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INVESTIGATIONS ON RADIOACTIVE FISSION PRODUCT CORRELATIONS: GAMMA SPECTROMETRY MEASUREMENTS ON SPENT FUEL ASSEMBLIES OF THE GARIGLIANO REACTOR

by

G. ADILETTA, N. BENATTI, R. GUIDOTTI, M. PAOLETTI GUALANDI, P. PERONI (ENEL-Italy)
A.M. BRESESTI, M. BRESESTI, D. D'ADAMO (Euratom)

1974

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The gamma ray activities of the fission products were correlated with values of burnup, plutonium content and power calculated by means of nuclear codes at the end of the cycle.

A proportionality was observed between Cs 137 activity and burnup : this result indicates the feasibility of relative measurements of burnup in fuel assemblies by means of non-destructive techniques.

On the other hand no correlation was found between the Cs 134/Cs 137 activity ratio and Pu/U mass ratio due to the important differences existing in the power history of the fuel assemblies.

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ABSTRACT

In the framework of a collaboration between ENEL and EURATOM (JRC Ispra), gamma spectrometry measurements with a Ge(Li) detector were performed on 11 fuel assemblies of the Garigliano BWR, to study the correlations existing between radioactive fission products and burnup, plutonium content and power at the end of cycle.

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1. INTRODUCTION

The work presented in this report was carried out in the framework of a collaboration agreement between ENEL and EURATOM Joint Research Center (Nr. 055-71-PIPGI), established for the development of techniques for the fissile material control.
The main point of the collaboration agreement concerns the study of the isotope correlations in irradiated fuels. ENEL is interested in this kind of study since these correlations can improve the consistency and accuracy of fissile material balances. The EURATOM Joint Research Center is interested in the same studies for reasons connected with the development of safeguards techniques.

The work performed on the spent fuel assemblies of the Garigliano reactor is directed to investigating the possibility of correlating data gathered through gamma spectrometry measurements of fuel assemblies with the data of burnup, plutonium content and power at the end of cycle.

The first information on these correlations was obtained during post-irradiation analyses, performed at the Joint Research Center, Ispra, on small samples of the fuel discharged from the Trino Vercellese PWR reactor at the end of the first irradiation cycle. In fact, during these analyses, linear correlations of radioactive fission products with burnup and plutonium content were observed[1]. On the basis of these indications a first gamma scanning experiment was carried out during 1972, under the EURATOM-ENEL Research Contract Nr. 071-66-6 TEEI, in the spent fuel pond of the Trino Vercellese PWR reactor on 12 fuel assemblies discharged after two irradiation cycles.

This experiment showed a proportionality between the measured Cs 137 activity and the burnup and between the measured Cs 134/Cs 137 activity ratio and the plutonium content in the fuel assemblies[2,3).
A correlation between Cs 134/Cs 137 activity ratio and burnup was also observed in SENA fuel assemblies[4)].
These results seemed to indicate the possibility of carrying out relative measurements of burnup and plutonium content in fuel assemblies.

In order to collect further information in this field ENEL and EURATOM Joint Research Center performed a new gamma scanning experiment in January 1973 in the spent fuel pond of the Garigliano reactor on 11 fuel assemblies discharged in April 1972.
The fission product activities determined by means of gamma spectrometry measurements are related to the values of burnup and of power at the end of cycle obtained from follow up calculations carried out by the FLARE code[5)] and to values of plutonium content generated by the BURSQUID code[6)].

The Garigliano nuclear power station is equipped with a 506 MWth (160 MWe) boiling water reactor. The reactor core consists of 208 fuel assemblies with a total UO_2 content of about 50 tons. The fuel assemblies of the first core consist of a 81-rod bundle in a 9 x 9 array (fuel diameter 11.9 mm, average enrichment 2.02 %) ; the reload fuel assemblies consist of a 64-rod bundle in a 8 x 8 array (fuel diameter 12.9 mm, average enrichment 2.30 %).
The active length of the fuel rods is about 270 cm.
The fuel cladding is zircaloy.

In total 11 fuel assemblies have been measured.
Nine fuel assemblies consisted of a 81-rod bundle ; 12 rods had an initial enrichment of 1.6 % and 69 rods an initial enrichment of 2.1 %, the average enrichment being 2.02 %.
The total UO_2 weight in a fuel assembly was 250 Kg.
The range of burnup determined by means of computer code calculations was between 15,500 and 18,500 MWD/MTU. The other two fuel assemblies consisted of a 64-rod bundle ; 12 rods had an initial enrichment of 1.83 % and 52 rods an initial enrichment of 2.42 %, the average enrichment being 2.30 %.
The total UO_2 weight in a fuel assembly was 226 Kg.
The burnup values of the two assemblies were about 7,000 and 14,000 MWD/MTU respectively.
The irradiation history and the locations of the fuel assemblies in the core are reported in Table 1.
Fig. 1 shows how it is possible to determine the actual locations of the fuel assemblies in the core during the different irradiation cycles.

TABLE 1

IRRADIATION HISTORY AND LOCATIONS OF THE GARIGLIANO FUEL ASSEMBLIES

Fuel Assembly	Rod Array	Locations of the Fuel Assemblies in the Core		
		1st Cycle 23-11-63/30-7-68	2nd Cycle 22-10-68/13-6-70	3rd Cycle 30-9-70/22-4-72
A 001	9 x 9	5621	5621	6609
A 083	"	6601	6601	6613
A 053	"	5922	5922	6607
A 219	"	6322	6322	5615
A 160	"	6607	Cooling Pond	6120
A 195	"	6819	6819	5702
A 217	"	5403	5403	5720
A 147	"	6520	6619	5205
A 154	"	6502	6603	5900
SB 51	8 x 8			5817
SA 16	"		6508	5815

2. COLLIMATION AND FUEL POSITIONING SYSTEM

The facilities for gamma scanning consist of a collimator system, a transfer system to move the assembly or the rods in front of the collimator, and a structure outside the spent fuel pool, where the measurement equipment is located (Fig. 2).

The collimation system is composed of a fixed lead collimator, with an opening of pre-established dimensions, located in a hole purposely bored through the spent fuel pool wall, and a movable collimator, with a variable opening, located on the structure outside the pool. The fixed collimator is located in a stainless steel outer tube which provides an exact fit with the pool wall and acts as a guide tube. A rectangular box is located inside the guide tube in horizontal position. At both ends of this box, the gap between the guide tube and the box itself is plugged with lead.
Inside the rectangular box the collimator is placed, the section of which decreases stepwise to minimize radiation streaming. The collimator slit is 20 mm high and 160 mm wide.
The movable collimator has an opening, the height of which can be varied. It consists of two steel boxes filled with lead, which are mounted one over the other upon a frame and can be moved away from one another by means of a fine-regulation screw, while maintaining the two faces perfectly parallel. The height of the opening during the measurements was about 1 mm.

The fuel assembly positioning system is located in the spent fuel pool in front of the collimator.

It consists of a guide sliding on rails in vertical direction and of a motor-operated chain drive. The guide can accommodate the assembly to be measured and can rotate around its main axis and move sideways. Rotation can be used to scan the assembly along 360 degrees ; the sideways movement permits the fuel to be positioned perfectly on the axis of the collimation system.

Corner measurements on the fuel assemblies were preferred to side measurements. In this way the effect on the detected activity of an imperfect positioning of the fuel assembly in front of the collimator, is much lower. In fact, previous experiments in the Garigliano reactor had shown that in the case of side measurements on fuel assemblies variation of about 30 % in the 1.60 MeV La 140 gamma activity was produced by an angular variation of 10^{o} in respect of the collimator-detector axis, while the same angular deviation caused a variation of about 1.5 % in the case of corner measurements.

