispra	18.15	3.62	16.59
	G7 P1	Ispra	14.79	2.92	11.97
	P1	Karlsruhe	15.60	2.93	18.18
	P2	Ispra	19.24	3.85	19.21
	P3	Ispra	22.39	3.94	23.36
	P3	Karlsruhe	22.50	3.98	23.90
	P4	Ispra	19.91	4.05	21.62
	P5	Karlsruhe	19.91	3.60	19.80
	M14 P1	Karlsruhe	14.31	2.73	10.91
	P3	Ispra	21.22	3.81	21.73
	P4	Karlsruhe	19.67	3.60	18.85
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	23.25	3.89	29.38
	P3(1)	Karlsruhe	23.89	3.87	28.78
	P4(1)	Ispra	23.35	3.95	26.36
	P5(1)	Ispra	22.03	3.66	20.99 ✓
	P5(2)	Ispra	19.36	3.33	16.25 ✓
	K14 P1	Ispra	18.27	3.22	18.86
	P3(1)	Ispra	23.44	3.86	27.76
	P4(1)	Karlsruhe	23.26	3.82	24.55

TABLE XV - Build up of Plutonium Isotopes ($kg/MTU_{initial}$)

FUEL ELEMENT	SAMPLES	LABORATORY	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	Total Pu
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	0.036	4.35	1.32	0.62	0.128	6.454
	P2	Ispra						
	P3	Ispra	0.156	5.02	2.36	1.28	0.564	9.380
	E3 P1	Ispra	0.041	4.28	1.36	0.62	0.139	6.440
	P2	Karlsruhe	0.104	4.62	1.99	1.02	0.380	8.114
	P3	Ispra	0.165	4.77	2.41	1.26	0.629	9.234
	P4	Ispra	0.125	4.94	2.22	1.18	0.464	8.929
	P4	Karlsruhe	0.103	4.91	2.24	1.19	0.488	8.931
	P5	Ispra	0.062	4.65	1.62	0.84	0.211	7.383
	G7 P1	Ispra	0.031	4.26	1.16	0.55	0.099	6.100
	P1	Karlsruhe	0.039	4.39	1.23	0.59	0.112	6.361
	P2	Ispra	0.079	4.70	1.81	0.94	0.279	7.808
	P3	Ispra	0.138	4.99	2.27	1.20	0.478	9.076
	P3	Karlsruhe	0.145	5.04	2.29	1.21	0.500	9.194
	P4	Ispra	0.106	5.08	2.04	1.11	0.366	8.702
	P5	Karlsruhe	0.079	5.08	1.95	1.05	0.320	8.479
	M14 P1	Karlsruhe	0.025	3.98	1.04	0.46	0.075	5.581
	P3	Ispra	0.107	5.02	2.08	1.12	0.384	8.711
	P4	Karlsruhe	0.076	4.96	1.86	1.00	0.299	8.195
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	0.190	4.52	2.47	1.30	0.773	9.253
	P3(1)	Karlsruhe	0.139	4.60	2.52	1.32	0.787	9.366
	P4(1)	Ispra	0.159	4.41	2.38	1.21	0.680	8.839
	P5(1)	Ispra	0.114	4.36	2.11	1.09	0.494	8.168
	P5(2)	Ispra	0.065	4.27	1.72	0.86	0.285	7.200
	K14 P1	Ispra	0.065	4.60	1.74	0.90	0.264	7.569
	P3(1)	Ispra	0.169	4.69	2.47	1.30	0.735	9.364
	P4(1)	Karlsruhe	0.126	4.51	2.37	1.21	0.658	8.874

TABLE XVI - Build up of Am and Cm Isotopes ($kg/MTU_{initial}$)

