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FEASIBILITY STUDY OF THE USE OF
RADIOACTIVE FISSION PRODUCT CORRELATIONS
FOR THE DETERMINATION OF BURNUP
AND HEAVY ISOTOPES COMPOSITION
OF BWR DODEWAARD FUEL

by

P. BRAND

(N.V. tot Keuring van Elektrotechnische Materialen Arnhem, the Netherlands)

and

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(Euratom)

1974



Joint Nuclear Research Centre
Karlsruhe Establishment - Germany

European Institute for Transuranium Elements

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ABSTRACT

Two fuel rods irradiated in the BWR Dodewaard power plant at 14 MWd/kg U were analyzed by non-destructive gamma spectrometric technique in the spent fuel pool of the Dodewaard plant in order to determine concentration ratios of selected fission products.

Subsequent destructive examination of seven pellet-size fuel samples of the two rods were performed at the laboratories of the European Institute for Transuranium Elements by gamma analysis and mass spectrometric isotope dilution analyses to determine the burnup (monitors: Nd-148 and Cs-137), heavy nuclide and radioactive fission product concentrations. The consistency of destructive results was checked by previously established correlations of fission product ratios versus burnup and heavy isotopes. It can be concluded that the ratio Cs-134/137 can be useful to determine burnup and heavy isotope composition of spent fuel.

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I. INTRODUCTION

The determination of burnup parameters of nuclear fuel, including the determination of the heavy isotopes composition, is of great importance for both the reactor operator and the safeguards authorities. There is still a need for a simple experimental method to obtain these burnup parameters. This report describes the results of an experimental investigation on irradiated fuel rods to determine the possibility of obtaining a non-destructive analysis method to be used in nuclear power plants. The analysis is based on the well-known nuclide correlation method, studied at the European Institute for Transuranium Elements, Karlsruhe (1).

The nuclide concentration ratio method has been preferred to the determination of single nuclide concentrations for the following reasons:

- Some isotopic ratios have been proven to be an excellent monitor for burnup and heavy isotopes measurements;
- The determination of the nuclide concentration ratios is based on the measurement of relative gamma intensities. Therefore some unknown parameters of the counting equipment (e.g. the absolute efficiency) is of no interest.

The investigation can be summarized as follows:

- Non-destructive gamma-spectrometrical analysis of two rods of an irradiated fuel assembly, which remained for cooling in the spent fuel pool of the Dodewaard plant;
- Destructive analysis of fuel samples of the two rods at the laboratories of the European Institute for Transuranium Elements (TUI) for the determination of burnup, heavy isotopes composition and isotopic ratio correlations;
- Comparison of the results of both analyses in order to determine the adequacy of the in-pool measuring method based on gamma-spectrometry.

Some nuclear data of the Dodewaard power station are summarized in section II.

The gamma-spectrometrical analysis will be described in section III and the destructive analysis performed at TUI is presented in section IV. The comparison of the results will be given in section V.

II. THE DODEWAARD NUCLEAR POWER PLANT

The reactor of the Dodewaard nuclear power plant is a direct cycle boiling-water reactor with natural circulation. In the reactor core 156 box-type fuel elements and 37 cruciform control rods are situated. A schematic cross-section of the core is given in Fig. 1 and the cross-section of a fuel assembly is shown in Fig. 2. From this last figure it can be seen that the fuel pin diameter is somewhat smaller than commonly used in most of the operating boiling-water reactors.

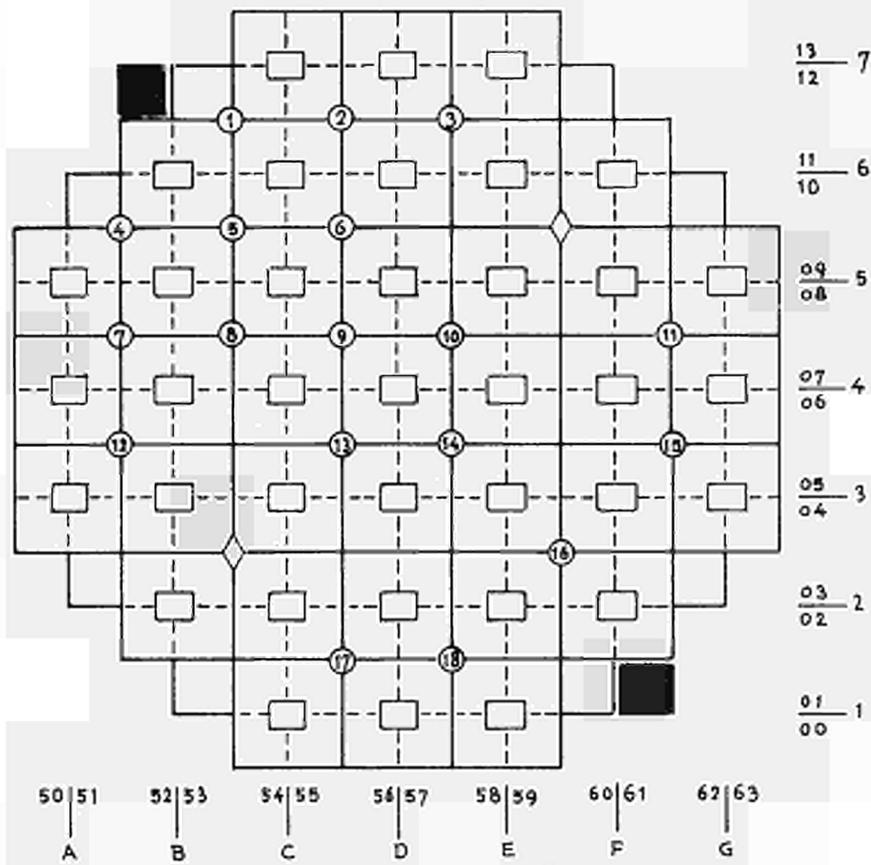
During the first part of the first cycle the over-reactivity of the core has partly been compensated by poison curtains (stainless steel blades with 5000 ppm boron), positioned in the narrow water gaps between the fuel elements.

Some nuclear core parameters are summarized in Table II.1.

Thermal power	163.4	MW
Primary steam flow	256000	kg/hr
Pressure at core top	70	bar
Number of fuel assemblies	156	
Number of control rods	37	
Effective fuel length	1793	mm
Number of fuel rods/assembly	36	
Rod clad O.D.	13.49	mm
UO ₂ pellet O.D.	11.48	mm
Zr-2 clad thickness	0.89	mm
Average assembly steam volume fraction	0.32	
Maximum steam volume fraction at channel exit	0.64	
Initial U-235 enrichment	2.5	w/o

Table II.1. Nuclear parameters of the Dodewaard reactor.

From the date of start of operation - 4th October 1968 - the reactor was on power during the first cycle for 798 days. The power history of the first cycle is schematically presented in Fig. 3.



-  Control rod
-  Instrumentation channel
-  Start-up monitor
-  Neutron source

Fig.1 Schematic cross-section of the core of the Dodewaard reactor

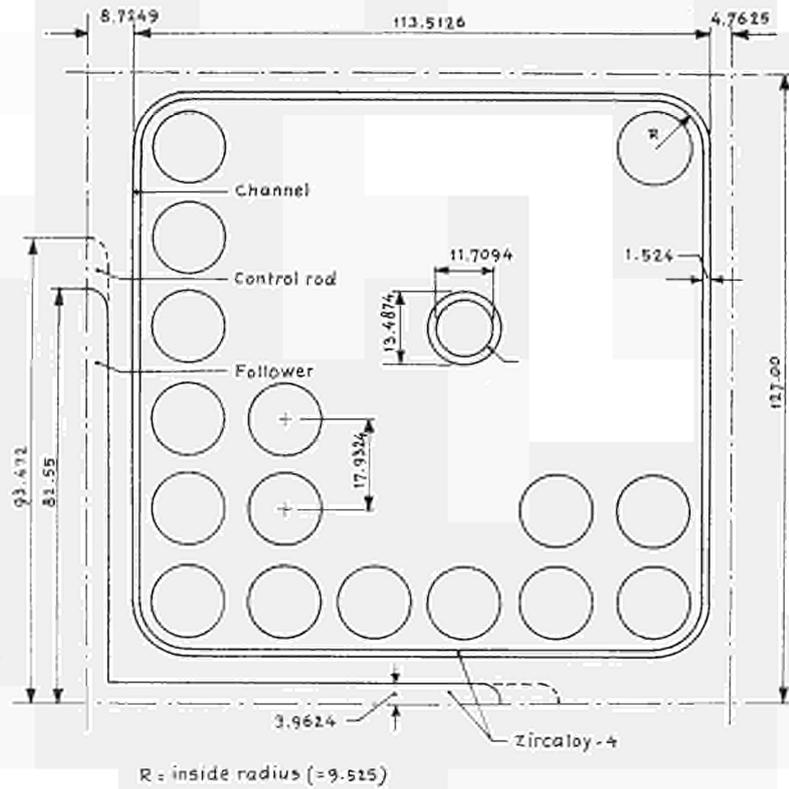


Fig.2 Schematic cross-section of the Dodewaard fuel assembly
(dimensions in mm)

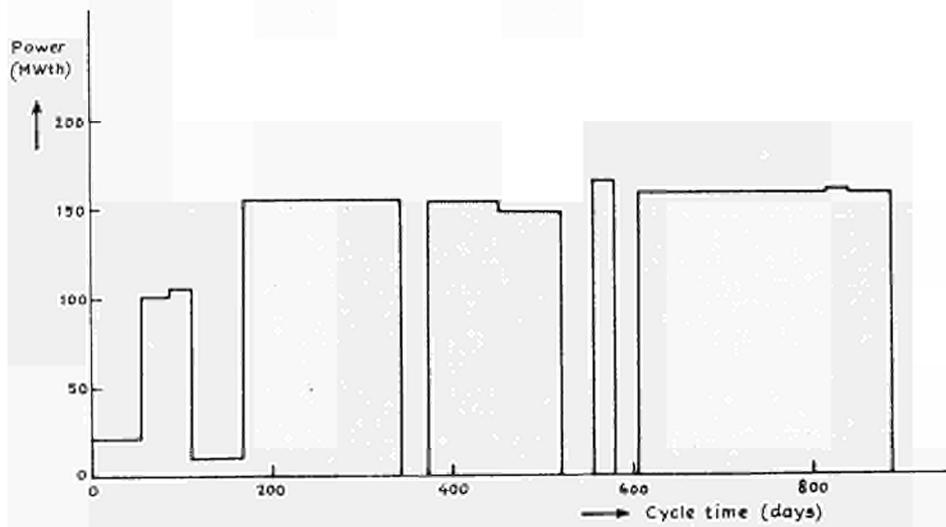


Fig.3 Power history of the Dodewaard nuclear power plant (First cycle)

III. NON-DESTRUCTIVE ANALYSIS

Two fuel rods of one of the four central fuel assemblies (see Fig. 1) were selected for an extensive program of post-irradiation examinations on physical and metallurgical properties. The position of the rods in the fuel assembly is shown in Fig. 4. The average burnup of the total fuel bundle was 14 MWd/kg U, the peak rod burnup was calculated to be 16.0 MWd/kg U and the peak pellet burnup was 20.2 MWd/kg U.

III.1. Procedure

III.1.1. Detector system

In the fuel pool of the Dodewaard reactor a gamma-scan installation was constructed for the determination of power and burnup distributions of fuel elements and fuel rods. A schematic cross-section of the installation is shown in Fig. 5. A fuel assembly or a fuel rod can be moved vertically by a jib crane.

The position of the source relative to the collimator axis can be read on the refuelling floor.

In the concrete wall of the fuel pool a hole (15 cm diameter and 115 cm long) was drilled in order to position the combination of detector, collimators and the lead discs for gamma-attenuation purposes. A NaJ scintillation detector will mostly be used for gamma-scanning, for gamma-spectrometry a GeLi semiconductor detector within a lead shielding can be installed. The important data with regard to the semiconductor detector system are summarized in Table III.1.

Detector	GeLi, planar, 2.8 cm ³
Collimators	cylindrical, 50/20/12 mm
Gamma attenuation	lead, 4.5 cm
Resolution	4.1 keV for Co-60
Dead time correction of multi-channel analyzer	< 10 %
Stability	< .1 % over 15 days
Channel width	2.368 keV

Table III.1. Gamma-spectrometry detector system.