3. GAMMA SPECTROMETRY MEASUREMENTS

The detection system consisted of a Ge(Li) detector connected to a 400 channel analyzer. The coaxial-type Ge(Li) detector had a volume of 30 cm^3 and a FWHM of about 3.5 KeV at 1.33 MeV. For the determination of the gamma photopeak areas, the sum of the channel contents between two selected limits were performed by the analyzer itself. The background at these limits was assumed as the average of the countings in the selected channel and in the two neighbouring channels, being the weight for the neighbouring channels taken as 0.5.

The variation of the background between the limits was assumed to be linear.
The total counting rate was kept below 10^4 counts/sec. The losses for pile-up effects in the amplifier were negligible and the dead-time losses in the converter were automatically corrected.
This was checked by means of the "two sources" technique.

A typical gamma spectrum, measured after a 9 months cooling time, is shown in Fig. 3.
The gamma rays considered for the aims of this experiment are listed in Table 2.

TABLE 2

GAMMA RAYS MEASURED IN THE FUEL ASSEMBLIES

Nuclide	Gamma Photopeak Energy (KeV)	Half-Life
Cs 137	662	30.1 y
Cs 134	605	2.05 y
	796 + 802	
Zr-Nb 95	757 + 766	65 d - 35 d
Zr 95	724	65 d

The gamma spectrum shows also several gamma photopeaks of the following nuclides :

Ru-Rh 106
Ce-Pr 144
Eu 154
Ag 110m
Co 60

The first two nuclides are fission products, the other three nuclides are formed by neutron capture respectively on the fission products Eu 153 and Ag 109 and Co 59 which is one of the impurities of the zircaloy. The 658 KeV gamma ray of Ag 110m can interfere in the determination of 662 KeV photopeak of Cs 137. The effect of the interference was established through the measurement of the 1384 KeV photopeak of Ag 110m. This evaluation was made on the basis of the known ratio between the Ag 110m gamma ray intensities at the two energies and by using a curve of detection efficiency as a function of the energy derived from the determination of the Cs 134 photopeaks of different energies.
Since the contributions are small the values of the 662 KeV photopeak areas were not corrected for the Ag 110m presence.

4. THEORETICAL EVALUATIONS

4.1 Power, Burnup and Plutonium Buildup of the Assemblies

In order to verify the validity of the correlations involving radioactive fission products, the results of the gamma spectrometry measurements have been related with data of power, burnup and plutonium buildup generated by means of computer code calculations. Data on burnup and plutonium buildup, produced during the reprocessing operations, are more accurate than theoretical data but are not available at present.

The data of power and burnup distribution have been calculated, for the fuel assemblies considered in the present experiment, by means of the tridimensional diffusion code FLARE[5], which is normally used by ENEL for the follow-up calculations of the Garigliano reactor core. The power distributions generated by FLARE had been previously compared with those experimentally determined by means of gamma scanning and satisfactory agreements had been observed. The same code, on the basis of the power distribution and of the energy produced in the considered time interval, gives the burnup distribution which is in general quite accurate.

The input data of the FLARE code (K_∞ , M^2, etc.) for each type of fuel assembly have been calculated by means of the BURSQUID code[6] on a two-dimension detailed model of the section of the fuel assembly, considered as isolated. These calculations, carried out for various irradiation levels, give also the plutonium concentrations utilized for the analysis of the correlations considered in this experiment.

4.2 Mutual Rod-Shielding and Decay Corrections

The gamma activity measurements taken on irradiated fuel assemblies for the purpose of measuring the activity of isotopes, can be influenced by mutual rod shielding[7]. This effect is due to the attenuation undergone by the gamma rays when they cross the fuel assembly regions lying on the path towards the detector. Because of the very strong shielding effect of the fuel rods only the peripheral rods contribute significantly to the measurement, so that substantially different activity distributions in the assembly can give rise to different measured values for the same average activity emitted.

In order to assess the magnitude of this effect and to calculate a correction coefficient that allows the measurement to be brought back to the actual value with a fair degree of approximation, the code ATTENUA has been developed which takes into account the geometry of the assembly, the energy of the gamma rays of interest, the activity distribution inside the assembly and the geometry of the measurement system. Since experience acquired has revealed that greater precision can be obtained by placing the assembly corner in front of the detector, the ATTENUA code takes into account only the corner positioning of the assembly. In addition the code theory is based on the assumption that the gamma rays reaching the detector are parallel. This assumption is valid when the distance between detector and assembly is larger than 10 feet as is the case in this experiment. The code divides the fuel assembly into layers parallel to the diagonal lying in the same direction as the detector axis. The thickness of the layers is given as an input, and the attenuations involved are computed analytically for each layer. In this manner, on the basis of a theoretical rod-by-rod activity distribution, the code can simulate the effect in question fairly accurately for each of the four corners of the assembly.

In order to compute a sufficiently accurate rod-by-rod activity distribution the ATTIVA code has been developed which receives in input at the different burnup levels, from the BURSQUID code, for each rod, the absolute values of the neutron flux, the fission cross-sections of fissile isotopes, and the data essential to the calculation of the absorption cross-sections of the various fission products.

From these data and on the basis of the power history of each assembly, the code calculates the rod-by-rod activity distribution for the fission nuclide considered, which will be given as input to the ATTENUA code to complete the analysis.

By means of the chain BURSQUID - ATTIVA - ATTENUA the activities of the different nuclides in a fuel assembly, which is assumed to be irradiated at a constant power, can be calculated. These activities can be compared with similar activities calculated for each fuel assembly considered and correction factors can be derived to take into account the effects of mutual shielding and decay.

4.3 Isotope Correlations

In order to estimate the range of validity of the isotope correlations involving radioactive fission products and to find out which are the parameters influencing these correlations, a series of calculations have been carried out, by means of the ATTIVA code, on a zerodimensional model.
This analysis, performed for the fuel of the Garigliano reactor, has given some indications which can assist in the interpretation of the experimental results.

Fig. 4 shows the correlation between Pu/U mass ratio and Cs 134/Cs 137 activity ratio for different values of voids fractions. The correlation is essentially linear and independent of the voids fractions except for high voids fractions (50 %) and burnup values above 20,000 MWD/MTU.
The calculations have also shown that the fuel enrichment has a slight influence on the behaviour of the correlation.

On the contrary the correlation is strongly influenced by the power level of the irradiation, as shown in Fig. 5 where it can be observed that to the same Cs 134/Cs 137 activity ratio correspond different Pu/U ratios for fuel assemblies which were irradiated at different power levels.

A theoretical analysis has been carried out also for the correlation between Pu/U mass ratio and Eu 154/ Cs 137 activity ratio. This correlation is essentially independent of the power level and the fuel enrichment ; however, this correlation deviates from linearity above 20,000 MWD/MTU in different ways depending on the voids fractions, as shown in Fig. 6. This behaviour can be explained considering the high value of the Eu 154 neutron capture cross section.

5. MEASUREMENT DATA

The procedure utilized for the measurements performed on the fuel assemblies is the following :

- Measurements were performed on each corner of the assemblies at 8 levels which were symmetrical with respect to the mid-point of the active length.
  The 8 levels were spaced at equal intervals of 34.5 cm.
  In the measurements of the 8 x 8 fuel assemblies 2 levels were displaced of 9 cm to avoid interferences with the grids.
  The countings of the 8 positions were summed in one spectrum which was processed to obtain the net areas of the various gamma photopeaks.
- The areas of the gamma-photopeaks for the four corners were summed to give an integral value for the fuel assembly. The integral values for the fuel assembly correspond to a total counting time of 3,200 sec.