FUEL ELEMENT	SAMPLES	LABORATORY	AMERICIUM			CURIUM	
			241 ($\times 10^{-2}$)	242 ($\times 10^{-4}$)	243 ($\times 10^{-1}$)	242 ($\times 10^{-3}$)	244 ($\times 10^{-3}$)
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	2.19(a)			7.40	2.22
	P2	Ispra					
	P3	Ispra	6.08(b)			15.65	33.97
	E3 P1	Ispra	2.93(b)			4.50	2.04
	P2	Karlsruhe					12.58
	P3	Ispra	9.00(b)			14.79	41.62
	P4	Ispra	7.30(b)			15.01	24.80
	P4	Karlsruhe				33.17	22.18
	P5	Ispra	2.37(b)			7.10	5.13
	G7 P1	Ispra	1.40(a)	2.88	0.086	2.94	1.04
	P1	Karlsruhe	2.21(a)				1.97
	P2	Ispra	2.48(a)	4.94	0.39	8.58	9.49
	P3	Ispra	17.58(b)			14.79	29.73
	P3	Karlsruhe				27.15	26.73
	P4	Ispra	13.98(b)			11.88	16.52
	P5	Karlsruhe				19.56	10.68
	M14 P1	Karlsruhe	0.67(a)				1.04
	P3	Ispra	9.39(b)			11.30	17.99
	P4	Karlsruhe				14.07	8.91
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	4.10(a)	7.29	1.54	14.54	59.81
	P3(1)	Karlsruhe				47.03	45.94
	P4(1)	Ispra	4.03(a)	19.30	1.38	14.36	47.15
	P5(1)	Ispra	5.53(b)	12.07	0.42	11.70	22.07
	P5(2)	Ispra	2.77(a)	7.10	0.36	7.56	6.95
	K14 P1	Ispra	2.52(a)			8.13	6.53
	P3(1)	Ispra	7.49(b)	7.57	1.40	17.75	52.59
	P4(1)	Karlsruhe				33.10	31.68

(a) Values obtained by mass spectrometry (Ispra) and alpha spectrometry (Karlsruhe)

(b) Values obtained by alpha spectrometry (Ispra)

obtained in different laboratories is a very effective tool for evaluating the total error in the measured quantities.

The agreement between the burnup values from ^{148}Nd measured at Ispra and Karlsruhe for three pairs of adjacent fuel sections is about 1.4% (see Table IX).

For sample G7 P1 the burnup value obtained at Karlsruhe seems to be extremely high (about 30.0% higher than the Ispra one).
Considering that three different methods used at Ispra give approximately 17,000 MWD/MTU, this value has not been taken into account.
Besides, a calculation made on the asymptotic G7 samples with the burnup code RIBOT [18] indicated a possible error of approximately 25% in the evaluation of the burnup of G7 P1 sample analysed at Karlsruhe.

The percentage differences between the values of the ^{235}U depletion, the ^{236}U and ^{239}Pu build up, the total Pu, the Pu/U mass ratios and the $^{148}Nd/^{238}U$ ratios obtained at Ispra and Karlsruhe for the four pairs of adjacent pellets are presented in Table XVII.

Agreement is very satisfactory for three samples.
For the sample G7 P1, the values found at the Karlsruhe Laboratory are much higher in comparison with those found at Ispra, as it occurred for the burnup value.

The average differences between the values of isotopic composition of U and Pu for the three adjacent pellets are the following :

^{235}U	3.8%	^{239}Pu	0.32%
^{236}U	0.06%	^{240}Pu	0.24%
^{238}U	0.02%	^{241}Pu	1.27%
^{238}Pu	14.3%	^{242}Pu	5.3%

TABLE XVII - Comparison between Values Determined at Ispra and Karlsruhe Laboratories

SAMPLE	^{235}U DEPLETION			^{236}U BUILD UP			^{239}Pu BUILD UP		
	Ispra	Karlsruhe	Percent Differ.	Ispra	Karlsruhe	Percent Differ.	Ispra	Karlsruhe	Percent Differ.
E3 P4	0.75	0.74	- 1.33	3.83	3.91	2.09	4.94	4.91	- 0.61
G7 P1	0.49	0.52	6.12	2.92	2.93	0.34	4.26	4.39	3.05
G7 P3	0.75	0.75	-	3.94	3.98	1.02	4.99	5.04	1.00
G14 $P3_1$	0.82	0.80	- 2.44	3.89	3.87	-0.51	4.52	4.60	1.77