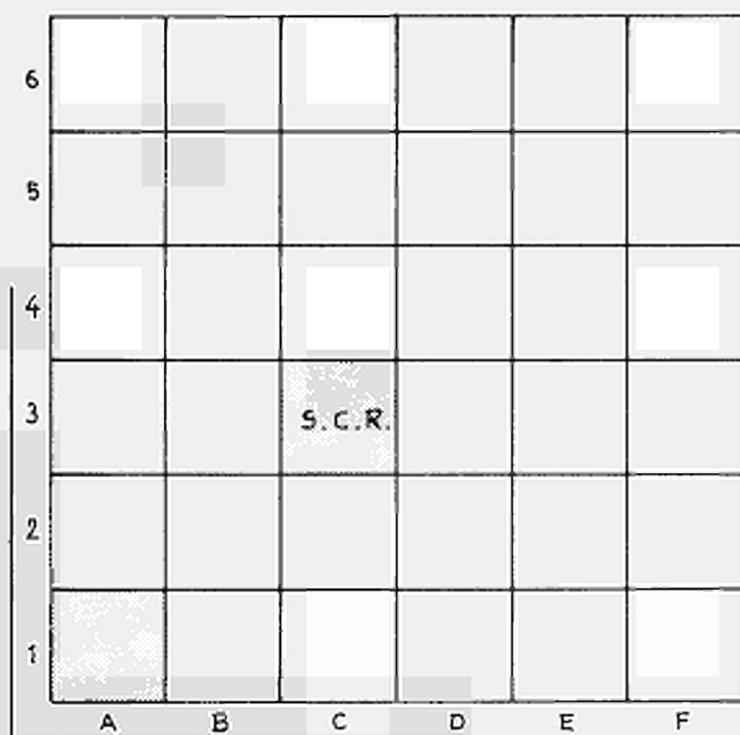


Fig.4 Position of the fuel rods selected for analysis
S.C.R.: Spacer Capturing Rod

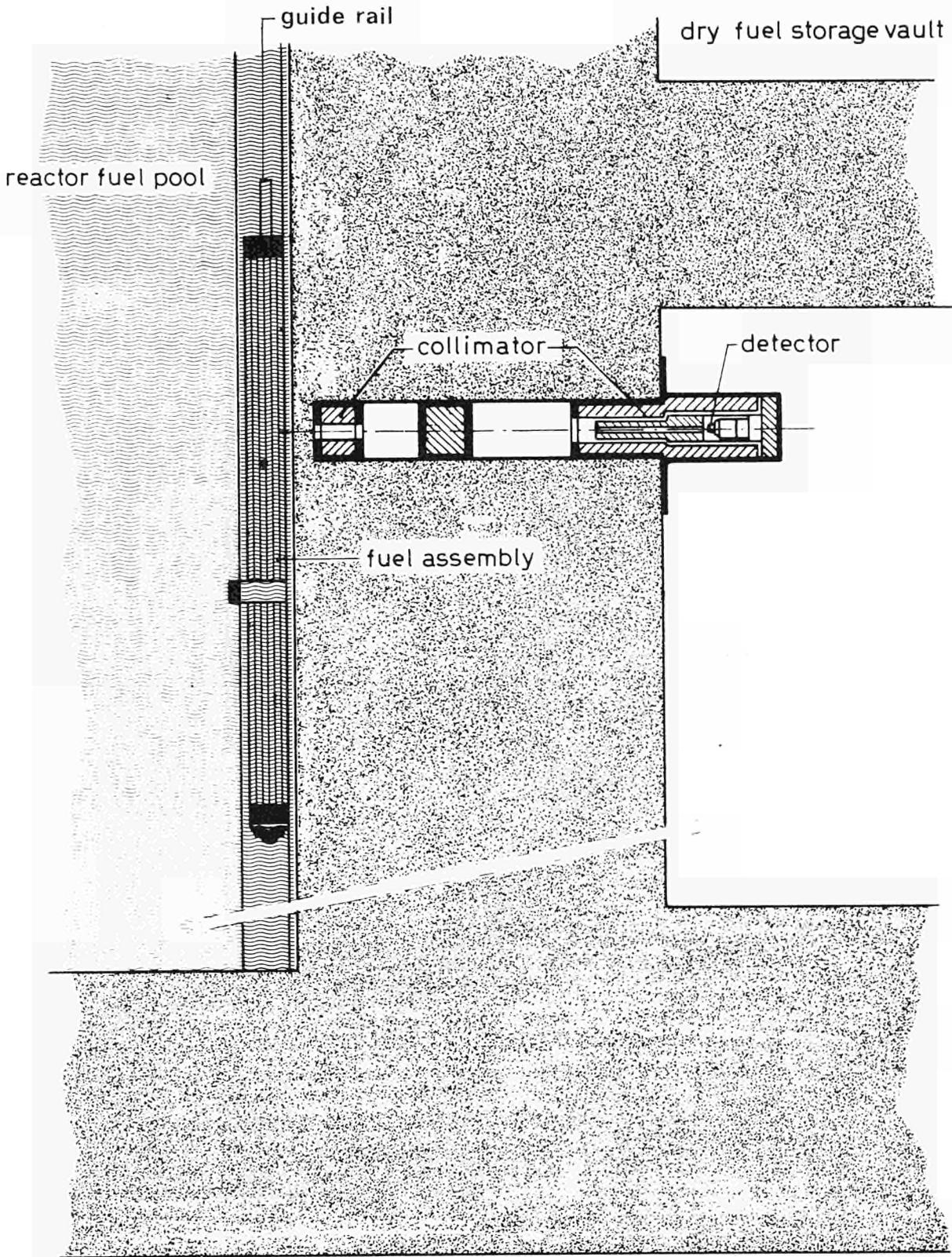


Fig.5 Schematic presentation of γ - scan facility

The main amplifier and the 1024 channel multi-channel analyzer were installed outside the reactor building to prevent the possibility of contamination of the equipment. A shielded signal cable of 120 m was used to connect the detector system and the pre-amplifier with the multi-channel analyzer.

III.1.2. Fuel rod translation system

For this experiment the corner rod, adjacent to the control rod, and the spacer capturing rod were selected. The fuel rod is positioned in the centre of the fuel rod-holder. This rod-holder can be moved vertically by the crane. The rod-holder is enclosed by a guide rail, installed in the fuel pool. The axis of the guide rail has been adjusted to the collimator axis, using a strong gamma source. The axial position of the fuel rod relative to the collimator axis was calibrated by measuring the local decreases of the Nb-95 gamma intensity due to the connector plugs of the spacer capturing rod (Fig. 6). From this measurement and the well-known distances of the connector plugs the axial position of the fuel rods could be derived with an accuracy of 4 mm.

III.1.3. Fuel sampling

The axial positions of the fuel samples are given in Table III.2. The fuel height is given with respect to the lower end of the active fuel column.

Label	A-15	A-16	A-17	A-18	C-16	C-17	C-18
Fuel rod (see Fig.4)	A-1	A-1	A-1	A-1	C-3	C-3	C-3
Fuel height (cm)	16.8	72.8	160.8	104.8	72.8	172.8	16.8

Table III.2. Axial position of the fuel samples.

The reasons for this selection are the following:

- from gamma-scanning of the rods large differences in burnup can be expected;
- the influence of the steam void fraction may be apparent because of the selection of positions with high and low moderator densities;

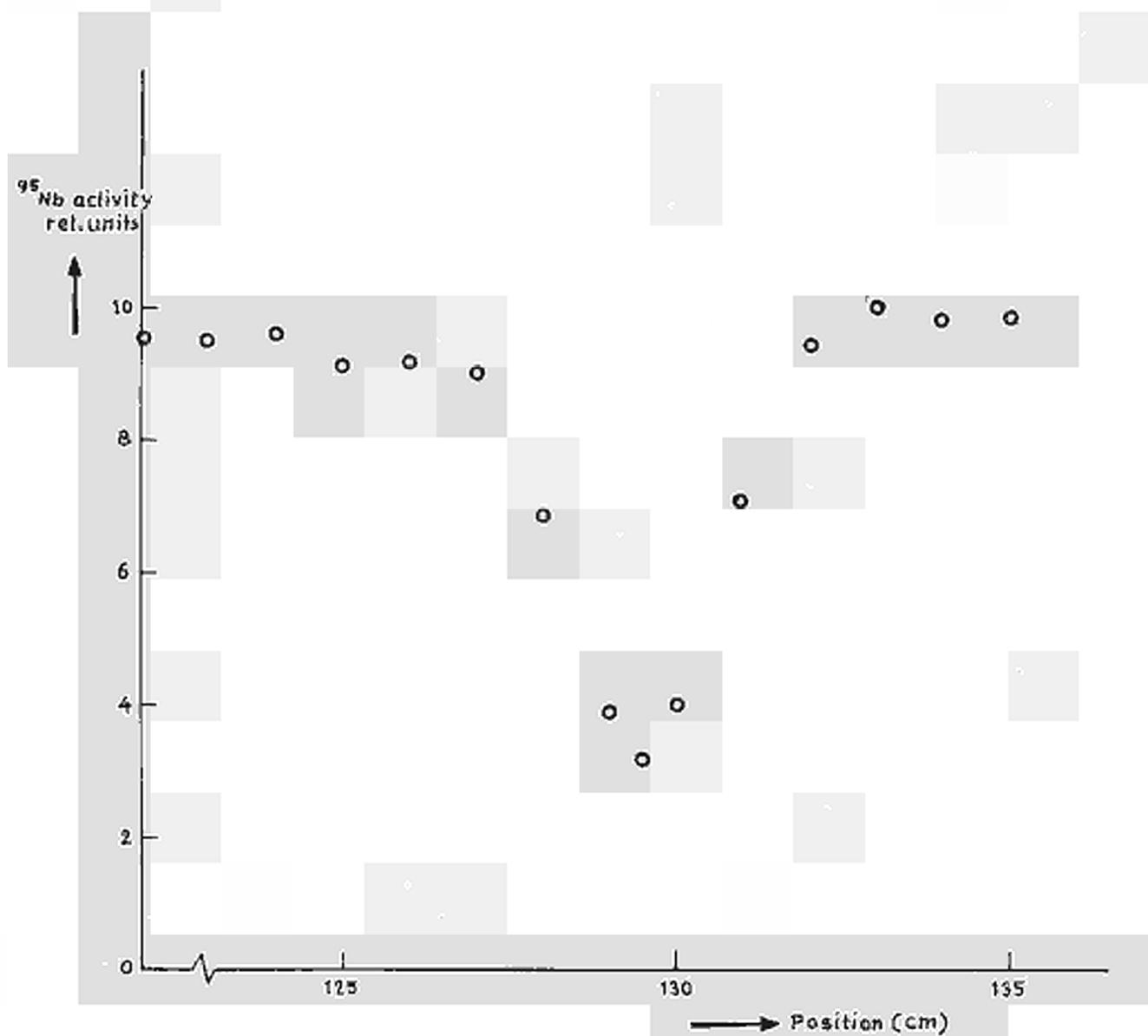


Fig.6 ⁹⁵Nb activity as a function of the position at the location of a core for plug of the spacing rod

- the influence of the control rod may be seen since the lower part of the fuel rods were longer adjacent to a control rod than the upper part;
- a possible influence of the spacers will be avoided;
- interference with the sampling for physical and metallurgical hot-cell investigations had to be avoided.

III.2. Data processing

III.2.1. The gamma spectrum

A typical gamma spectrum, obtained with the equipment described above, is presented in Fig. 7. From this figure it can be seen that the response to low-energy gammas is small in comparison with the response to higher gamma energies ($> .5$ MeV). The cause of this distortion of the gamma spectrum is the absorption in the different materials between source and detector (see Table III.3.).

Absorbers between source and detector	Water	Stainless Steel	Lead	Concrete
Absorber thickness (cm)	approx. 12	2.8	4.5	approx. 8

Table III.3. Absorbers between source and detector.