One of the 11 fuel assemblies (assembly Nr. A 001) was selected as a reference assembly. In order to detect possible variations in the detection efficiency, this fuel assembly was subjected to measurements at different times during the experiment. No systematic variation of the detection efficiency was observed during the measurements of the 9 x 9 fuel assemblies.

Table 3 gives the results of the measurements on each corner of the reference fuel assembly.
Table 4 shows the reproducibility of the integral measurements on the reference fuel assembly for the gamma activities and for the gamma activity ratios.

It can be observed that the reproducibility is quite satisfactory for the measurements of the Cs 137 activity (standard deviation 1.7 %) and of the Cs 134/ Cs 137 activity ratios (standard deviations of 1.2 % and 0.5 % for the Cs 134 gamma photopeaks of 605 KeV and 796 + 802 KeV respectively). The standard deviations of the activity ratios are smaller than would be expected from the combination of the standard deviations of the single activities. This can be explained by the fact that the error due to the uncertainty in the positioning is strongly reduced when activity ratios are considered.

Table 5 shows the gamma ray activities measured on each corner of the 9 x 9 fuel assemblies.
Table 6 shows the gamma ray activities measured on each corner of the 8 x 8 fuel assemblies, together with the activities of the reference assembly A 001. These values are not directly comparable with the other measurements performed on the 9 x 9 fuel assemblies,

having been obtained in slightly different geometrical conditions.

Table 7 and 8 give the integral measurements of gamma activities and gamma activity ratios on the fuel assemblies with 9 x 9 and 8 x 8 rod array respectively.

In Tables 7 and 8 the values of burnup and Pu/U ratio generated by the BURSQUID code are also reported.

All the activities of the Table 3 to 8 are referred to the date of January 23, 1973.

TABLE 3

MEASUREMENTS ON THE REFERENCE FUEL ASSEMBLY

FUEL ELEMENT	CORNER	Cs 134 605 KeV x 10^4	Cs 137 662 KeV x 10^4	Cs 134 796 + 802 KeV x 10^4	Zr 95 724 KeV x 10^3	Zr + Nb 95 757 + 766 KeV x 10^4
A001	1	1.910	3.093	3.220	9.749	6.807
	2	2.123	3.363	3.779	9.823	6.655
	3	1.900	3.022	3.191	10.444	7.082
	4	1.543	2.709	2.675	10.987	7.159
		7.476	12.187	12.865	41.003	27.703
A001	1	1.705	3.037	3.141	9.744	6.878
	2	2.037	3.233	3.540	9.425	6.404
	3	1.724	2.815	2.987	9.778	6.789
	4	1.498	2.605	2.556	10.070	6.738
		6.964	11.690	12.224	39.017	26.809
A001	1	1.879	3.057	3.119	9.940	6.834
	2	2.054	3.177	3.488	9.640	6.287
	3	1.748	2.937	3.007	9.423	6.848
	4	1.545	2.634	2.667	10.541	6.943
		7.226	11.805	12.281	39.544	26.912
A001	1	1.751	2.923	3.159	10.582	6.816
	2	2.099	3.301	3.614	10.183	6.494
	3	1.805	2.937	3.041	9.650	6.811
	4	1.534	2.677	2.646	10.367	7.137
		7.189	11.838	12.460	40.782	27.258
A001	1	1.831	3.014	3.179	9.510	6.779
	2	2.073	3.248	3.531	9.992	6.412
	3	1.873	3.159	3.301	10.198	7.216
	4	1.566	2.788	2.854	10.788	7.514
		7.343	12.209	12.865	40.488	27.921
A001	1	1.852	2.954	3.153	9.530	6.622
	2	2.085	3.271	3.569	9.859	6.492
	3	1.760	3.006	3.106	10.015	6.861
	4	1.644	2.745	2.765	10.683	7.081
		7.341	11.976	12.593	40.087	27.056
A001	1	1.903	3.058	3.158	9.788	6.722
	2	2.140	3.309	3.576	9.361	6.664
	3	1.820	3.016	3.186	10.014	7.232
	4	1.614	2.759	2.809	10.452	7.304
		7.477	12.142	12.729	39.615	27.922

TABLE 4

REPRODUCIBILITY OF THE INTEGRAL MEASUREMENTS ON THE REFERENCE FUEL ASSEMBLY

Fuel Element	Cs 134 605 KeV x 10^4	Cs 137 662 KeV x 10^5	Cs 134 796+802 KeV x 10^5	Cs134(605) / Cs 137	Cs134(769+802) / Cs 137	Zr+Nb95 757+766KeV x 10^5	Zr 95 724 KeV x 10^4
A001	7.476	1.219	1.286	0.6133	1.055	2.770	4.100
	6.964	1.169	1.222	0.5957	1.045	2.681	3.902
	7.226	1.180	1.228	0.6124	1.041	2.691	3.954
	7.189	1.184	1.246	0.6072	1.052	2.726	4.078
	7.343	1.221	1.286	0.6014	1.053	2.792	4.049
	7.341	1.198	1.259	0.6128	1.051	2.706	4.009
	7.477	1.214	1.273	0.6159	1.048	2.792	3.961
Mean Value	7.288	1.198	1.257	0.6084	1.049	2.737	4.007
S.D. %	2.48	1.73	2.09	1.20	0.47	1.73	1.80

TABLE 5

MEASUREMENTS ON THE FUEL ASSEMBLIES WITH A 9 x 9 ROD ARRAY

FUEL ELEMENT	CORNER	Cs 134 605 KeV x 10^4	Cs 137 662 KeV x 10^4	Cs 134 796 + 802KeV x 10^4	Zr 95 724 KeV x 10^3	Zr 95 + Nb 95 757 + 766 KeV x 10^4
A083	1	1.498	2.752	2.798	10.588	6.899
	2	1.418	2.263	2.551	10.110	6.989
	3	2.205	3.356	3.026	11.078	7.191
	4	2.144	3.433	3.865	10.476	6.887
		7.265	11.804	12.240	42.252	27.966
A053	1	2.018	3.348	3.508	8.962	6.088
	2	1.749	2.961	3.131	9.864	6.680
	3	1.680	2.850	2.918	10.356	6.935
	4	1.920	3.077	3.313	9.898	6.444
		7.367	12.236	12.870	39.080	26.147
A219	1	2.358	3.505	3.889	10.041	6.585
	2	1.931	3.267	3.413	9.520	6.510
	3	1.500	2.593	2.549	9.365	6.491
	4	1.915	3.033	3.050	9.863	6.362
		7.704	12.398	12.901	38.789	25.948
A160	1	2.173	3.614	3.587	7.784	5.420
	2	2.042	3.581	3.455	8.091	5.301
	3	1.694	3.320	3.024	8.062	4.985
	4	1.923	3.492	3.310	7.562	5.143
		7.832	14.007	13.376	31.499	20.849
A195	1	1.278	2.804	2.250	5.638	3.925
	2	1.767	3.466	3.098	6.606	3.991
	3	2.003	3.454	3.350	6.967	4.612
	4	1.459	2.982	2.598	7.163	4.545
		6.507	12.706	11.296	26.374	17.073
A217	1	1.952	3.488	3.402	6.713	4.639
	2	1.437	2.875	2.384	5.923	4.150
	3	1.351	2.771	2.192	5.400	3.798
	4	1.808	3.286	3.100	6.012	4.041
		6.548	12.420	11.078	24.048	16.628
A147	1	2.081	3.924	3.581	4.977	3.224
	2	1.485	3.380	2.646	3.562	2.360
	3	1.374	3.259	2.353	4.037	2.356
	4	1.775	3.517	3.053	4.301	2.851
		6.715	14.080	11.633	16.877	10.791
A154	1	1.183	3.121	2.107	2.839	2.089
	2	1.446	3.326	2.413	3.424	2.217
	3	2.030	3.851	3.482	5.173	3.061
	4	1.689	3.485	2.907	3.962	2.544
		6.348	13.783	10.909	15.398	9.911