SAMPLE	TOTAL Pu			$Pu/_U$ MASS RATIO			$^{148}Nd/\ ^{238}U$		
	Ispra	Karlsruhe	Percent Differ.	Ispra	Karlsruhe	Percent Differ.	Ispra	Karlsruhe	Percent Differ.
E3 P4	8.929	8.931	0.02	9.30	9.32	0.22	5.89	5.91	0.34
G7 P1	6.100	6.361	4.28	6.25	6.56	4.96	3.23	4.31	33.44
G7 P3	9.076	9.194	1.30	9.47	9.60	1.37	6.02	5.95	- 1.16
G14 $P3_1$	9.253	0.366	1.22	9.73	9.85	1.23	7.31	7.09	- 3.00

Contrary to what occurred in the preceeding post-irradiation experiments [1, 2, 3, 4] the ^{241}Am build up values present a rather complex situation.
An accurate analysis of the experimental data led to the conclusion that the values can be split into two sets.
The first set (A) includes mass-spectrometry data obtained at Ispra and α-spectrometry data obtained at Karlsruhe.
The second set (B) is only composed of the α-spectrometry data obtained at Ispra.
In order to give an idea of the different distribution and of the discrepancy between the A and B sets of data, the ^{241}Am build up values have been plotted against the burnup and presented in Fig. 4 and Table XVIII.
Considering that the values belonging to A are obtained by two laboratories with different experimental techniques, a greater degree of confidence should be placed in these values.
This conclusion is also supported by the results obtained with the RIBOT code previously mentioned.

The ^{242}Cm data also present a similar situation. All Karlsruhe build up values are higher than those obtained at Ispra (see Fig. 5a). The check with the RIBOT code also indicated the possible presence of a systematic error in Karlsruhe determinations.
The ^{244}Cm build up data, both from Ispra and Karlsruhe Laboratories, lie, on the contrary, on a single line with the only exception of the E3P2 sample (see Fig. 5b).

3.3.3. Isotope correlations

A consistency check of the most important quantities determined by the post-irradiation analyses has been performed by means of the Isotope Correlation Technique. Both spectrum-dependent and independent correlations have been used to test the experimental data.

TABLE XVIII - Build up of ^{241}Am

FUEL ELEMENT	SAMPLES	LABORATORY	SET A ($\times 10^{-2}$)	SET B ($\times 10^{-2}$)
BE 124 3.00 wt% 235U	D1 P1	Karlsruhe	2.19	
	P2	Ispra		
	P3	Ispra		6.08
	E3 P1	Ispra		2.93
	P2	Karlsruhe		
	P3	Ispra		9.00
	P4	Ispra		7.30
	P4	Karlsruhe		
	P5	Ispra		2.37
	G7 P1	Ispra	1.40	
	P1	Karlsruhe	2.24	
	P2	Ispra	2.48	
	P3	Ispra		17.58
	P3	Karlsruhe		
	P4	Ispra		13.98
	P5	Karlsruhe		
	M14 P1	Karlsruhe	0.67	
	P3	Ispra		9.39
	P4	Karlsruhe		
BE 210 2.83 wt% 235U	G14 P3(1)	Ispra	4.10	
	P3(1)	Karlsruhe		
	P4(1)	Ispra	4.03	
	P5(1)	Ispra		5.53
	P5(2)	Ispra	2.77	
	K14 P1	Ispra	2.52	
	P3(1)	Ispra		7.49
	P4(1)	Karlsruhe		