III.2.2. Evaluation of the spectral data

The gamma spectra were evaluated by the computer program SANA (2) which program is based on the GASPAN code (3). SANA computes the peak integral by fitting the peak to a gaussian function and integrating after subtracting a best fit to the background and the Compton continuo. From the resulting values of the peak integrals combined with an evaluation of possible gamma emitting isotopes, it follows that only a few isotopes can possibly be used for further analysis:

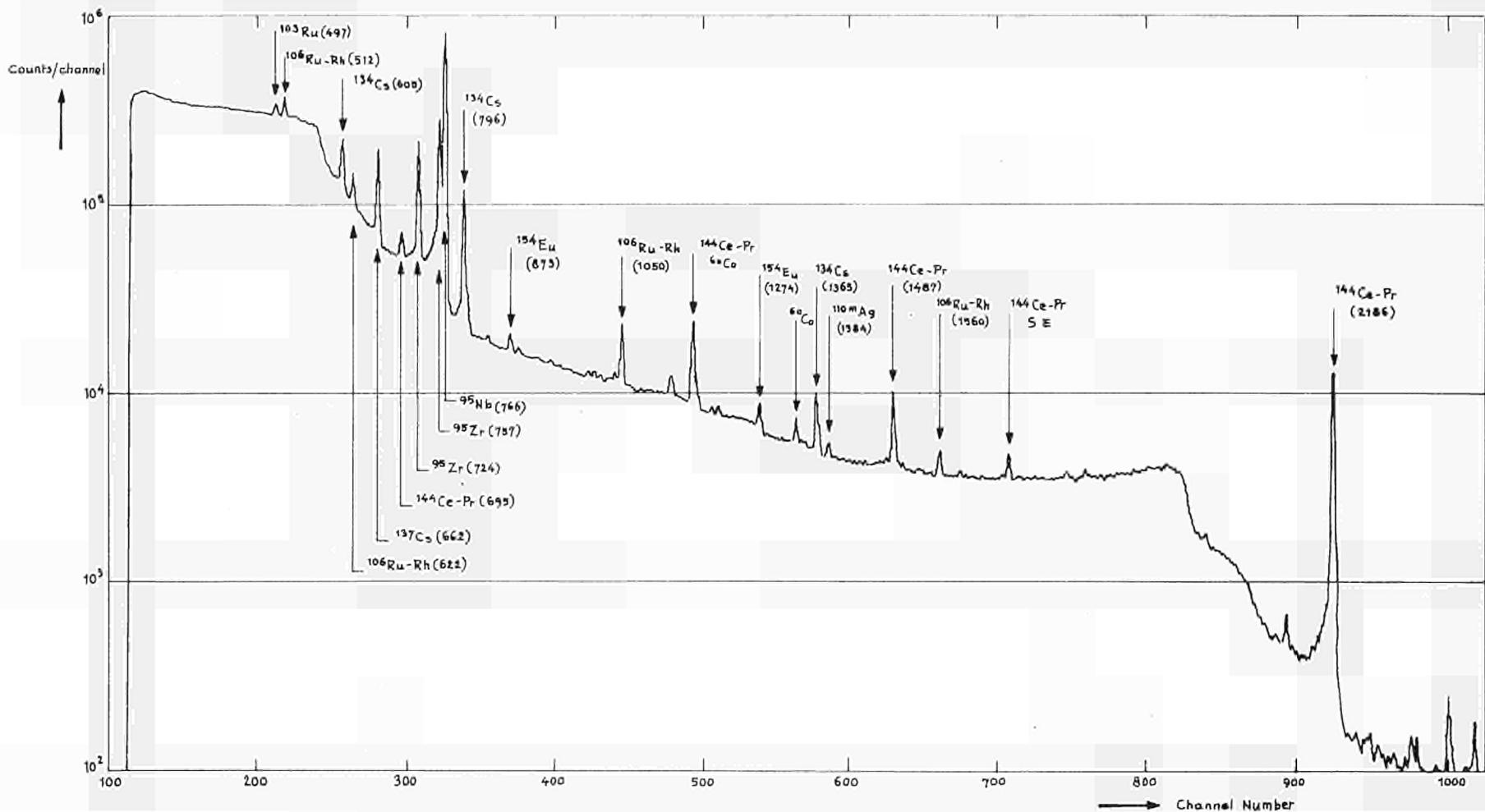


Fig.7 Gamma spectrum of a fuel rod, measured at Dodewaard BWR power station

- Ru-Rh-106
- Cs-134
- Cs-137
- Ce-Pr-144
- Eu-154
- Zr-95

From these, the nuclide Zr-95 is not only a fission product but also an activation product in the Zircaloy cladding. Therefore it is not a useful nuclide for in-pool fission product analysis.

In order to derive the nuclide concentration ratios from the count rates, the following formula has to be applied:

$$R_{i,j,k,l} = \frac{A_{i,k} \cdot F_i \cdot \lambda_j \cdot \epsilon_{j,l} \cdot \alpha_{j,l}}{A_{j,l} \cdot F_j \cdot \lambda_i \cdot \epsilon_{i,k} \cdot \alpha_{i,k}} \quad (1)$$

where

$R_{i,j,k,l}$: ratio of concentration of nuclides i and j, based on gamma energies k and l respectively.

$A_{i,k}$ ($A_{j,l}$) : count rate of nuclide i (j) for gamma energy k (l).

F_i (F_j) : decay correction factor for in-pile and out-of-pile decay for nuclide i (j).

λ_i (λ_j) : decay constant of nuclide i (j).

$\epsilon_{i,k}$ ($\epsilon_{j,l}$) : relative efficiency of the counting equipment for gamma energy k (l) of nuclide i (j).

$\alpha_{i,k}$ ($\alpha_{j,l}$) : branching ratio of the gamma transition of nuclide i (j) with energy k (l).

The decay correction factor F can be calculated as follows:

$$F = \frac{\lambda \cdot e^{\lambda \theta} \cdot \sum_{p=1}^n T_p}{\sum_{p=1}^n (1 - e^{-\lambda T_p}) \cdot (e^{-\lambda t_p})} \quad (2)$$

where

θ : time elapsed from shut-down of the reactor to the measurement

n : number of reactor cycles

T_p : irradiation time of cycle p

t_p : time elapsed from end of cycle p to end of cycle n .

For the calculation of the decay correction factor a constant neutron flux during power operation has been assumed.

The efficiency of the counting equipment for a specific gamma line of a nuclide can be written as follows:

$$\epsilon_{i,k} = \eta_k \cdot \mu_k \cdot \nu_{i,k} \quad (3)$$

where

η_k : detector intrinsic efficiency for gamma energy k

μ_k : gamma attenuation for energy k caused by the absorbing materials between source and detector

$\nu_{i,k}$: gamma self-absorption in the fuel rod for gamma energy k of nuclide i .

The efficiency factor $\epsilon_{i,k}$ can also be expressed as the product of two functions:

$$\epsilon_{i,k} = H(d) \cdot G(E) \quad (4)$$

where $H(d)$ is only dependent on the distribution of the nuclide within the fuel rod and $G(E)$ is only energy-dependent. If the fission products distribution in the fuel pellets is the same for the nuclides considered, then only the energy-dependent factor has to be considered in the evaluation of the nuclide concentration ratios. For nuclides with the same atomic number (isotopes) this assumption is justified in case these isotopes stem either directly from fission or from short-living parents.

In all other cases possible effects of migration have to be considered carefully.

Table III.4. presents the data used for the calculation of the in-pile and out-of-pile decay correction factor.

In Table III.5. some nuclear data of the nuclides of possible interest are given.

T_p	p = 1,2,3,4 : 343 d, 148 d, 25 d, 282 d						
t_p	p = 1,2,3,4 : 545 d, 367 d, 208 d, 0 d						
Sample	A-15	A-16	A-17	A-18	C-16	C-17	C-18
θ (days)	207	206.5	205.5	220	213	218	213

Table III.4. Data used for the calculation of the decay correction factor.

Nuclide	γ -Energy (MeV)	Branching-ratio (%)	Decay-constant (days ⁻¹)
Zr-95	0.7242	44.2	1.058×10^{-2}
	0.7568	54.5	
Ru-Rh-106	0.6221	9.88	1.882×10^{-3}
	1.050	1.51	
	1.128	0.399	
	1.562	0.157	
Cs-134	0.6046	97.5	9.254×10^{-4}
	0.7958	85.4	
	1.3652	3.31	
Cs-137	0.6616	84.6	6.307×10^{-5}
Ce-Pr-144	0.6965	1.51	2.436×10^{-3}
	1.4891	0.29	
	2.1857	0.74	
Eu-154	0.5917	4.9	1.186×10^{-4}
	0.7233	19.7	
	0.8732	11.7	
	0.9963	10.1	
	1.0048	17.4	
	1.2745	34.7	
	1.5964	1.67	

Table III.5. Nuclear data of some fission and activation products.

In order to obtain the relative efficiency as a function of energy, the three main gamma lines of Cs-134 have been selected for the following reasons:

- Cs-134 has high values for the branching ratios;
- the count rates of the three gamma lines are sufficient;
- of the three lines only the 604.6 keV line may slightly be disturbed (approximately 1%) by the gamma lines of Sb-Te-125 (606.8 keV and 600.7 keV);
- of the nuclides of interest only in one case the isotopic ratio can be determined, i.e. Cs-134/Cs-137. Thus the above mentioned assumption of a constant value of the function $H(d)$ may be justified (both isotopes do not have long lived parents).

For each of the seven measurements the calculated results of the three peak integrals of Cs-134 are divided by the corresponding branching ratio and normalized to the value of the 604.6 keV line. Because the self-absorption, the attenuation factor and the detector efficiency are approximately exponential functions, the relative efficiency is assumed to be a function of the following form:

$$\epsilon(E) = A (1 - B e^{-CE}) \quad (5)$$

where E is the energy and A , B and C are constants.

The function $\epsilon(E)$ has been calculated for each of the measurements in order to compensate for the possible fluctuations of the attenuation factor, due to the deviations of the fuel rod position. The function is determined numerically using the regula falsi. The efficiency function for one of the samples is shown in Fig. 8.

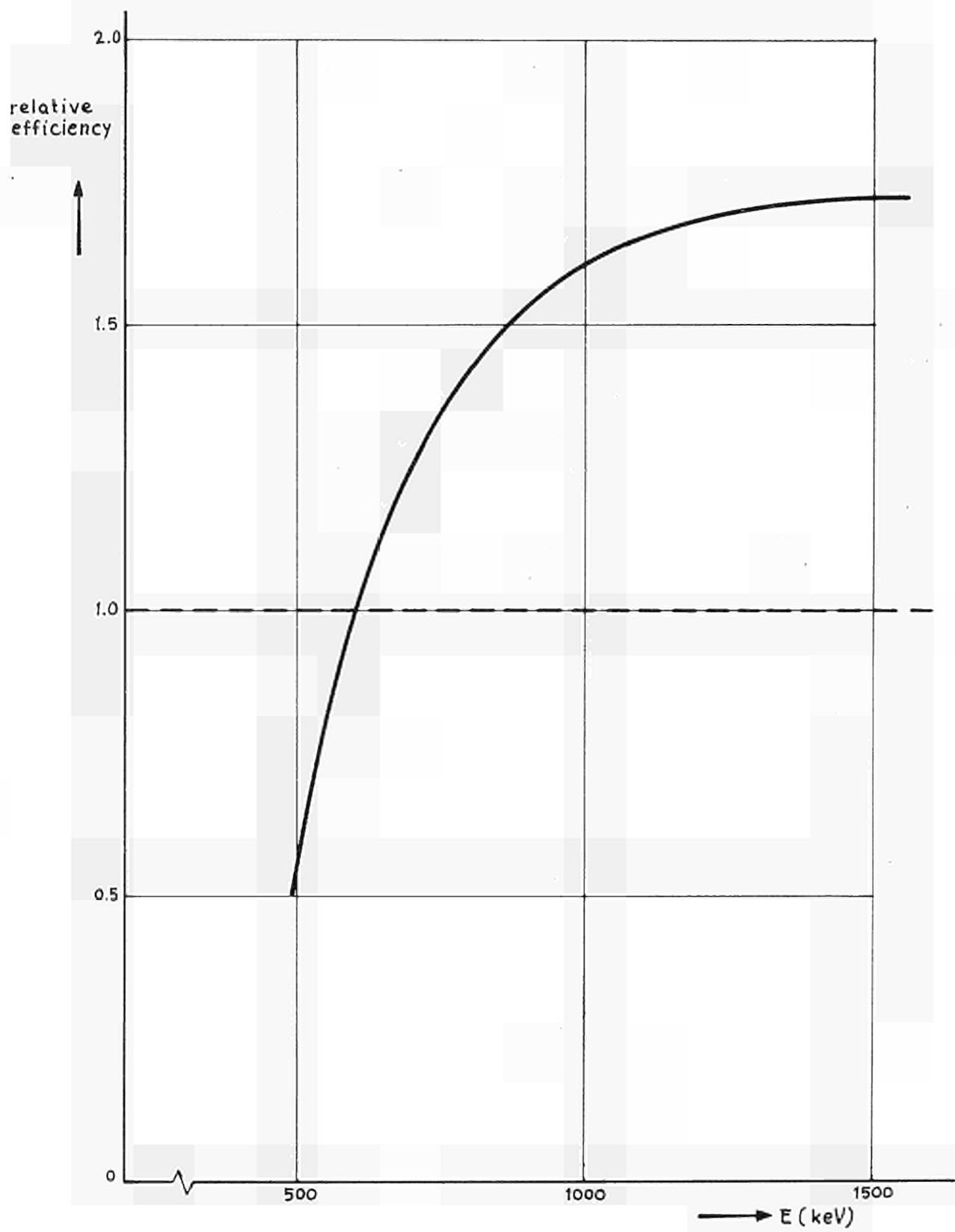


Fig. 8 Relative efficiency as a function of energy (Sample C-16)

III.2.3. Results

The results of the computation of the peak integrals by SANA for the gamma-lines considered, are presented in Table III.6.