TABLE 6

MEASUREMENTS ON THE FUEL ASSEMBLIES WITH A 8 x 8 ROD ARRAY

FUEL ELEMENT	CORNER	Cs 134 605 KeV x 10^4	Cs 137 662 KeV x 10^4	Cs 134 796+802 KeV x 10^4	Zr 95 724 KeV x 10^3	Zr 95 + Nb 95 757+766 KeV x 10^4
SB51	1	0.4942	1.370	0.7861	13.504	9.434
	2	0.5395	1.503	0.9079	13.862	9.551
	3	0.5214	1.545	0.9511	13.382	9.549
	4	0.9652	1.538	0.5646	13.215	9.449
		2.520	5.956	3.210	53.963	37.983
SA16	1	1.558	2.840	2.770	11.605	8.048
	2	1.768	2.978	3.033	11.478	8.027
	3	1.801	2.995	3.033	11.736	7.946
	4	1.595	2.850	2.826	11.150	7.571
		6.722	11.663	11.662	45.969	31.592
A001 (Refer-ence)	1	1.921	3.158	3.281	10.458	7.054
	2	2.258	3.579	3.778	10.524	6.913
	3	1.905	3.098	3.265	10.894	7.320
	4	1.548	2.901	2.802	10.445	7.443
		7.632	12.736	13.126	42.321	28.730

TABLE 7

INTEGRAL MEASUREMENTS ON THE FUEL ASSEMBLIES WITH A 9 x 9 ROD ARRAY

FUEL ELEMENT	THEOR.PREDICTIONS		Cs 134 605 KeV	Cs 137 662KeV	Cs 134 796+802	Cs 134(605) / Cs 137	Cs134(796+802) / Cs 137	Zr+Nb 95 757+766KeV	Zr 95 724KeV
	BURNUP MWD/MTU	Pu/U $\times 10^{-2}$	$\times 10^4$	$\times 10^5$	$\times 10^5$			$\times 10^5$	$\times 10^4$
A001	15591	0.644	7.288	1.198	1.257	0.6084	1.049	2.737	4.007
A083	15568	0.643	7.265	1.180	1.224	0.6157	1.037	2.797	4.225
A053	15930	0.651	7.367	1.224	1.287	0.6019	1.051	2.615	3.908
A219	15861	0.649	7.704	1.240	1.290	0.6213	1.040	2.595	3.879
A160	17960	0.691	7.832	1.401	1.338	0.5590	0.9550	2.085	3.150
A195	16480	0.664	6.507	1.271	1.130	0.5119	0.8891	1.707	2.637
A217	16425	0.662	6.548	1.242	1.108	0.5272	0.8921	1.663	2.405
A147	18277	0.697	6.715	1.408	1.163	0.4769	0.8260	1.079	1.688
A154	18153	0.695	6.348	1.378	1.091	0.4607	0.7917	0.991	1.540

TABLE 8

INTEGRAL MEASUREMENTS ON THE FUEL ASSEMBLIES WITH A 8 x 8 ROD ARRAY

FUEL ELEMENT	THEOR.PREDICTIONS		Cs 134 605KeV	Cs 137 662KeV	Cs 134 796+802	Cs 134(605) / Cs 137	Cs134(796+802) / Cs 137	Ze+Nb95 757+766KeV	Zr 95 724KeV
	BURNUP MWD/MTU	Pu/U $\times 10^{-2}$	$\times 10^4$	$\times 10^5$	$\times 10^5$			$\times 10^5$	$\times 10^4$
SB 51	7298	0.326	2.520	0.5956	0.3210	0.4231	0.5389	3.798	5.396
SA 16	14350	0.531	6.722	1.166	1.166	0.5765	1.000	3.159	4.597
A001 (Refer.)	15591	0.644	7.632	1.274	1.313	0.5990	1.031	2.873	4.232

6. CORRELATION BETWEEN Cs 137 ACTIVITY AND BURNUP

In Table 9 the ratios between Cs 137 activity and burnup are reported for the fuel assemblies with a 9 x 9 rod array. Correction factors have been introduced in the experimental values of Cs 137 activity to take into account the variations in the Cs 137 decay and in the gamma ray attenuation between the different fuel assemblies. The correction factors have been normalized to the value 1 for the reference fuel assembly A001. The corrections are smaller than 2 % for all the fuel assemblies with a 9 x 9 rod array.

The decay corrections have been calculated by means of the computer code ATTIVA taking into account the irradiation histories of the fuel assemblies.
The attenuation corrections have been calculated by means of the computer code ATTENUA taking into account the rod-by-rod burnup distribution, having assumed that the Cs 137 activity distribution coincides with the burnup distribution given by the BURSQUID code.
The calculations have shown that for the fuel assemblies with a 9 x 9 rod array the 662 KeV gamma ray attenuation is constant within 0.5 %.

In Table 10 the ratios between Cs 137 activity and burnup are reported for the fuel assemblies with a 8 x 8 rod array and for the reference fuel assemblies A001.
Corrections have been introduced in the Cs 137 activity of the fuel assemblies with a 8 x 8 rod array to take into account the differences in decay and gamma ray attenuation with respect to the reference fuel assembly.

A correction has also been introduced to take into account the different densities of the fuel in the assemblies with a 9 x 9 and a 8 x 8 rod array ; this correction is required because the measured activity is expressed as gamma rays per volume unit while the burnup is expressed as energy produced per weight unit.
The total corrections introduced in the Cs 137 activities for the fuel assemblies SB 51 and SA 16 are smaller than 2 %.

The ratios between Cs 137 activity and burnup reported in Table 9 are quite constant (standard deviation 1.5 %). Also the ratios reported in Table 10 are constant within 1 %.
These results indicate the possibility of determining the burnup relative distribution in the core by means of non-destructive measurements on the fuel assemblies. On the basis of the results of Table 10 it seems possible also to compare fuel assemblies with different rod arrays and with large burnup differences. In fact, the fuel assemblies SB 51 and SA 16 have burnup values of about 7,000 and 14,000 MWD/MTU respectively.

The corrections to be introduced in the Cs 137 activity to take into account differences in rod array and irradiation history are normally small and it seems possible in most cases to operate without introducing any correction.

The results obtained in this experiment agree with the results of the Trino Vercellese experiment[2,3]. In fact in the Trino Vercellese experiment the ratic between Cs 137 activity and burnup was found constant with a standard deviation of 2.3 %.