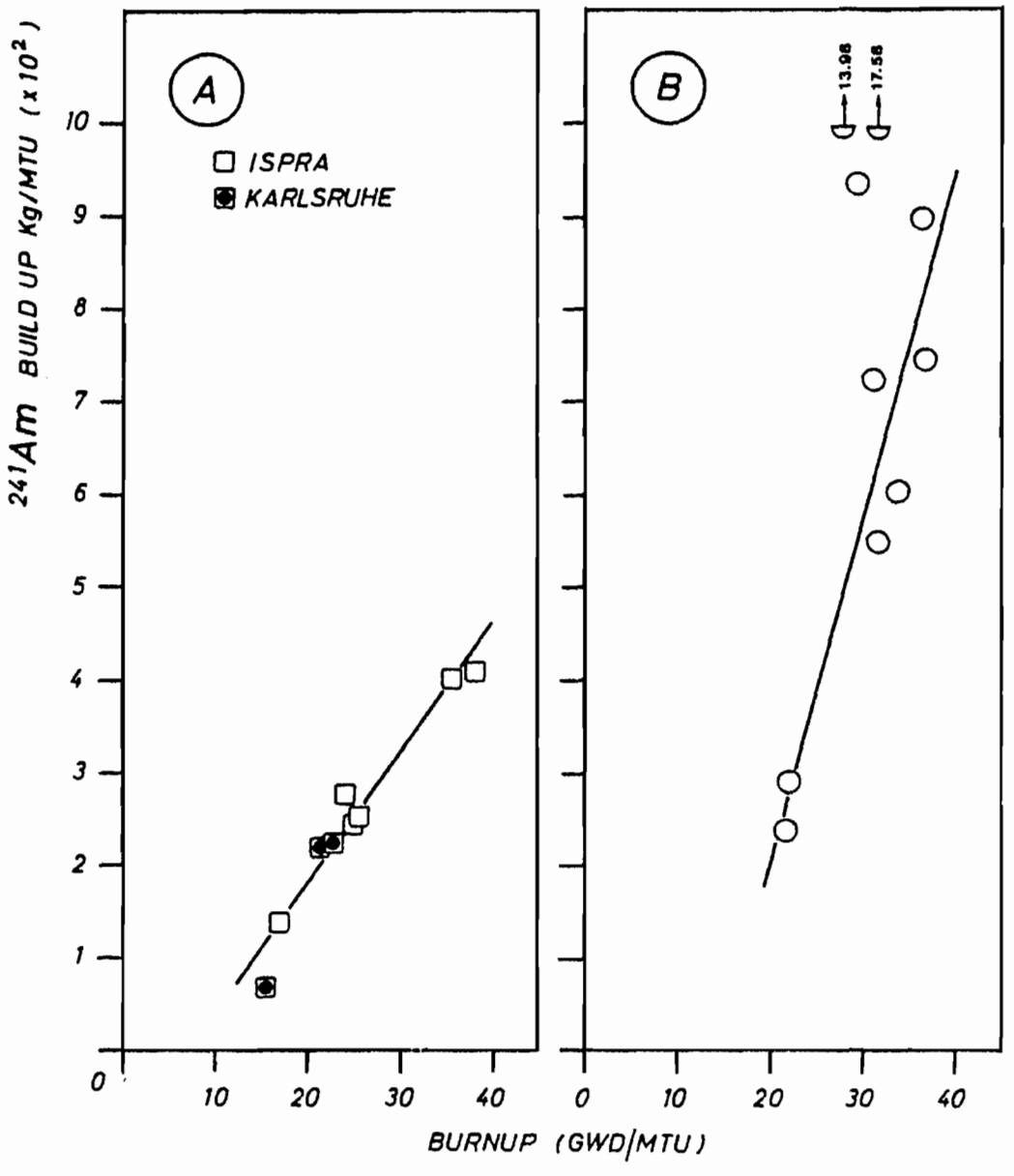


Fig. 4 - ^{241}Am build up plotted against burnup

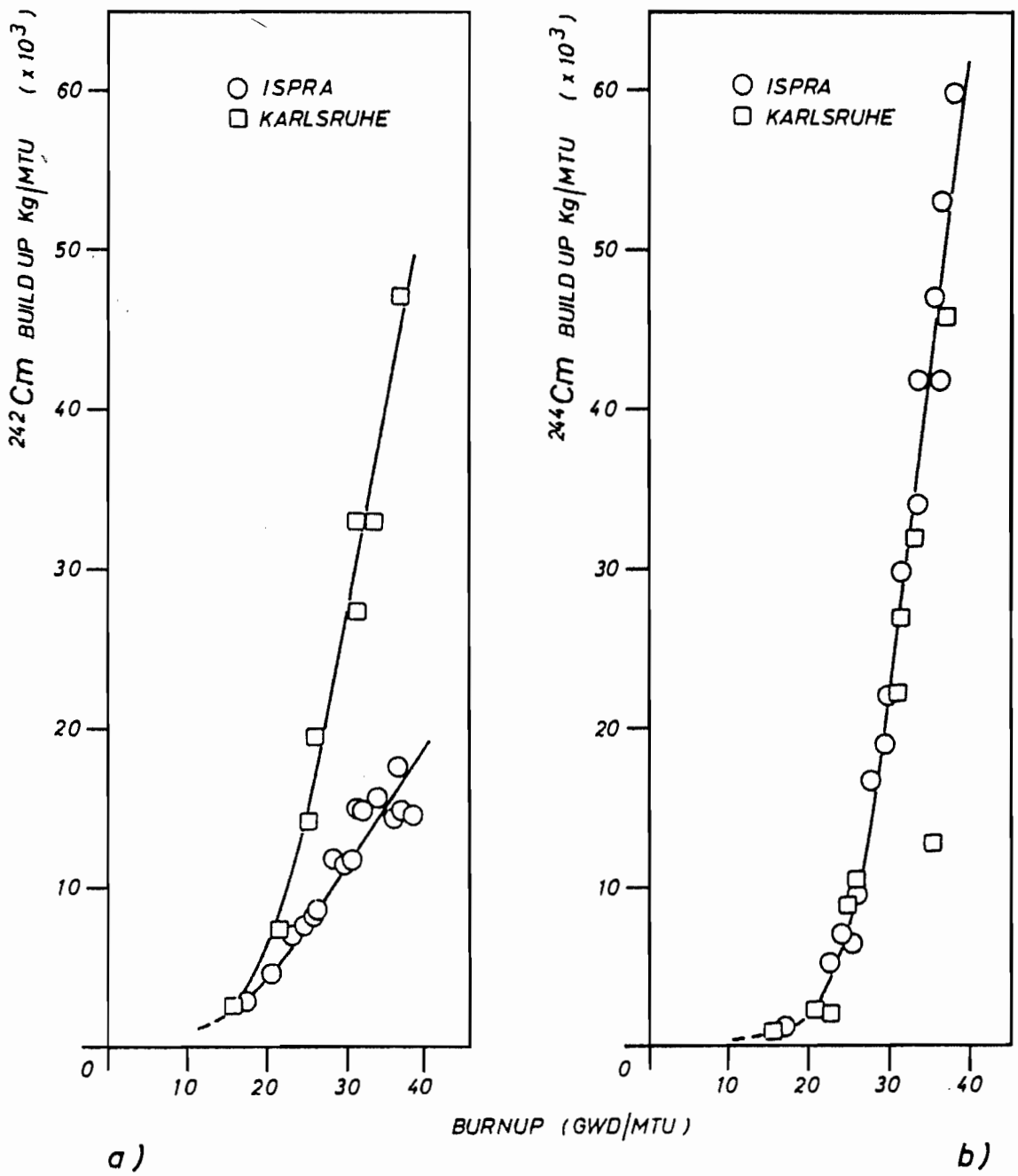


Fig. 5 - Cm isotopes build up plotted against burnup

Examples are given in Figs. 6 and 7.
In Fig. 6 the Pu/U mass ratio is plotted against the ^{235}U depletion.

$$D_5 = \frac{W_o^5 - W^5}{W_o^5}$$

where W_o^5 and W^5 are the percentage weight of ^{235}U in the fresh and irradiated fuel respectively. The different lines drawn show the different Pu production rates of the various fuel rods according to the radial position in the fuel assembly.
The experimental points belong to the BE 124 fuel element. The values coming from the G7 rod which was irradiated in asymptotic position are fitted on a stright line, while The points belonging to the other rods, irradiated in perturbed positions, are grouped on a different line.