The calculated relative efficiency for the different samples and for the gamma energies considered are presented in Table III.7. The value for the 604.6 keV line is, of course, 1.0.

The results of the computation of the nuclide concentration ratios, using equation (1), are given in Table III.8. Because of the computing method of the efficiency function, the ratios of Cs-134/Cs-137 will deliver the same values regardless which of the gamma lines of Cs-134 has been used. The consistency of the method of deriving nuclide ratios with a relative efficiency function can be checked by considering two gamma lines of an isotope, which have not been used to determine the function. For this reason the two values of the nuclide ratio of Ru-Rh-106/Cs-137 are given.

From Table III.8. it can be seen that:

- the values of the ratio Cs-134/Cs-137 show large differences for some of the samples. It can be concluded that in case this ratio will be used, it may be a sensitive burnup monitor;
- the values of the ratios of Ru-Rh-106/Cs-137 for both gamma energies show a fair agreement.

Sample	Counting time (s)	Calculated count rate (s ⁻¹)						
		Cs-137	Cs-134	Cs-134	Cs-134	Ru-Rh-106	Ru-Rh-106	Ce-Pr-144
		661.6 keV	604.6 keV	795.8 keV	1365.2 keV	622.1 keV	1050 keV	696.5 keV
A-15	20000	9.6101	5.5119	7.0510	0.3615	3.2315	0.7693	1.4176
A-16	20000	10.4268	8.7163	11.0569	0.5522	3.8888	0.9743	1.4172
A-17	20000	9.6223	5.8344	7.0869	0.3752	2.9289	0.7181	1.2802
A-18	4000	10.3160	8.2903	9.7765	0.5153	3.5885	0.9018	1.3628
C-16	20000	8.0126	5.9138	7.3760	0.3444	2.8604	0.6586	1.0399
C-17	20000	3.4704	1.0802	1.3695	0.0710	0.7984	0.2118	0.4908
C-18	20000	8.0696	4.1499	5.2405	0.2577	2.4442	0.6018	1.2253

Table III.6. Results of the calculation of the peak integrals.

Energy (keV) Sample	661.6 Cs-137	795.8 Cs-134	622.1 Ru-Rh-106	1050 Ru-Rh-106	696.5 Ce-Pr-144
A-15	1.1668	1.4605	1.0542	1.7705	1.2553
A-16	1.1653	1.4483	1.0544	1.7309	1.2518
A-17	1.1338	1.3868	1.0430	1.6953	1.2073
A-18	1.1184	1.3464	1.0379	1.6345	1.1841
C-16	1.1657	1.4240	1.0554	1.6383	1.2483
C-17	1.1601	1.4474	1.0520	1.7626	1.2459
C-18	1.1648	1.4417	1.0543	1.7084	1.2502

Table III.7. The calculated results of the total relative efficiency for all samples and for some gamma-energies.

Sample	Cs-134/Cs-137	Ru-Rh-106/Cs-137 622.1 keV	Ru-Rh-106/Cs-137 1050 keV	Ce-Pr-144/Cs -137 696.5 keV
A-15	.06764	.3114	.2888	.7693
A-16	.09841	.3445	.3440	.7091
A-17	.06939	.2761	.2725	.6985
A-18	.09186	.3211	.3352	.7220
C-16	.08741	.3335	.3237	.6898
C-17	.03684	.2165	.2243	.7584
C-18	.06086	.2830	.2814	.8051

Table III.8. Isotopic concentration ratios.

IV. DESTRUCTIVE ANALYSES

Destructive analyses were performed on seven pellet-size fuel samples in order to determine:

- the concentration of heavy isotopes;
- the content of Nd-148 in order to evaluate the burnup;
- the isotopic composition of the retained fission gases;
- the concentration of selected radioactive fission and neutron capture nuclides.

The sequence of the analyses performed on the samples is illustrated by the flow diagram in Fig. 9. The procedures utilized for the analyses are briefly described below; more detailed information can be found in reference (4).

IV.1. Procedure

a) Dissolution of the samples and fission gas analysis

The dissolution of the samples was effectuated in hot cells by means of concentrated HNO_3 .

The fission gases krypton and xenon, released during the dissolution, were collected, washed and analyzed using a mass spectrometer of the type CH_4 , Varian-MAT. The fuel solutions were diluted to about 0.24 mg/g so that they could be handled outside the hot cells. Aliquots of these solutions were then subjected to mass, alpha and gamma spectrometry.

b) Analysis of heavy nuclides

The heavy isotopes U-235, U-236, U-238, Pu-239, Pu-240, Pu-241 and Pu-242 were measured by means of the mass spectrometric isotope dilution technique. For this purpose uranium and plutonium were separated from interfering materials by sorption on an anion exchanger and by washing with 8 M HNO_3 . Excessive uranium was removed in order to get about the same quantities of uranium and plutonium for the subsequent mass spectrometry.

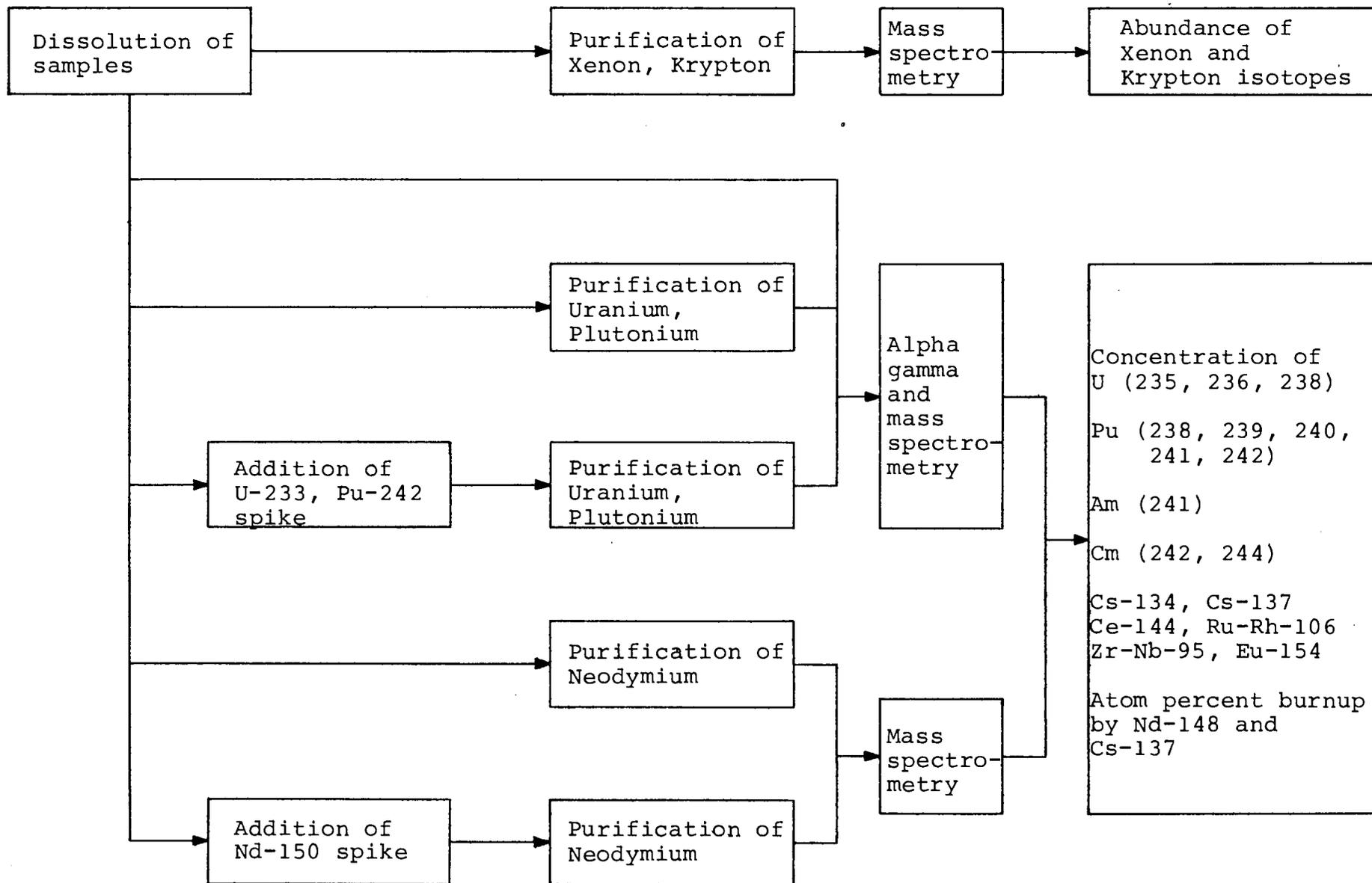


Fig. 9: Flow diagram of post-irradiation examination.

For isotopic dilution U-233 and Pu-242 spikes were used after double calibration against standards of the National Bureau of Standards.

The less abundant heavy isotopes Pu-238, Am-241, Cm-242 and Cm-244 were analysed by alpha spectrometry.

c) Nd-148 analysis

Mass spectrometry combined with isotope dilution analysis was also used for Nd-148 determination. Neodymium was purified as the α -hydroxoisobutyric acid complex by chromatographic elution from a cation exchanger. Nd-150 was used as spike and its calibration was performed against a natural neodymium standard of the BCMN, Geel.

d) Analysis of radioactive fission nuclides

Gamma spectrometry was performed on an aliquot of diluted fuel solution in order to evaluate the concentration of Cs-134, Cs-137, Ce-144, Ru-Rh-106, Zr-Nb-95 and Eu-154. A weighed amount of each fuel solution was counted three times with a Ge-Li detector connected, via a preamplifier and amplifier, to a 4096 channel analyzer. The absolute efficiency of the detector was determined by means of I.A.E.A. standard sources.

IV.2. Data processing and results

The experimental data coming out from alpha, mass and gamma spectrometric measurements were evaluated by means of appropriate computer programs.

Alpha, mass and gamma spectra, stored on paper punched tapes were handled by the programs ALPHASPEC (5), DATA-DATA-2 (5) and GASPAN (3).

The results of these evaluations are further reduced by the BURNUP program (5) which gives the concentration of heavy and fission nuclides and the burnup by Nd-148 and Cs-137.

The results and a short description of the data processing procedure for each type of measurement are given below.

a) Alpha_spectrometry

The area of alpha peaks is evaluated by the program ALPHASPEC on the basis of an empirical method which partially takes into account the overlapping of tails. The alpha decay energies partly overlap, so that only the activity ratios $(\text{Pu-238} + \text{Am-241})/(\text{Pu-239} + \text{Pu-240})$, $\text{Cm-242}/(\text{Pu-239} + \text{Pu-240})$ and $\text{Cm-244}/(\text{Pu-239} + \text{Pu-240})$ can be determined. Consequently, a measurement of the activity ratio $(\text{Pu-239} + \text{Pu-240})/\text{Pu-238}$ in the samples separate from Am-241 and Cm-242,244 is necessary.

The mean value of 3 reported measurements together with the single standard deviation of the percent alpha activity before and after separation are given for each sample in Table IV.1.

The concentration of the single nuclide was consequently computed by the BURNUP program using the ratio Pu-239/Pu-240 determined by mass spectrometry.

b) Mass_spectrometry

The DATA-DATA-2 computer program evaluates mass spectra by computing the peak heights by a least square fit through the peak top range. Correction for ion current decrease is accomplished by a linear interpolation to the starting point of several scans. The isotopic ratios are then calculated from the peak heights in respect to the most abundant isotope. The isotopic ratios of the fission gases Kr and Xe related to the isotopes Kr-86 and Xe-134 are listed in Table IV.2. The results of the isotopic ratios of uranium, plutonium and neodymium are presented in Table IV.3.

In addition, the program BURNUP performs the calculations for the isotope dilution analysis and burnup.