TABLE 9

RATIOS BETWEEN Cs 137 ACTIVITY AND BURNUP FOR THE FUEL ASSEMBLIES WITH A 9 x 9 ROD ARRAY

Fuel Assembly	Burnup Theor.Pred. MWD/MTU	Cs 137 662 KeV $\times 10^5$	$\frac{\text{Cs 137}}{\text{Burnup}}$
A001	15591	1.198	7.68
A083	15568	1.177	7.56
A053	15930	1.217	7.64
A219	15861	1.241	7.82
A160	17960	1.423	7.92
A195	16480	1.280	7.77
A217	16425	1.254	7.63
A147	18277	1.427	7.81
A154	18153	1.398	7.70
Mean Value			7.73
Standard Deviation %			1.5

TABLE 10

RATIOS BETWEEN Cs 137 ACTIVITY AND BURNUP FOR THE FUEL ASSEMBLIES WITH A 8 x 8 ROD ARRAY

Fuel Assembly	Burnup Theor.Pred. MWD/MTU	Cs 137 662 KeV $\times 10^5$	$\frac{\text{Cs 137}}{\text{Burnup}}$
SB 51	7298	0.596	8.16
SA 16	14350	1.181	8.23
A001 Reference	15591	1.274	8.17

7. CORRELATION BETWEEN Cs 134/Cs 137 ACTIVITY RATIO AND PLUTONIUM CONTENT

Table 11 reports the ratios between the experimental values of Cs 134/Cs 137 and the theoretical values of Pu/U, determined by means of the BURSQUID code. The experiment previously performed on the fuel assemblies of Trino Vercellese reactor had shown a proportionality between Cs 134/Cs 137 activity ratio and plutonium content in the fuel assemblies[2,3]. This proportionality has not been found for the fuel assemblies of the Garigliano reactor as shown in Table 11. This is due to the fact that the fuel assemblies of the Garigliano reactor experienced different power histories as shown in column 5 of Table 11 where the power values during the last year of irradiation are reported. The half-life of Cs 134 being relatively short (2.05 y), the activity of this nuclide measured on the fuel assemblies is strongly influenced by the power level in the last part of the irradiation. These observations confirm the results of the theoretical evaluations which showed a strong influence of the power history on the Cs 134/Cs 137 activity ratio (see paragraph 4.3).

Before rejecting the possibility of utilization of this correlation, the use of relatively simple computer calculations has been investigated to take into account the effect of the different power histories. In the last column of Table 11 the experimental values for the ratios between Cs 134/Cs 137 and Pu/U are reported, corrected for the decay during the irradiation and for the self-shielding effects. The correction factors have been calculated by means of the ATTIVA and ATTENUA codes.

These data show that the computer calculations in this relatively simple form cannot predict with sufficient accuracy the effect of the power history. As it does not seem convenient to perform more complex calculations for these corrections the use of this correlation cannot be considered generally valid.

TABLE 11

RATIOS BETWEEN Cs 134/Cs 137 AND Pu/U

Fuel Assembly	Pu/U Theor.Pred. Mass Ratio $\times 10^{-2}$	$\frac{\text{Cs 134 (796+802 KeV)}}{\text{Cs 137 (662 KeV)}}$ Activity Ratio	$\frac{\text{Cs 134/Cs 137}}{\text{Pu/U}}$ $\times 10^{2}$	Power During the Last Year	$\frac{\text{Cs 134/Cs 137}}{\text{Pu/U}}$ Corrected Values $\times 10^{2}$
A001	0.644	1.049	1.629	1.02	1.629
A083	0.643	1.037	1.613	1.02	1.616
A053	0.651	1.051	1.614	0.99	1.639
A219	0.649	1.040	1.602	1.00	1.617
A160	0.691	0.955	1.382	0.79	1.623
A195	0.664	0.889	1.339	0.62	1.505
A217	0.662	0.892	1.348	0.62	1.505
A147	0.697	0.826	1.185	0.40	1.526
A154	0.695	0.792	1.140	0.35	1.499

TABLE 12

CORRELATIONS BETWEEN Zr 95 AND Nb 95 ACTIVITIES AND POWER AT THE END OF CYCLE

Fuel Assembly	Power at the End of Cycle (Flare)	Zr-Nb 95 757+766KeV x 10^5	Zr 95 724KeV x 10^4	Zr-Nb 95 / Power x 10^5	Zr 95 / Power x 10^4	Zr-Nb 95 / Power Normalized	Zr 95 / Power Normalized	Zr 95 / Power Nuclear Codes
A001	1.23	2.737	4.007	2.225	3.258	1	1	1
A083	1.25	2.797	4.225	2.238	3.380	1.006	1.037	0.986
A053	1.16	2.615	3.908	2.254	3.369	1.013	1.034	1.022
A219	1.15	2.595	3.879	2.237	3.344	1.005	1.026	1.030
A160	0.86	2.085	3.150	2.397	3.621	1.077	1.111	1.059
A195	0.72	1.707	2.637	2.338	3.612	1.051	1.109	1.081
A217	0.72	1.663	2.405	2.310	3.340	1.038	1.025	1.077
A147	0.44	1.079	1.688	2.452	3.836	1.102	1.177	1.069
A154	0.37	0.991	1.540	2.608	4.053	1.172	1.244	1.098

8. CORRELATION BETWEEN Zr 95 AND Nb 95 ACTIVITIES AND POWER AT THE END OF CYCLE

In Table 12 the power values at the end of cycle for the 9 x 9 fuel assemblies (FLARE code), the activities of Zr-Nb 95 (757+766 KeV) and Zr 95 (724 KeV) and the ratios between activities and power are reported.
The values of these ratios normalized to 1 for the reference fuel assembly A001 are also reported in Table 12 and compared with the theoretical values of the ratios between Zr 95 and power calculated by means of the BURSQUID + ATTIVA codes and FLARE code.

Table 12 indicates that the ratios between activities and power increase with a decrease of power. This effect can be expected since the assemblies which at the end of cycle had a low power level, in the previous period had a higher power level. Due to the fact that Zr 95 had a 65 d. half-life, the Zr-Nb activity emitted by those assemblies was increased by the contributions of previous higher power periods.

This experimental observation is also in agreement with the theoretical predictions obtained by means of the computer codes (see columns 7, 8 and 9 of Table 12). The theoretical and experimental values are in a satisfactory agreement for all the fuel assemblies except the fuel assemblies A147 and A154 which have the lowest values of power at the end of cycle.

9. CONCLUSIONS

The experiment performed on the fuel assemblies of the Garigliano reactor has shown that non-destructive measurements of Cs 137 activity can be utilized for the relative determination of burnup in fuel assemblies. The corrections to be introduced to take into account differences in rod array and irradiation history appear to be small.

On the contrary the correlation between Cs 134/Cs 137 activity ratio and plutonium content is not valid in the Garigliano reactor due to important differences existing in the power history of the fuel assemblies.

On the basis of this result it seems convenient to investigate the possible use of the Eu 154/Cs 137 activity ratio as plutonium indicator. In fact, the half-life of Eu 154 (16 y) makes this nuclide less sensitive than Cs 134 to the differences in the power history of the fuel assemblies. Moreover the theoretical predictions indicate a linearity between Eu 154/Cs 137 and Pu/U for burnup values up to 20,000 MWD/MTU.