In Figure 7 the ratio $^{240}Pu/^{239}Pu$ is plotted against $^{235}U/^{238}U$ for all analysed pellets.
This correlation appears to be less spectrum-dependent than the previous one. The experimental points aggregate on a curve and it appears to be almost impossible to separate them according to their radial position. This correlation is generally a function only of the initial enrichment.

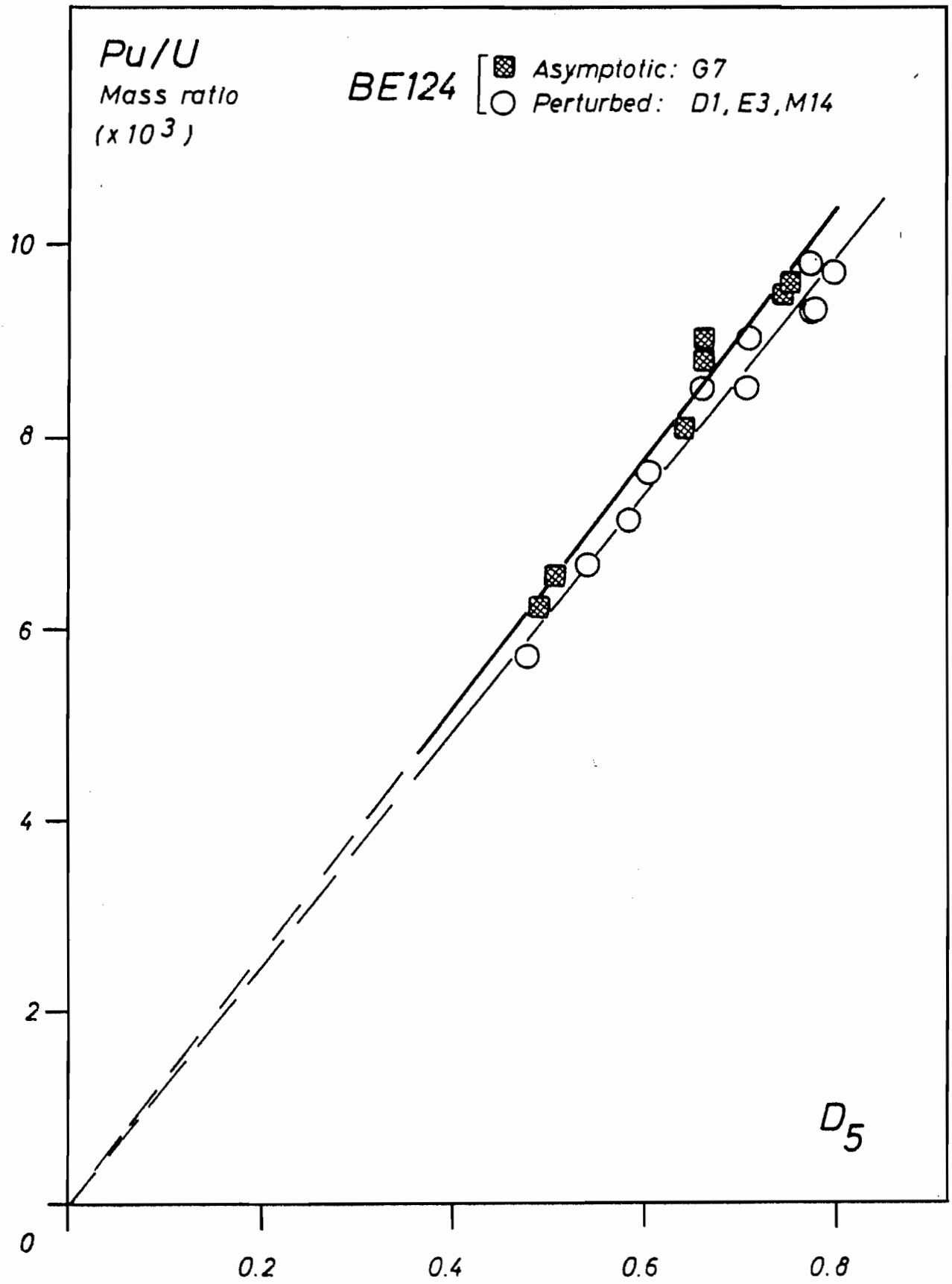


Fig. 6 - Correlation between Pu/U Mass ratio and ^{235}U depletion

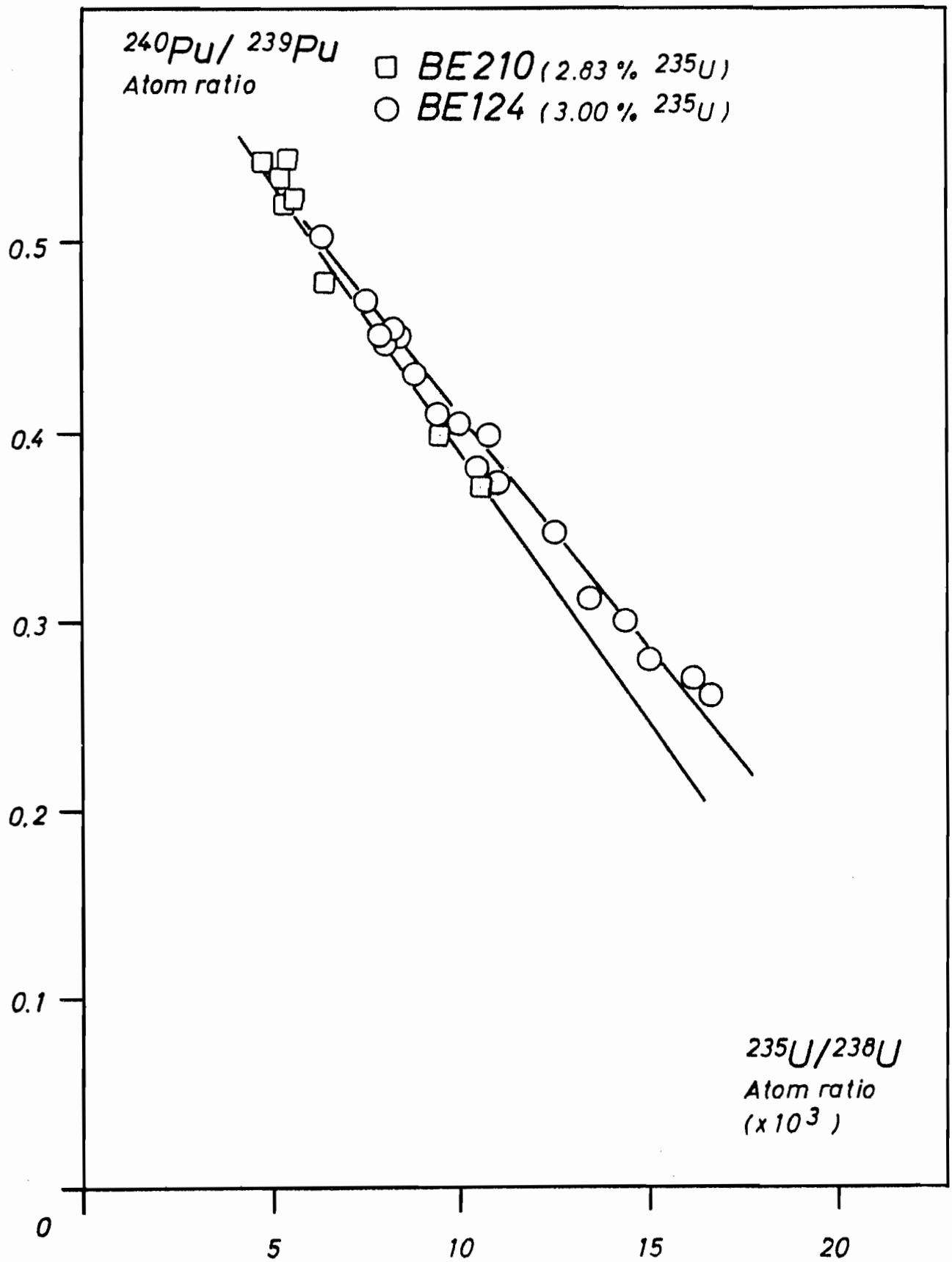


Fig. 7 - Correlation between the atom ratios $^{240}Pu/^{239}Pu$ and $^{235}U/^{238}U$