The results given by this program are:

- the isotopic composition of uranium, plutonium, americium and curium referred to the initial heavy atoms. The Pu-241

Sample	Date of measurement	Before separation				After separation	
		Pu-239+240 %	Pu-238+Am-241 %	Cm-242 %	Cm-244 %	Pu-239+ 240 %	Pu-238 %
A-15	08.18.72	26.57±0.34	27.56±0.37	3.25±6.18	42.62±0.48	57.81±1.13	42.19±1.55
A-16	08.24.72	18.34±1.31	27.08±0.91	7.86±2.51	46.73±0.52	43.83±1.05	55.06±0.97
A-17	08.24.72	25.59±0.73	26.58±1.49	4.59±6.66	43.24±0.16	53.65±0.10	45.24±0.75
A-18	08.24.72	19.17±0.24	27.29±0.16	7.39±2.94	46.14±0.58	45.69±0.01	54.31±0.92
C-16	08.23.72	20.38±0.47	29.07±0.23	5.41±0.94	45.14±0.25	46.19±0.26	53.03±0.53
C-17	08.18.72	51.23±1.59	25.45±4.10	-	23.31±2.31	74.63±1.05	25.37±3.08
C-18	08.23.72	33.89±1.59	28.61±0.39	1.36±1.68	36.15±1.24	60.85±0.82	38.64±0.63

Table IV.1. Percent of activity per alpha-emitting nuclide of plutonium, americium and curium with relative standard deviation.

Sample	Kr-83/Kr-86	Kr-84/Kr-86	Xe-131/Xe-134	Xe-132/Xe-134	Xe-136/Xe-134
A-15	0.2533	0.5709	0.3413	0.6466	1.4671
A-16	0.2511	0.5834	0.3308	0.6818	1.4853
A-17	0.2545	0.5724	0.3397	0.6497	1.4529
A-18	0.2537	0.5812	0.3306	0.6757	1.4686
C-16	0.2567	0.5776	0.3327	0.6749	1.4352
C-17	0.2723	0.5568	0.3603	0.6091	1.2612
C-18	0.2629	0.5639	0.3466	0.6341	1.3890

Table IV.2. Isotopic atom ratios of krypton and xenon.

Sample	U-235/U-238	U-236/U-238	Pu-240/Pu-239	Pu-241/Pu-239	Pu-242/Pu-239	Nd-150/Nd-148
A-15	0.01223 \pm 1.11	0.00247 \pm 3.02	0.3114 \pm 0.56	0.1125 \pm 0.73	0.02388 \pm 1.50	0.4422 \pm 1.69
A-16	0.01102 \pm 3.10	0.00273 \pm 4.85	0.3319 \pm 0.47	0.1485 \pm 0.72	0.03713 \pm 1.05	0.4502 \pm 1.95
A-17	0.01237 \pm 2.32	0.00246 \pm 5.78	0.2997 \pm 0.34	0.1146 \pm 0.63	0.02349 \pm 1.14	0.4449 \pm 0.67
A-18	0.01169 \pm 1.81	0.002535 \pm 3.86	0.3127 \pm 1.0	0.1503 \pm 0.53	0.03229 \pm 2.23	0.4540 \pm 0.67
C-16	0.01281 \pm 1.22	0.00263 \pm 2.34	0.2778 \pm 0.55	0.1218 \pm 0.95	0.02405 \pm 0.77	0.4588 \pm 0.22
C-17	0.01895 \pm 0.99	0.00144 \pm 2.63	0.1562 \pm 1.10	0.0404 \pm 1.78	0.00343 \pm 5.18	0.4325 \pm 0.32
C-18	0.01505 \pm 1.90	0.001946 \pm 4.25	0.2318 \pm 0.30	0.08328 \pm 0.94	0.01172 \pm 1.94	0.4370 \pm 1.03

Table IV.3. Isotopic atom ratio of uranium, plutonium and neodymium with relative standard deviation (1σ).

and Cm-242 are corrected for decay at the reactor shut-down date;

- the amounts of U, Pu, and Nd in atoms and weight;
- the atom percent burnup of fuel samples by the Nd-148 determination.

The relations used for the data reduction have been extensively discussed in (4).

The results of burnup determination and of the contents of uranium and plutonium isotopes are summarized in Table IV.4.

c) Gamma spectrometry

The gamma spectra were evaluated by the code GASPAN which computes the peak surface (counts per second) by fitting a Gaussian function, superposed to a linear one, to the experimental data.

The counting rates given by GASPAN (Table IV.5) were successively treated with the BURNUP program in order to get

- the concentration (atoms per gram of fuel) of Cs-137, Cs-134, Ru-106, Ce-144, Zr-95 and Eu-154;
- the atom percent burnup by Cs-137.

The following relation has been used to evaluate the atoms per gram of fuel of each radionuclide:

$$N = \frac{\bar{c} \cdot F}{E \cdot D \cdot W \cdot P \cdot \lambda \cdot A} \quad (6)$$

where:

- N : number of atoms per gram of fuel corrected for in-pile and out-of-pile decay (g^{-1}).
- \bar{c} : average count rate of the sample (s^{-1}).
- E : efficiency of the detector system.
- D : branching ratio of the gamma transition.
- W : weight of sample (g).

Sample	$F_T(a/o)$	Uranium			Plutonium				
		$235/\Sigma N_i^o$ $\times 10^{-2}$	$236/\Sigma N_i^o$ $\times 10^{-3}$	$238/\Sigma N_i^o$ $\times 10^{-1}$	$238/\Sigma N_i^o$ $\times 10^{-5}$	$239/\Sigma N_i^o$ $\times 10^{-3}$	$240/\Sigma N_i^o$ $\times 10^{-3}$	$241/\Sigma N_i^o$ $\times 10^{-4}$	$242/\Sigma N_i^o$ $\times 10^{-5}$
A-15	1.543	1.185	2.385	9.654	1.708	3.385	1.054	4.087	8.083
A-16	1.862	1.060	2.625	9.620	3.599	3.982	1.322	6.351	14.79
A-17	1.546	1.194	2.371	9.653	1.965	3.417	1.024	4.205	8.027
A-18	1.820	1.124	2.437	9.614	3.633	4.457	1.394	6.699	-
C-16	1.684	1.232	2.534	9.618	3.387	4.482	1.245	5.862	10.78
C-17	0.724	1.838	1.394	9.700	0.4479	2.498	0.3901	1.084	0.8568
C-18	1.220	1.455	1.882	9.668	1.330	3.431	0.7955	3.069	4.022

Table IV.4. Results of F_T determination and of the content of uranium and plutonium isotopes.

Sample	Count rate (c.p.s.)					
	Cs-137	Cs-134	Ru-106	Ce-144	Zr-95	Eu-155
A-15	103.696±0.21	37.722±0.84	20.942±0.19	7.13 ±1.4	5.87 ±1.36	0.751± 1.89
A-16	116.607±0.12	53.581±0.66	24.121±1.56	6.268±1.71	3.437±3.85	0.681± 2.63
A-17	99.891±0.35	35.035±0.64	18.01 ±1.07	5.293±1.98	2.784±0.89	0.889± 4.16
A-18	125.044±0.29	58.409±0.1	24.905±0.43	6.38 ±2.91	3.545±1.31	0.927± 7.68
C-16	108.298±0.45	49.636±0.74	23.804±0.13	6.526±1.37	5.366±2.12	0.962±12.7
C-17	49.199±0.42	8.763±0.47	7.872±2.59	3.471±1.63	2.721±1.10	0.282±16.6
C-18	80.172±0.32	24.813±0.63	16.279±2.21	5.537±0.67	4.659±2.15	0.563± 5.73

Table IV.5. Results of the peak integrals with relative standard deviation (1 σ).

- P : fuel concentration in the solution (g/g).
λ : decay constant (s⁻¹).
F : correction factor, for in-pile and out-of-pile decay (see eq.2).
A : self-absorption factor.

The constants used in calculations relevant to the selected gamma-line are given in Table IV.6.

The number of atoms per gram of fuel are presented in Table IV.7.

The atom percent burnup by Cs-137, $F_T(C_S)$ expressed as heavy atoms burnt per initial heavy atom, was calculated from the relation:

$$F_T(C_S) \text{ (a/o)} = \frac{N}{\bar{y} \cdot \Sigma H_O} \cdot 100 \quad (7)$$

where:

- N : number of atoms of Cs-137 as derived by the precedent relation.
 \bar{y} : effective fission yield of Cs-137.
 ΣH_O : initial heavy atom amount per gram of fuel.

The effective fission yield was calculated taking into account the following yields (7):

- 6.0699 % for U-235
6.6123 % for Pu-239

and assuming that about 70% of the fissions derive from U-235 and 30% from Pu-239.

The results of the burnup determination are also included in Table IV.7.

Nuclide	Energy (keV)	Branching ratio _g	Half-life (days)	Detector effic. %	Self-absorption
Cs-137	662	84.6	10990	0.035	1.056
Cs-134	796; 801	94.13	749	0.026	1.051
Ru/Rh-106	622	10.57	368.3	0.039	1.057
Ce/Pr-144	696	1.51	284.5	0.032	1.054
Zr/Nb-95	725	44.2	65.5	0.030	1.053
Eu-154	1275	34.7	5845	0.012	1.048

Table IV.6. Constants used in the calculation of the fission product concentrations.

Sample	Atoms per gram of fuel						$F_T(C_S)$ (a/o)
	Cs-137 $\times 10^{18}$	Cs-134 $\times 10^{17}$	Ru-106 $\times 10^{17}$	Ce-144 $\times 10^{18}$	Zr-95 $\times 10^{18}$	Eu-154 $\times 10^{17}$	
A-15	2.167	1.409	5.123	1.782	2.315	0.961	1.559
A-16	2.548	2.224	7.009	1.933	2.962	0.914	1.833
A-17	2.133	1.419	5.104	1.591	2.320	1.166	1.534
A-18	2.731	2.429	7.272	1.981	3.152	1.244	1.965
C-16	2.224	1.829	5.765	1.617	2.171	1.209	1.600
C-17	1.049	0.335	1.976	0.891	1.131	0.367	0.754
C-18	1.708	0.948	4.083	1.420	1.935	0.734	1.229

Table IV.7. Results of $F_T(C_S)$ and fission products concentration.

V. DISCUSSION

The main aim of the task was to compare the validity of results obtained by nondestructive γ -assay and by destructive analysis, which comprised radiometric and mass spectrometric techniques. In order to check the consistency of data resulting from destructive analyses an isotopic correlation technique was used successfully in the past (4, 7, 8). By this way a higher confidence can be established for data referring to the destructive methods. On this basis the results obtained from the nondestructive measurements can be evaluated.

V.1. Consistency check of results of mass spectrometric destructive analysis

From our past experience it proved that isotope correlations between the isotope ratios Kr-84/83 or Xe-132/131 and the burnup (F_T) or the heavy isotope content are best suited to check the results. These correlations showed to be independent of the individual reactor, therefore data earlier obtained from post-irradiation analysis of fuel pellets of BWR GARIGLIANO and BWR LINGEN were used to establish the correlations, in order to check consistency of the DODEWAARD data. The correlations applied are listed in Table V.1. together with the linear least square fit equations as obtained from the GARIGLIANO and LINGEN data.

F_T	= 3.181	Xe-132/131	- 4.837
Pu-240/239	= 0.624	Kr-84/83	- 1.096
Pu-240	= 0.00297	Xe-132/131	- 0.00462
Pu-241	= 0.00138	Xe-132/131	- 0.00221
Pu-242	= 0.000555	Xe-132/131	- 0.000976

Table V.1. Linear least square fit equations for GARIGLIANO and LINGEN data used for consistency check of DODEWAARD results.

The deviations of the DODEWAARD results are not significant and according to our experience mainly caused by the usual change of reactor operation parameters (Fig. 10), except for the correlation F_T versus Xe-132/131. Here all DODEWAARD values of F_T are systematically higher (Fig. 11).

Fig. 12. shows the ratio of Pu-240/239 versus the ratio of U-235/238. From this figure it can be seen that some correlations of heavy isotopes are influenced by the neutron spectrum, i.e. the influence of the control rod. The correlation does not seem to be dependent on the void fraction because the positions of the samples cover a large range of average void fractions from a few percent up to 65%.

V.2. Validity of destructive gamma spectrometric measurements

The possibility to use radioactive fission products for the determination of burnup or for establishing isotope correlations as observed, e.g. for the fission gases, was investigated before (9) and indicated a dependence on fuel irradiation history.

Cs-137 distinguishes itself from other radioactive fission products due to its long halflife (30.08 y.) and its high abundance which makes the precise measurements of the characteristic γ -line of 662 keV possible without interference of other fission products. However, at high fuel temperatures cesium tends to migrate along temperature gradients in the fuel matrix and is deposited at colder positions. Hiller suggested a fuel temperature threshold of the migration effect of Cs of approximately 2000°C (10).

The burnup results obtained by the destructive analysis of Cs-137 are in rather good agreement with those obtained by the Nd-148 method except value A-18 (Fig. 13). As this burnup value resulting from the Nd-148 method agrees with the correlations mentioned above, it is assumed that the different value derived from the Cs-137 measurement may be caused by a cesium migration.