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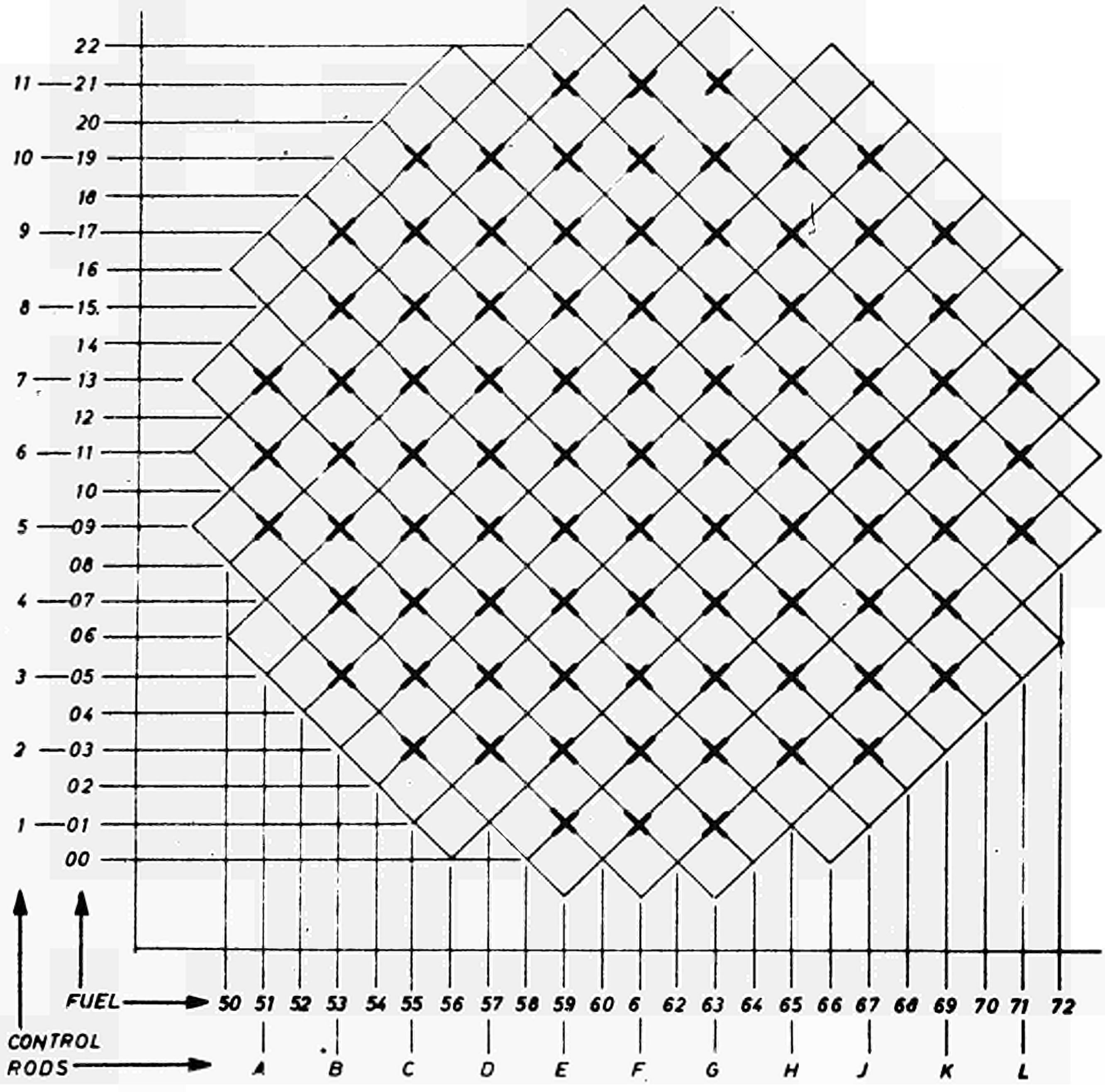

Fig. 1 : Coordinate System of the Locations of the Fuel Assemblies in the Core of the Garigliano Reactor.