4. REFERENCES

1) A. ARIEMMA et al., "Experimental and Theoretical Determination of Burnup and the Heavy Isotope Content in a Fuel Assembly Irradiated in the Garigliano Boiling Water Reactor", EUR 4638 e. (1971)

2) A.M. BRESESTI et al., "Post Irradiation Analysis of Trino Vercellese Reactor Fuel Elements", EUR 4909 e. (1972)

3) P. BARBERO et al., "Post Irradiation Examination of the Fuel Discharged from the Trino Vercellese Reactor after the 2nd Irradiation Cycle", EUR 5605 e. (1976)

4) P. BARBERO et al., "Post Irradiation Analysis of the Gundremmingen BWR Spent Fuel", EUR 6301 e. (1978)

5) L. KOCH, G. COTTONE and H.W. GEERLINGS, "^{148}Nd Analyse zur Abbrandbestimmung von Kernbrennstoffen", Radioch. Acta, 10, 122 (1968)

6) B. BRANDALISE et al., "Automatic Evaluation of Isotope Analysis of Nuclear Fuels Including Isotope Dilution, Mass-Spectrometry, Alpha-Spectrometry and Isotope Correlation Technique with a Appropriate Databank", EUR 5669 en (1977)

7) W.J. MAEK et al., "Discrepancies and Comments Regarding ^{235}U and ^{239}Pu Thermal Fission Yields and the Use of ^{148}Nd as a Burn-Up Monitor", ICP-1092 (1976)

8) M.E. MEEK and B.F. RIDER, "Compilation of Fission Product Yields",
NEDO 12154-1 (1974)

9) M.F. JAMES, "Energy Release in Fission",
Journ. Nuclear Energy 23, 517-36, (1969)

10) R.J. NODVIK et al., "Supplementary Report on Evaluation of Mass Spectrometric and Radiochemical Analysis of Yankee Core 1 Spent Fuel, Including Isotopes of Elements Thorium through Curium",
WCAP 6086 (1969)

11) A. CRICCHIO, "Status and Requirements for Plutonium, Americium and Curium Nuclear Data Relevant to Plutonium Recycling in Light Water Reactors",
EUR 5698 e. (1977)

12) R. BERG et al., "Value and Use of Isotopic Correlations in Irradiated Fuels",
Proc. Symp. Practical Applications of R&D in the Field of Safeguards, 185 - Rome, 7-8 March, 1974

13) P. BRAND, A. CRICCHIO and L. KOCH, "Feasibility Study of the Use of Radioactive Fission Product Correlations for the Determination of Burnup and Heavy Isotopes Composition of BWR Dodewaard Fuel",
EUR 5141 e. (1974)

14) C. FOGGI and P. FRANDOLI, "Correlations Between Heavy Isotopes in Irradiated Fuels of Light Water Power Reactor",
EUR 5071 e. (1974)

15) C. FOGGI, F. FRENQUELLUCCI and G. PERDISA, "Isotopic Correlations Based on Fission Product Nuclides in LWR Irradiated Fuels. A Theoretical Evaluation",
IAEA - SM - 201, 44 (1975)

16) S. GUARDINI, G. GUZZI, "The Use of Isotopic Correlation Technique as Consistency Check and Elaboration of Post Irradiation Examination Data"., Proc. Symp. Isotopic Correlations and Their Application to the Nuclear Fuel Cycle - Stresa, 9-11 May 1978.

17) H. UMEZAWA et al., "Correlation of Amounts of Transplutonium Nuclides with Burn-up in the JPDR-1 Spent Fuel", Journ. Nucl. Science and Techn. 10(8), 489-92, August 1973.

18) S. GUARDINI, Personal Communication (1979).