Correlations of reactor parameters and the Cs-134/137 are of special interest because they can be obtained non-destructively. However, this type of correlations varies slightly with

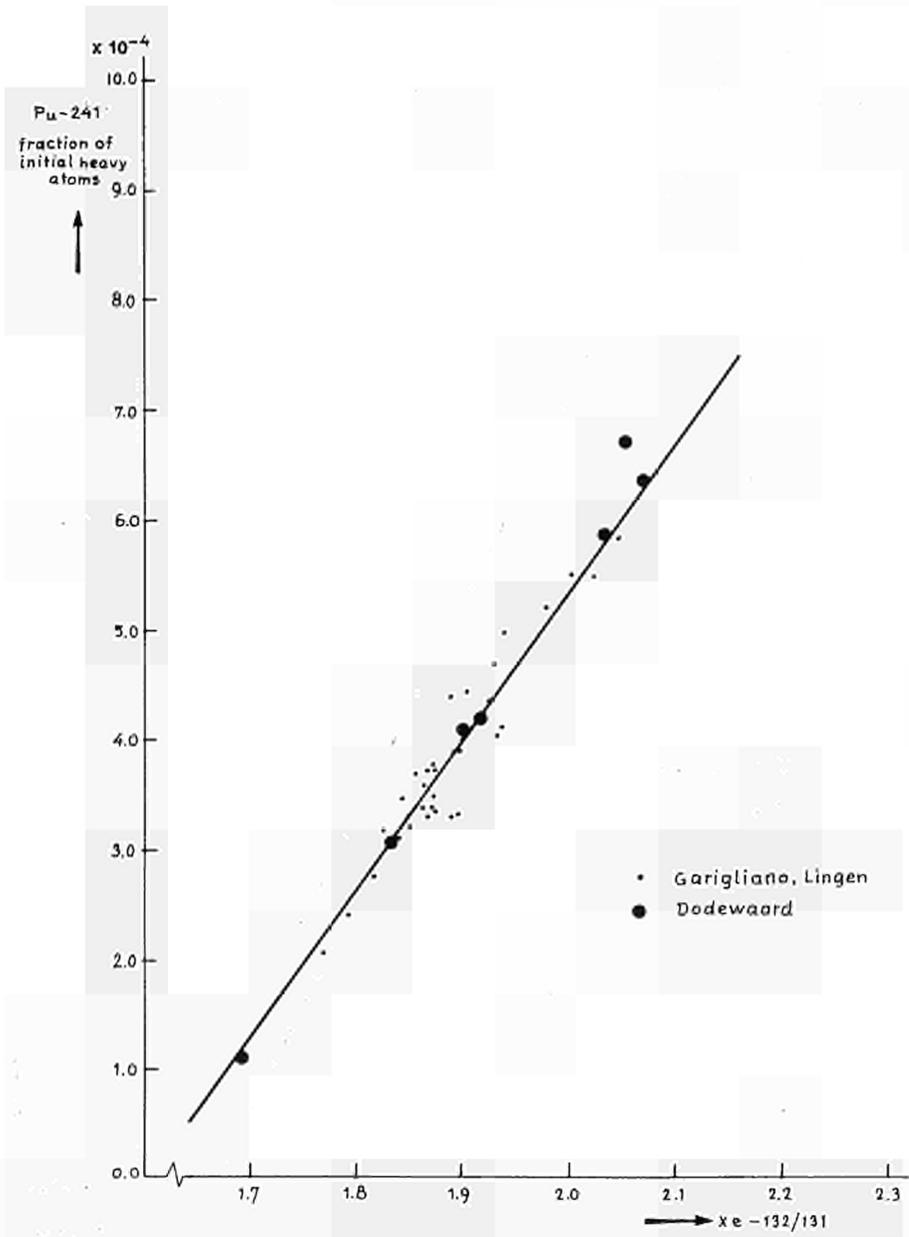


Fig.10 Correlation of Pu-241 vs. Xe-132/131

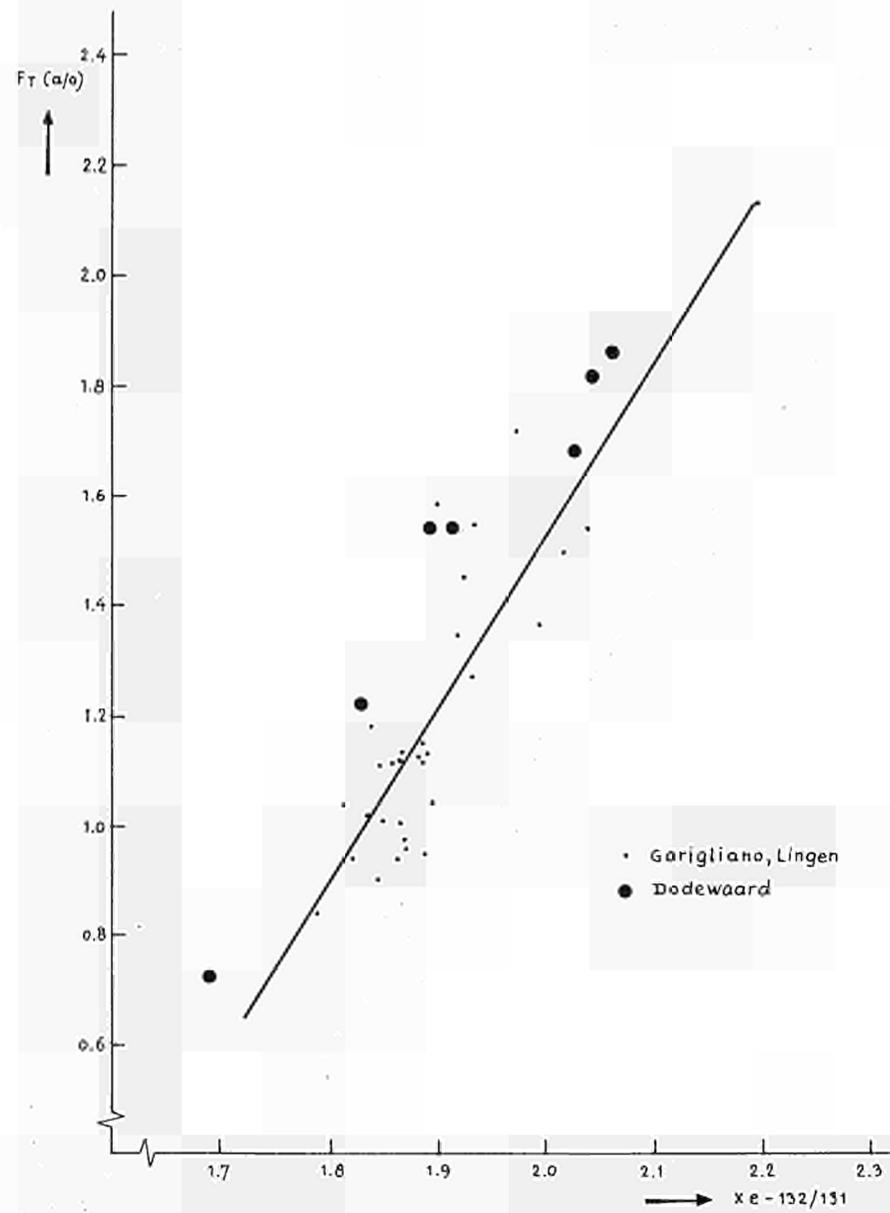


Fig.11 Correlation of F_T vs. Xe-132/131

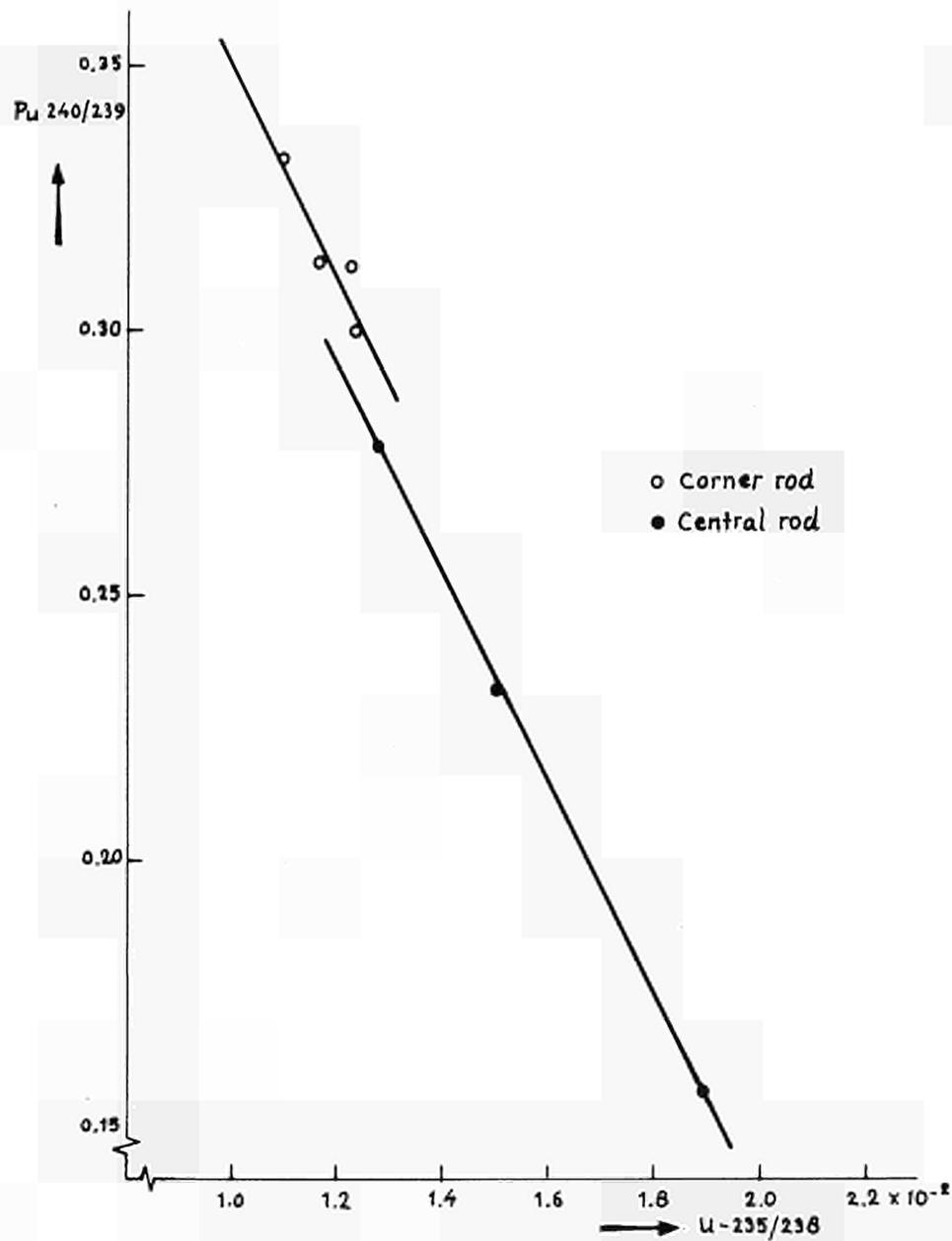


Fig. 12 Correlation of $Pu-240/239$ vs. $U-235/238$

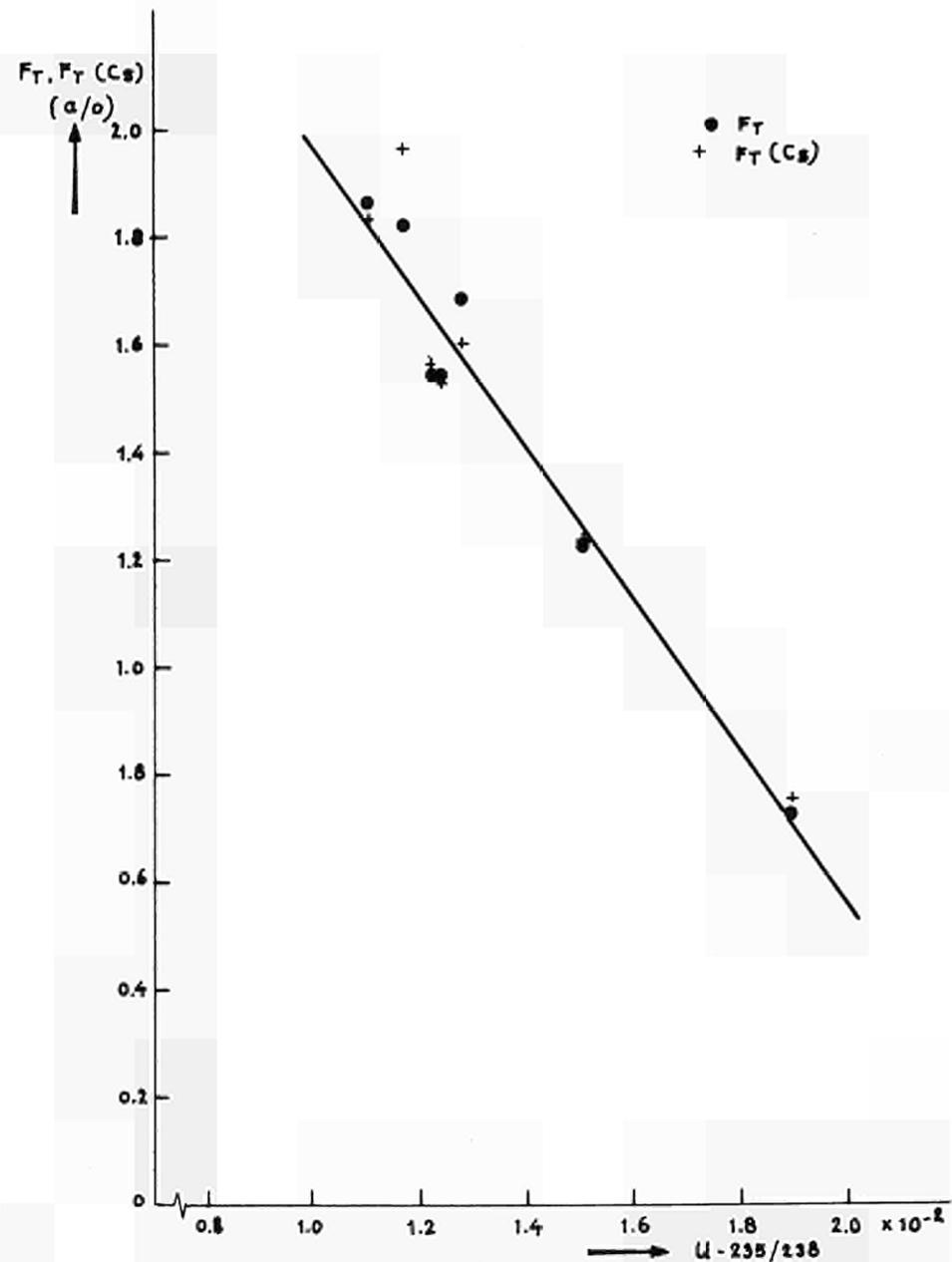


Fig. 13 Comparison of F_T and $F_T(C_9)$ vs. $U-235/238$

the individual reactor. The corresponding linear least square fit equations are given in Table V.2. for each of the same three BWR's and for all together.

F_T	=19.2	(<u>+1.71</u>)	Cs-134/137 + 0.176	(<u>+0.121</u>)	DOD
F_T	=21.8	(<u>+2.56</u>)	Cs-134/137 - 0.364	(<u>+0.181</u>)	GAR
F_T	=30.0	(<u>+3.62</u>)	Cs-134/137 - 0.487	(<u>+0.199</u>)	LIN
F_T	=17.3	(<u>+2.30</u>)	Cs-134/137 + 0.13	(<u>+0.15</u>)	ALL
Pu-240=	0.017	(<u>+0.00085</u>)	Cs-134/137 - 0.000123	(<u>+0.000060</u>)	DOD
Pu-240=	0.0272	(<u>+0.0016</u>)	Cs-134/137 - 0.000892	(<u>+0.000114</u>)	GAR
Pu-240=	0.0188	(<u>+0.0016</u>)	Cs-134/137 - 0.000859	(<u>+0.000086</u>)	LIN
Pu-240=	0.015	(<u>+0.0013</u>)	Cs-134/137 + 0.000041	(<u>+0.000085</u>)	ALL
Pu-241=	0.00983	(<u>+0.00029</u>)	Cs-134/137 - 0.000222	(<u>+0.000021</u>)	DOD
Pu-241=	0.0101	(<u>+0.00042</u>)	Cs-134/137 - 0.000298	(<u>+0.000030</u>)	GAR
Pu-241=	0.0102	(<u>+0.00034</u>)	Cs-134/137 - 0.000195	(<u>+0.000019</u>)	LIN
Pu-241=	0.00804	(<u>+0.00052</u>)	Cs-134/137 - 0.000110	(<u>+0.000034</u>)	ALL
Pu-242=	0.00239	(<u>+0.00030</u>)	Cs-134/137 - 0.000077	(<u>+0.000020</u>)	DOD
Pu-242=	0.00473	(<u>+0.00017</u>)	Cs-134/137 - 0.00033	(<u>+0.000012</u>)	GAR
Pu-242=	0.00303	(<u>+0.00034</u>)	Cs-134/137 - 0.00010	(<u>+0.000019</u>)	LIN