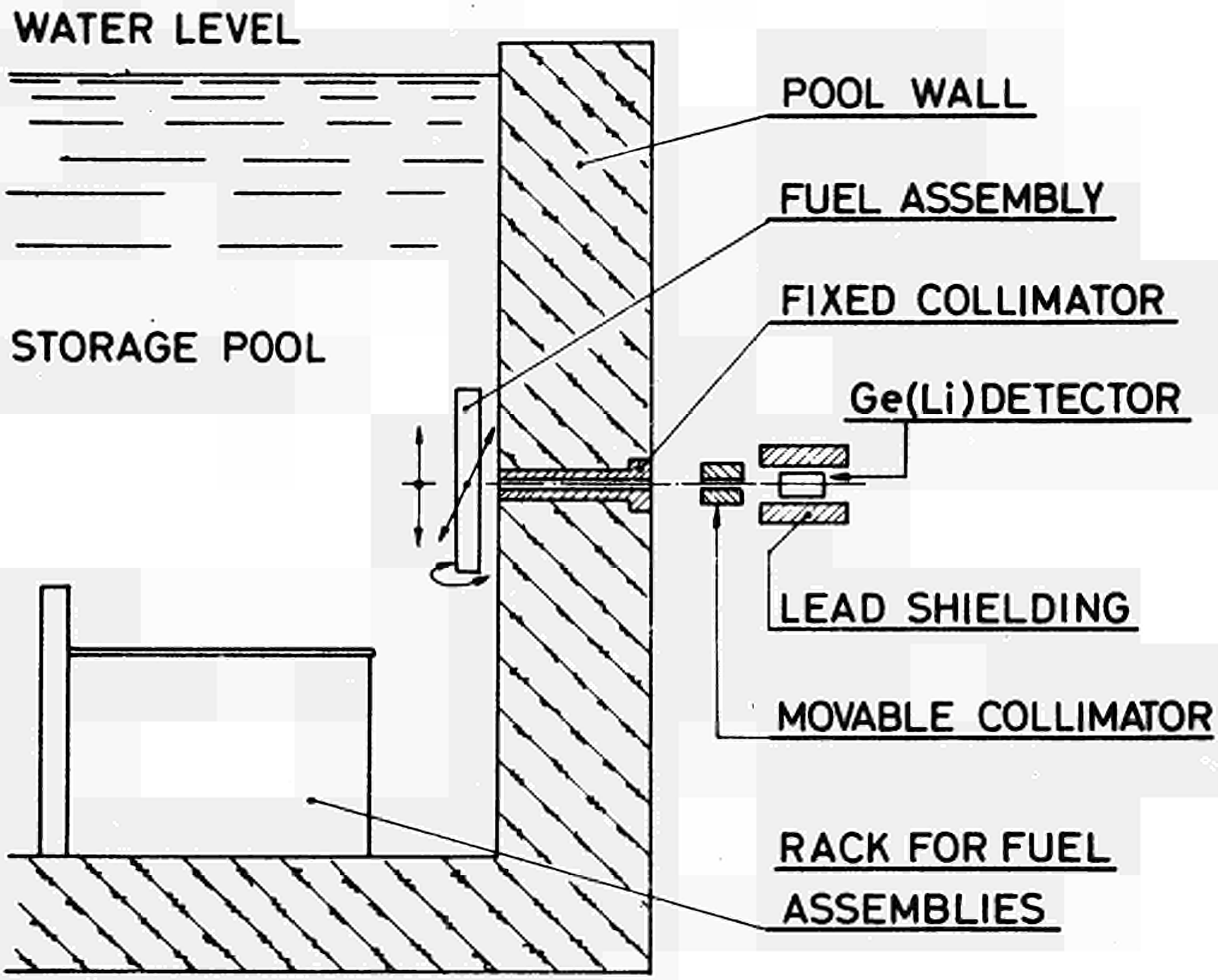

Fig. 2 : Sketch of the Gamma Scanning System

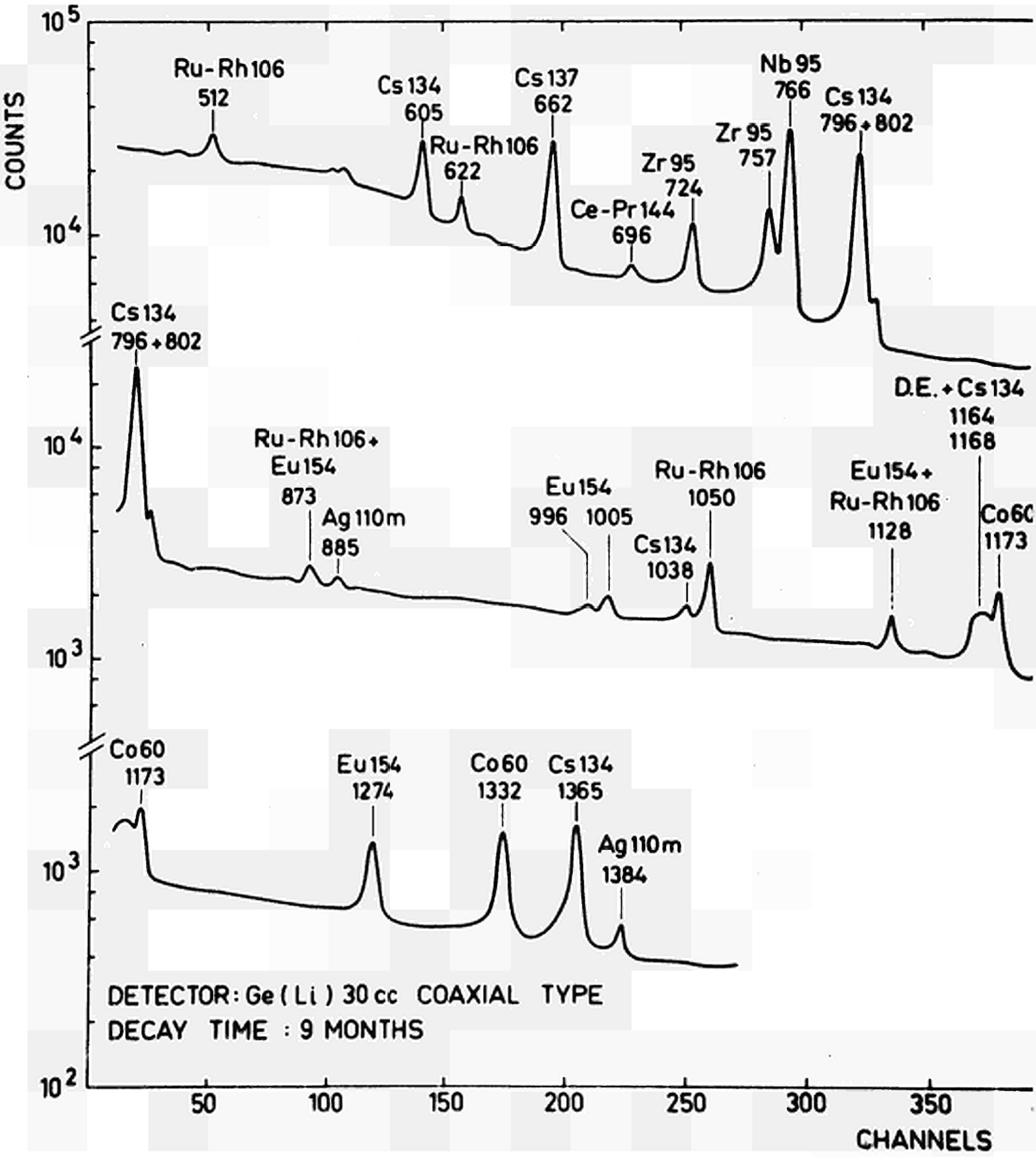

Fig. 3 : Gamma Spectrum of a Spent Fuel Assembly of the Garigliano BWR.

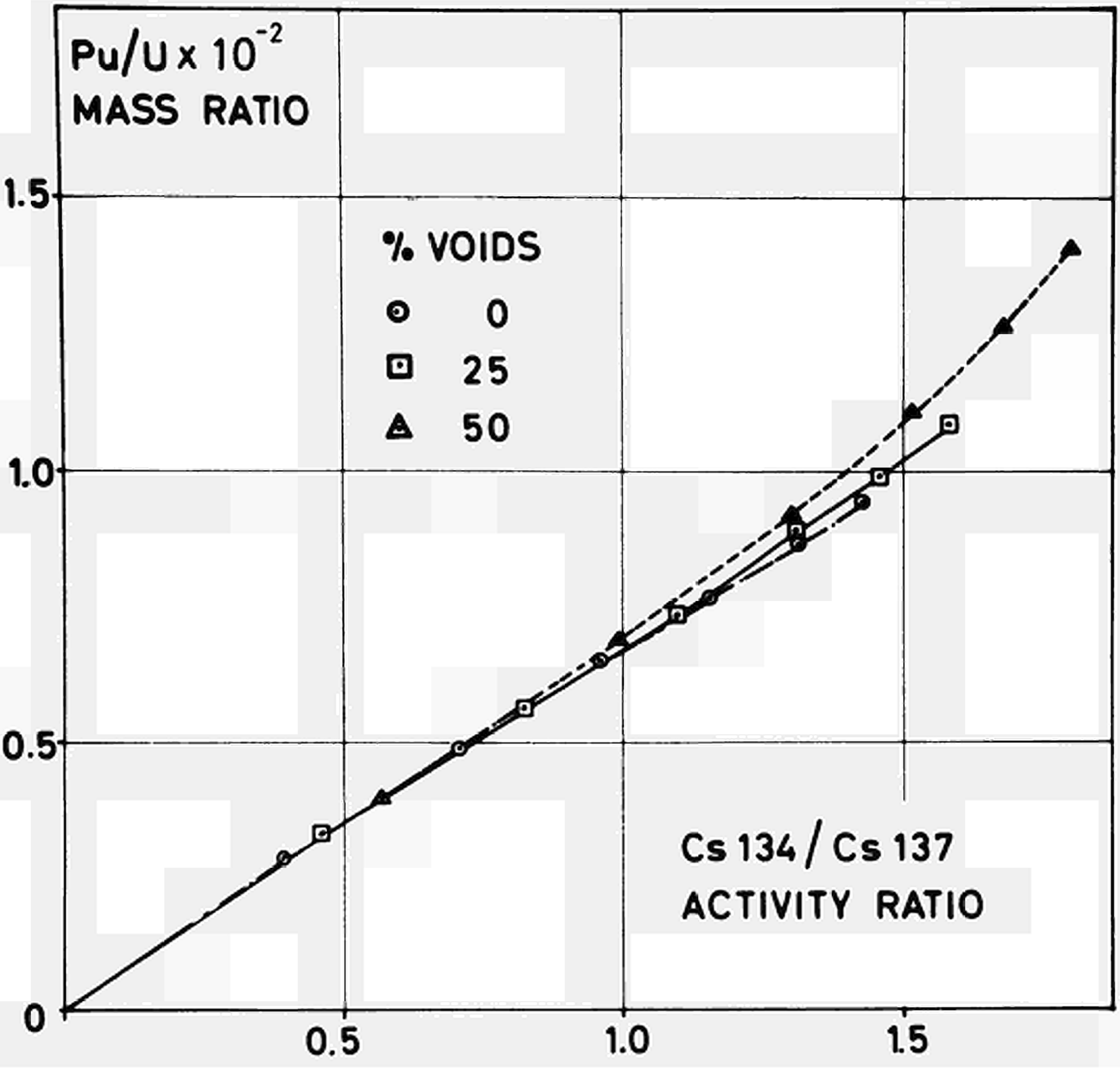

Fig. 4 : Influence of the Voids Fraction on the Correlation Between Cs 134/Cs 137 and Pu/U. Theoretical Evaluation. Each Step corresponds to 5,000 MWD/MTU.

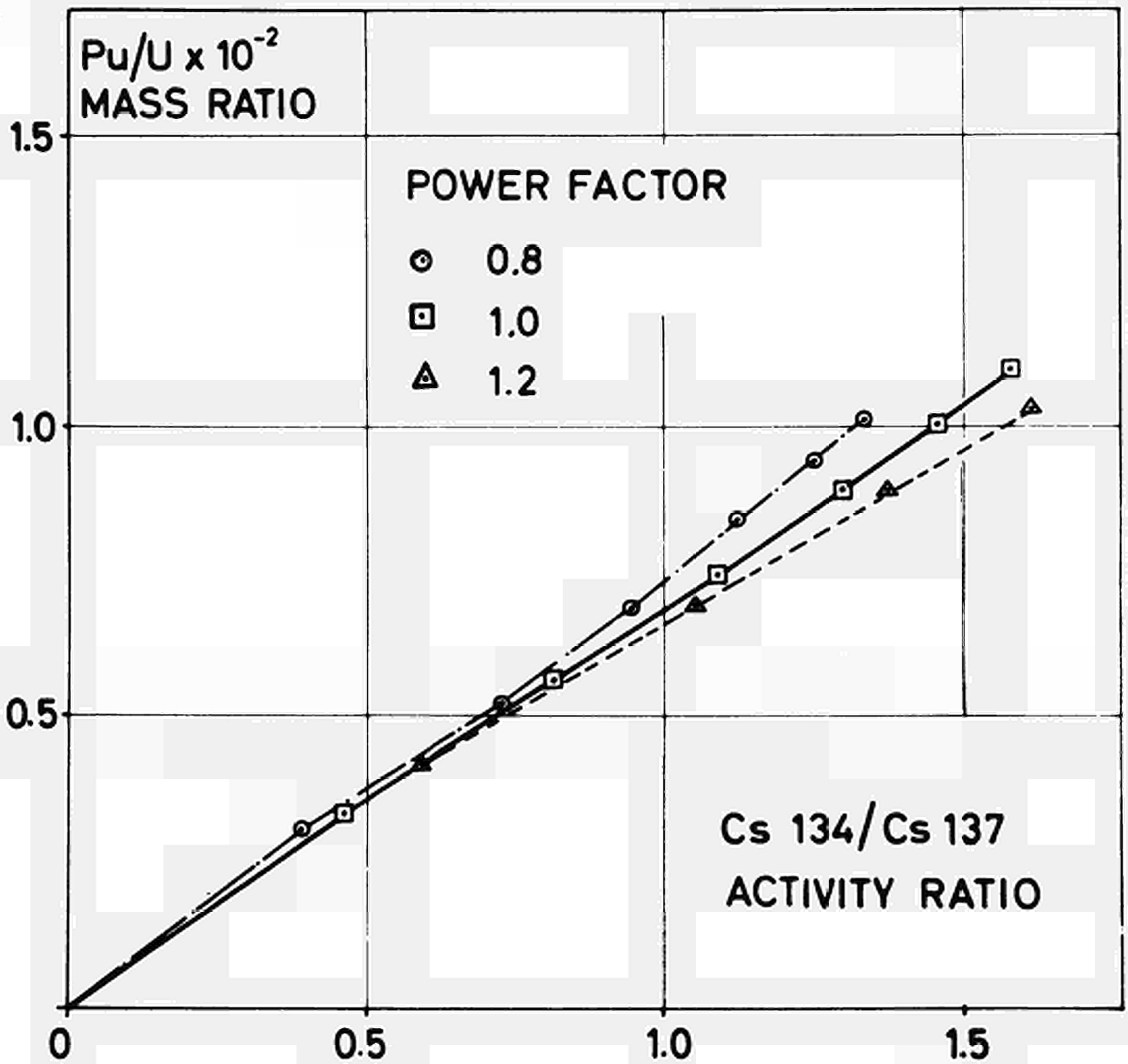

Fig. 5 : Influence of the Power Factor on the Correlation Between Cs 134/Cs 137 and Pu/U. Theoretical Evaluation. Each Step corresponds to 5,000 MWD/MTU.

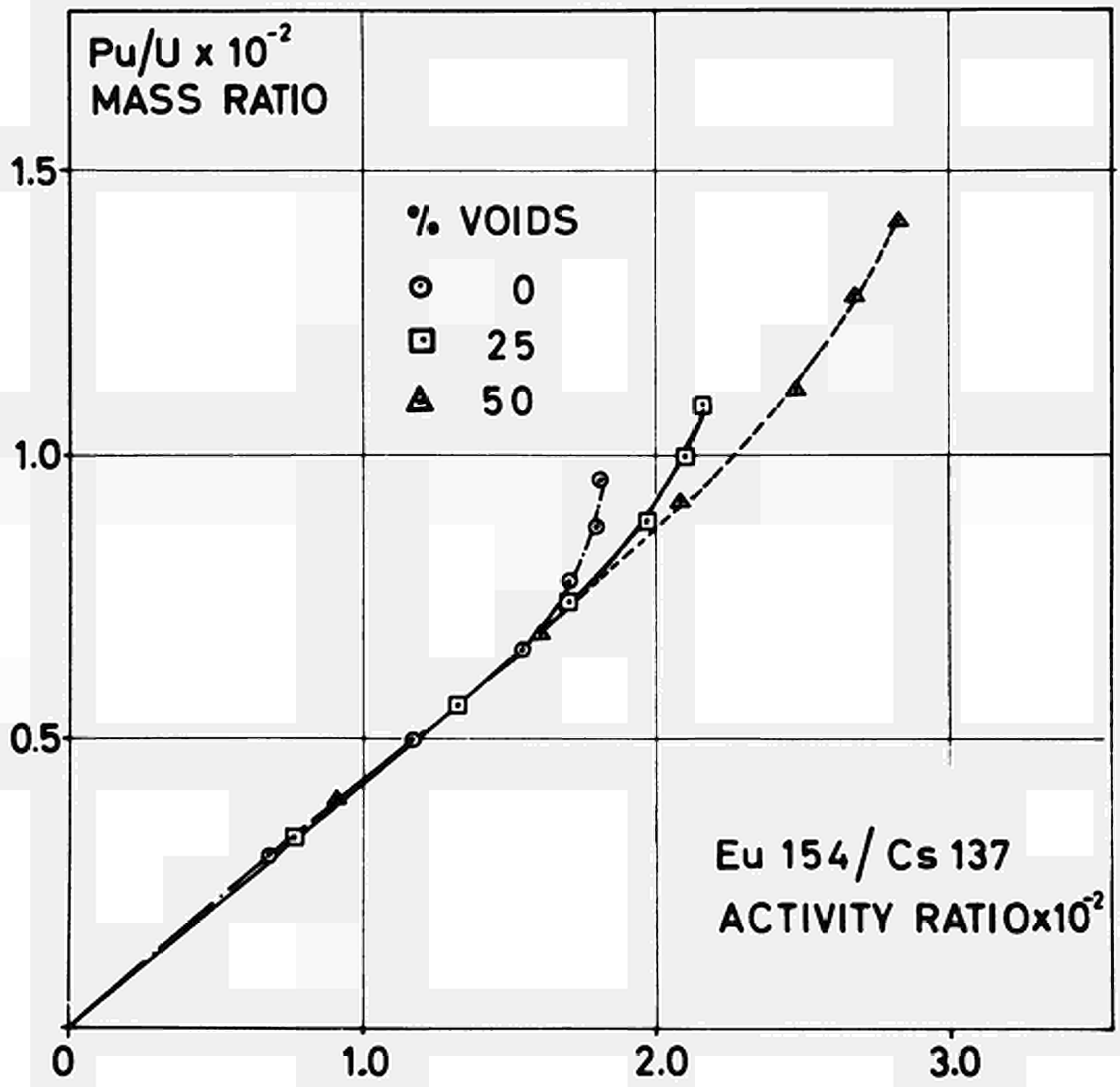

Fig. 6 : Influence of the Voids Fraction on the Correlation Between Eu 154/Cs 137 and Pu/U. Theoretical Evaluation.
Each Step corresponds to 5,000 MWD/MTU.

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Alfred Nobel

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