Pu-242=	0.00253	(<u>+0.00029</u>)	Cs-134/137 - 0.000086	(<u>+0.000018</u>)	ALL

Table V.2. Linear least square fit equations for correlations of F_T and plutonium isotope content versus Cs-134/137 ratio for DODEWAARD, GARIGLIANO, LINGEN data and for all together.

The differences are caused by the parameters governing the Cs-134 build-up, which are related to the reactor design and to the power history.

The consequences of the power history on the Cs-134 activity can be evaluated by adding a factor to equation (2) which will correct for the time- and flux dependent build-up of Cs-133. The effect of the reactor design on the build-up of Cs-134 will be discussed below.

V.3. Comparison of results of non-destructive and destructive analysis

From the nuclides measured by non-destructive analyses the ratio of Cs-134/137 is most suited to be correlated with reactor parameters. Compared with the values of Cs-134/137 as measured in the dissolved fuel samples, a deviation on the average of about 7% can be observed (Table V.3.).

Sample	Deviations		
	Cs-134/137	Ru-106/Cs-137	Ce-144/Cs-137
A-15	3.90	2.42	- 3.22
A-16	11.39	20.17	- 7.04
A-17	4.16	13.43	- 6.80
A-18	3.22	17.16	- 0.41
C-16	5.96	22.34	- 5.39
C-17	13.41	13.16	- 11.95
C-18	8.97	15.55	- 3.22

Table V.3. Deviations, (Non Destr. - Destr.).100/Non Destr., between non destructive and destructive gamma spectrometric measurements.

Fig. 14, 15, 16 and 17 illustrate the correlations of F_t and plutonium isotope content versus the Cs-134/137 ratio, measured non-destructively.

It can be seen that the correlations cover a large range of burnup and plutonium isotope content values.

The discrepancy between Cs-134/137 ratio of the two methods may be caused by two reasons:

- The SANA program does not completely exclude the influence of the 802.1 keV line on the 795.8 keV line of Cs-134.

Consequently the results of the Cs-134 activity, measured

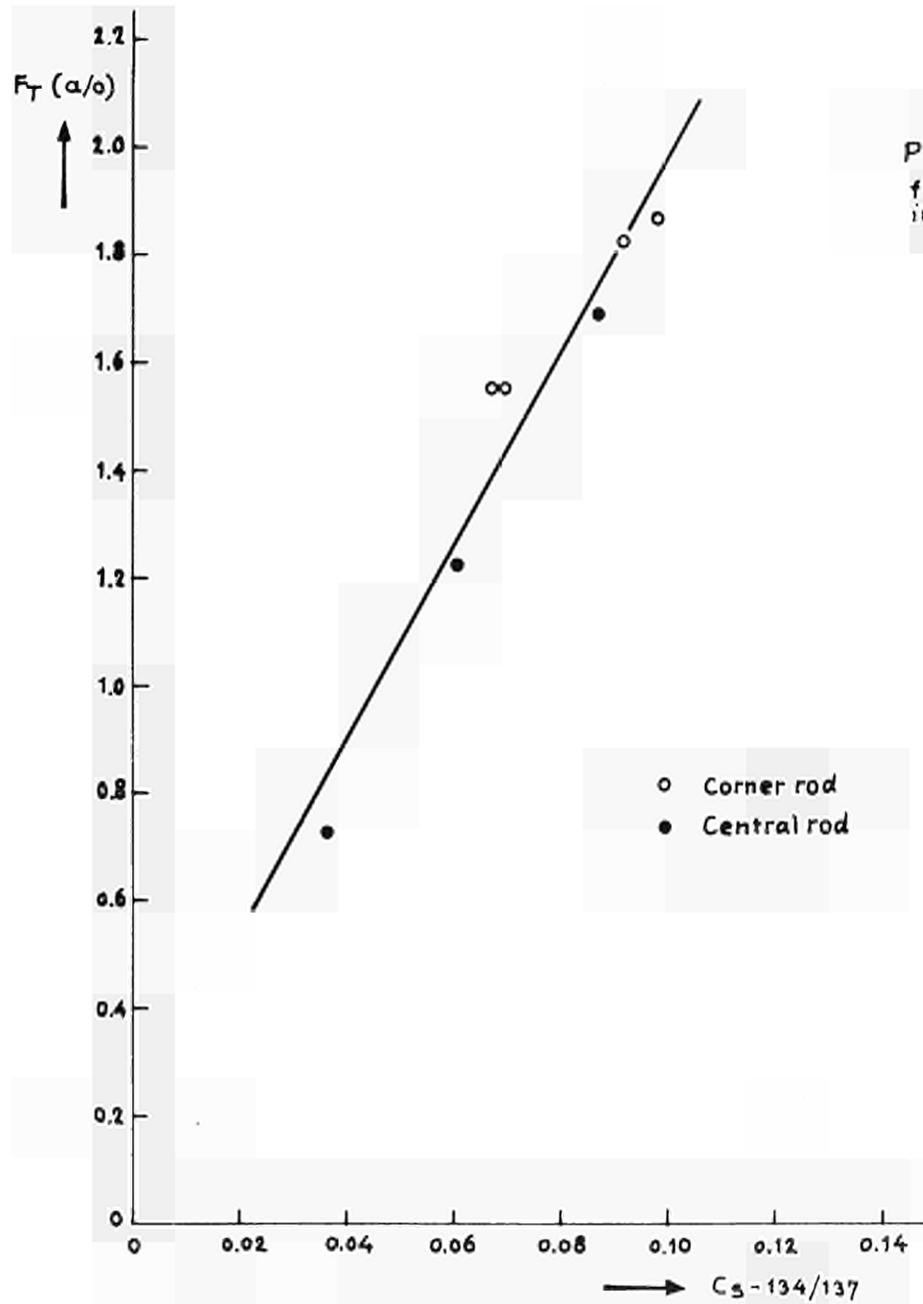


Fig.14 Correlation of F_T vs. $Cs-134/137$

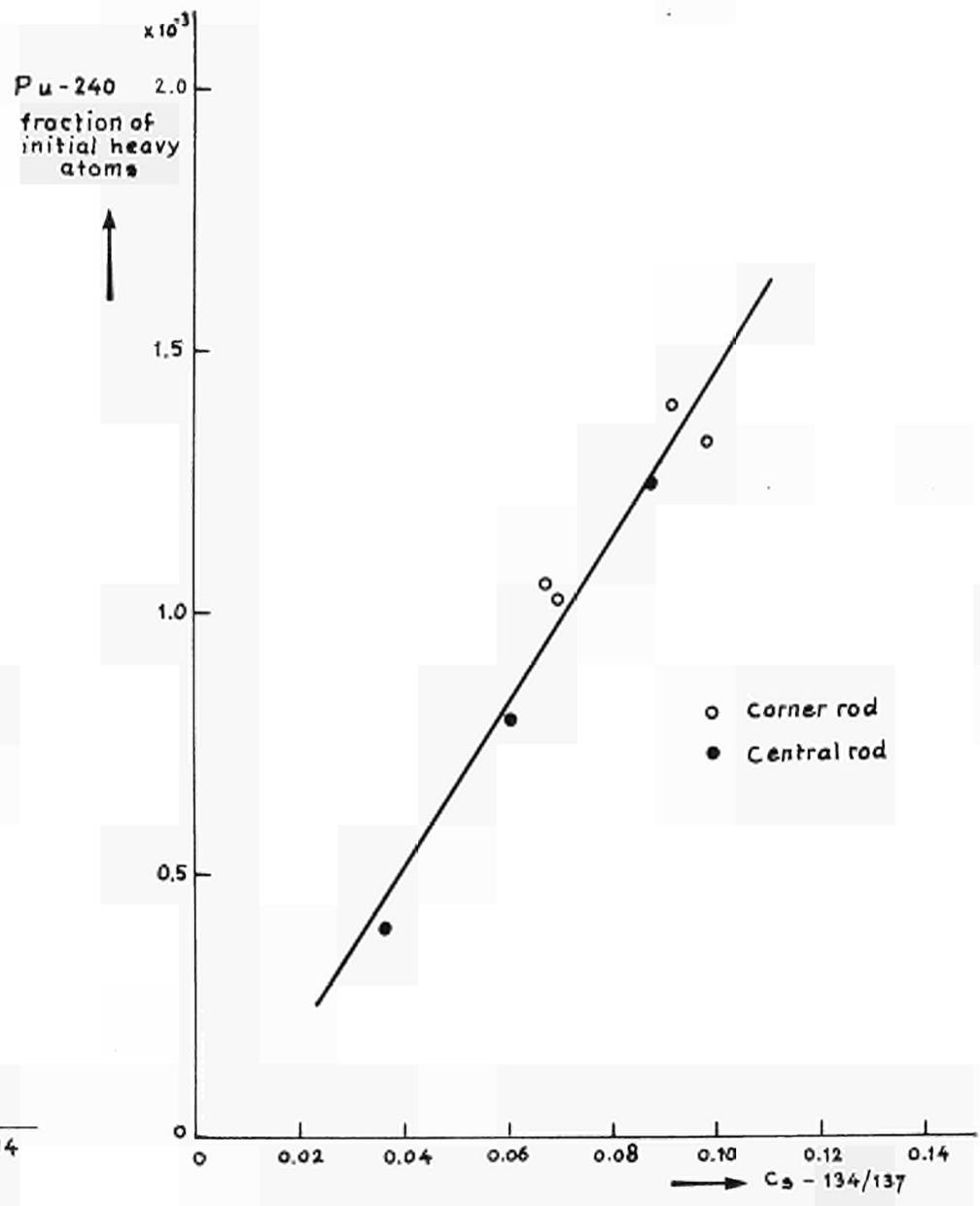


Fig.15 Correlation of $Pu-240$ vs. $Cs-134/137$

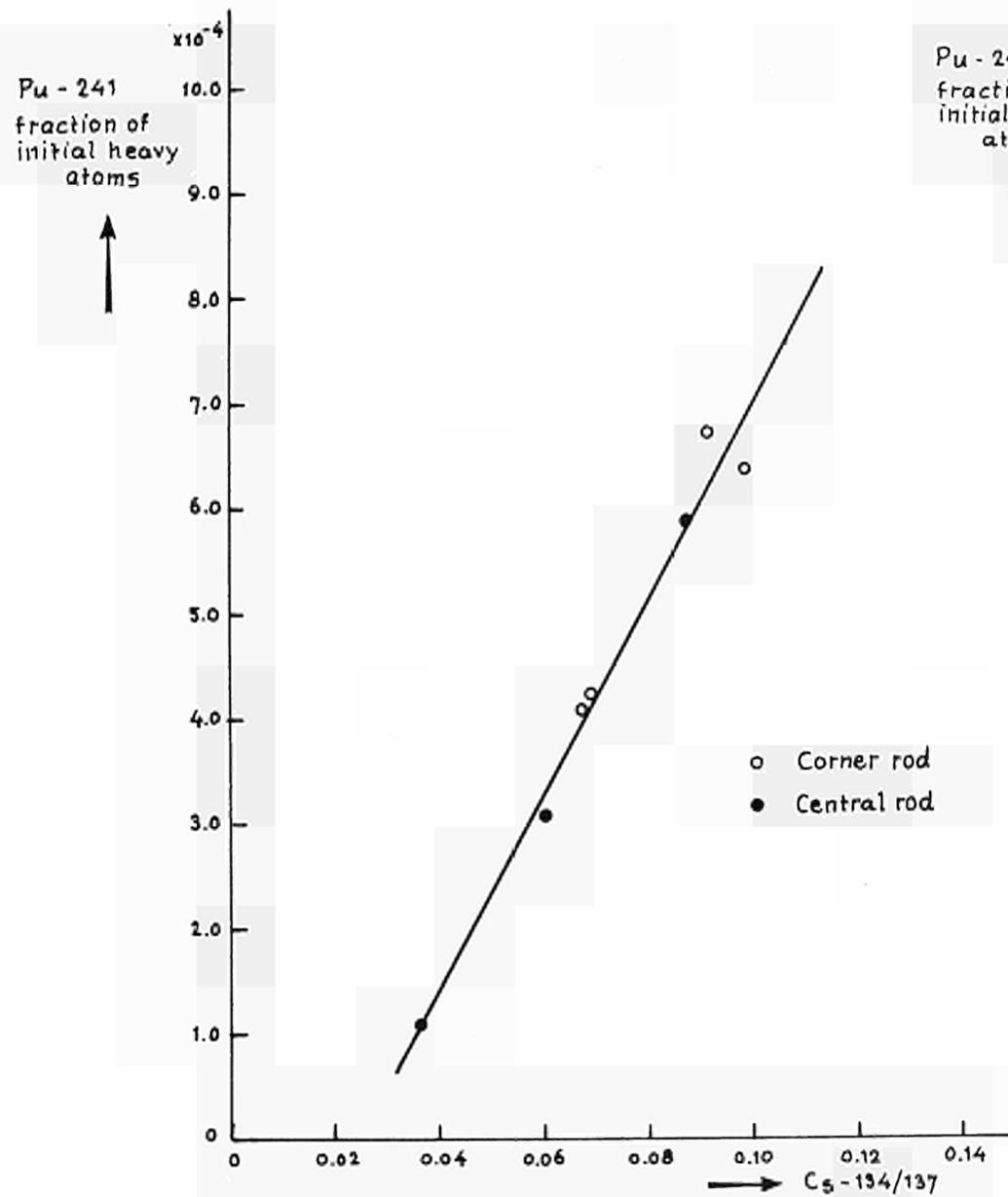


Fig.16 Correlation of Pu-241 vs.Cs-134/137

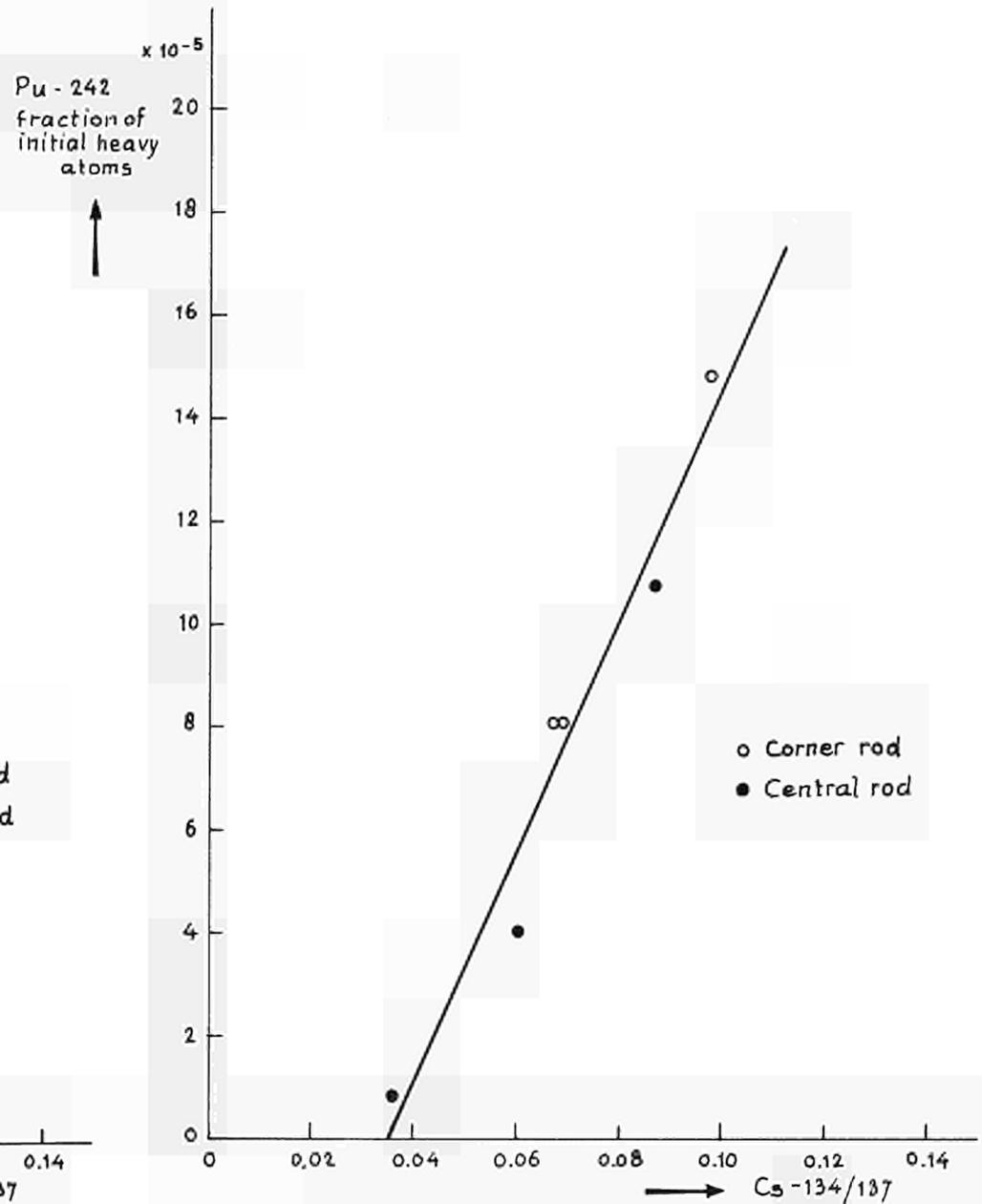


Fig.17 Correlation of Pu-242 vs.Cs-134/137

non-destructively, will be a few percent too high. The Cs-134 activity of the dissolved fuel has been determined on the basis of the 802.1 + 795.8 keV lines because the resolution of the detector did not allow a separation of two lines.

- The radial Cs-134 distribution in the fuel may be different from that of the Cs-137, because Cs-134 is produced increasingly with advancing burnup. The only way of production of Cs-134 is by neutron capture of Cs-133.

With increasing burnup the Cs-137 + Cs-133 production moves to the center of the fuel rod, because the fissile material in its outer part becomes depleted.

The Cs-134 production, however, continues in this region because formerly produced Cs-133 is still available. The difference in absorption of the characteristic gamma lines of 662 keV (Cs-137) and 796 keV (Cs-134) is about of the same magnitude as observed experimentally (1,2 and 0.95 cm resp.). This hypothesis is confirmed by the results obtained for the ratio Ru-106/Cs-137 (Table V.3.).

The more pronounced deviations of the Ru-106/Cs-137 ratios may be caused by the different position within the fuel pellet of deposition of Ru-106 and of Cs-137, Ru-106 mainly stemming from plutonium fissions.

For the deviations of the Ce-144/Cs-137 ratios a similar explanation can be given, because the Ce-144 stems to a larger extent from uranium fissions. Moreover the 696.5 keV line of Ce-144 may be influenced by Eu-154.

A quantitative answer to the phenomena described above, can be given only by an experimental investigation, where micro-samples taken from different radial positions are analysed.

VI. CONCLUSIONS

From the experiments described and from the comparisons, some conclusions and recommendations can be given:

- Once the correlations Cs-134/137 (non-destructive analysis) versus burnup and heavy isotopes (destructive analysis) have been established, the reactor operator will have a useful method for the analysis of spent fuel, provided the correlations cover the range of different enrichments, burnup and power ratings. The correction for the influence of the power history on the Cs-134 build-up has to be investigated and applied.
Present changes in BWR and PWR design, i.e. a smaller fuel pin diameter and lower linear power ratings, could enhance the usefulness of the method because the influence of the different radial distribution of Cs-134 and Cs-137 will be decreased and the probability of cesium migration will be very low.
- The accuracy of the non-destructive gamma analysis can be improved by a more rigid construction of the scanning equipment and by a decrease of the absorbing material between the fuel sample and the detector. The count rate can be optimized by an increased distance sample-detector.
- In order to classify and correct for the deviations of the Cs-134/137 ratios, measured non-destructively and destructively, a further analysis of microsamples at different radial positions in the fuel pellet is recommended.

ACKNOWLEDGEMENTS

The authors wish to acknowledge the valuable help of Mr. Hoekstra and Mr. Van Bernebeek for their assistance in the non-destructive measurements, Mr. De Meester and Mr. Heitz for their work in the hot chemical laboratories and Mr. Molinet for his work in the mass spectrometric laboratories.

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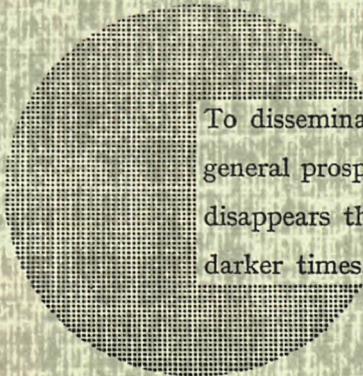
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