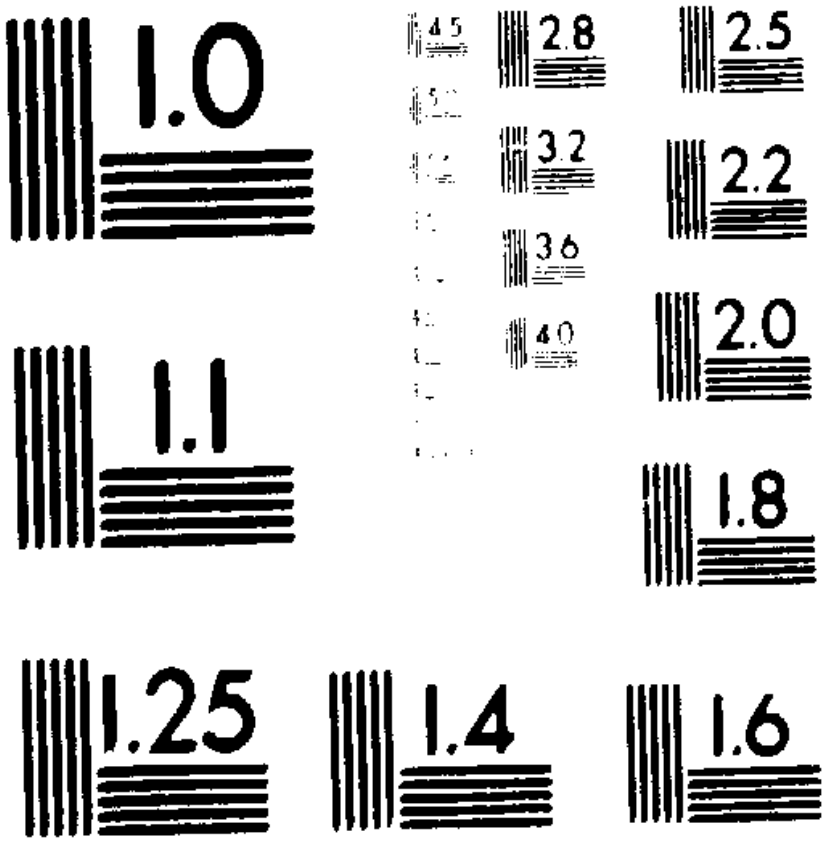

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COMITATO NAZIONALE PER LA RICERCA E PER LO SVILUPPO
DELL'ENERGIA NUCLEARE E DELLE ENERGIE ALTERNATIVE

REIMIEP 1987
AN INTERLABORATORY U-235 ENRICHMENT DETERMINATION BY GAMMA MEASUREMENT ON SOLID UF_6 SAMPLE

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RT/COMB/87/15

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Progetto Enea: Sicurezza impianti di ritrattamento e salvaguardia (CE)

RIASSUNTO

La tecnica della spettrometria gamma, basata sulla misura della riga a 185 KeV dell'U^{235} é ormai correntemente usata per la determinazione dell'arricchimento dell'Uranio in diversi materiali del ciclo del combustibile nucleare quali Uranio metallico, "pellets" di UO_2, UF_6 liquido o solido.

Il presente rapporto descrive l'utilizzo di tale tecnica e i relativi risultati nella determinazione dell'abbondanza isotopica in U^{235} di un campione certificato di UF_6 solido.

Tali misure sono state condotte nell'ambito della partecipazione al " UF_6 Interlaboratory Measurement Evaluation Programme" indetto dal CBNM/Geel con il supporto dell'ESARDA.

SUMMARY

Gamma spectrometry technique, based on the measurement of U^{235} 186 KeV flux, is now currently used for the determination of Uranium enrichment indifferen material of nuclear fuel cycle, namely: Uranium metallic, UO_2 pellets, UF_6 liquid or solid.

The present paper describes the use of such a technique and the obtained results in determining the U^{235}/U atomic isotopic abbundance on a certified UF_6 solid sample.

The measurements have been carried out in the framework of the partecipation to the "UF_6 Interlaboratory Measurement Evaluation Programme" organized by CBNM/Geel with the support of the ESARDA (European Safeguards Research and Development Association).

INTRODUCTION

The UF_6 Interlaboratory Measurement Evaluation Programme 1986 was organized by CBNM/Geel with the support of the European Safeguards Research and Development Association.

The purposes of such a programme are to provide the present state of practice in UF_6 isotopic measurements, to give the participants the opportunity to verify their measurement results relative to those of other laboratories and to locate the measurement results relative to a certified value.

In this framework two different test materials were prepared and characterized, namely:

a) 6g UF_6 in monel bottles for mass spectrometry measurement;

b) 80g UF_6, as a solid sample, contained in monel can with specified cladding, for gamma measurement (fig. 1).

The present paper describes the gamma measurements carried out at COMB-MEPIS Division, for determining the U^{235}/U atomic isotopic abundance on the aforementioned sample.

U^{235} ENRICHMENT MEASUREMENT

The U^{235} enrichment determination is made relative to calibration standards consisting of bulk quantities of reference material with well known U^{235} isotope abundances. The reference material (EC NRM 171), used for system calibration, consists of a set of 5 reference sample with different U^{235} isotope abundances, which are certified with an accuracy of $\leq 0.1\%$ by CBNM [ref. 1].

Since the reference sample cans and the UF_6 container were of different material, two calibration disks, in aluminium and monel, were provided in order to perform measurements with identical windows.

MEASURING HEAD DESCRIPTION

The U^{235} enrichment measurement principles are based on the infinite thickness condition which makes the 186 Kev measured flux dependent only on U^{235} enrichment of the sample.

To satisfy such infinite thickness condition a cylindrical collimator was designed according to the following formula [ref. 2]:

$$D_s = D_c \left(1 + 2s/H_c + 2r_{min}/\sqrt{(D_c^2 + H_c^2)} \right)$$

where:

- D_s is the sample diameter
- r_{min} is the "quasi-infinite" thickness of the sample
- H_c is the collimator height
- D_c is the collimator diameter
- s is the distance between the surface of the sample material and the surface of the collimator including the container wall.

Among the possible values which satisfy the formulas, shown in tab I along with the sample diameter seen by the detector, a lead collimator with 15 mm of diameter and 30 mm of height was chosen in order to obtain the highest gamma transmission for the given sample density (figg. 2a, 2b).

EXPERIMENTAL SET-UP

A HPGe detector, with relative efficency of 10%, has been used for the measurements. The detector head was inserted in a shielded chamber and a plastic collar containing the collimator (fig.3) allowed to perform the measurements with the same geometry. A filter cadmium disk, 1 mm

thick, was placed in front of the detector. The multichannel analyzer was a SILENA CATO 8K with a SILENA amplifier 7611/L. A typical spectrum obtained during the measurements is shown in fig. 4.

Three different methods to evaluate the net peak area of the 186 KeV peak were taken into consideration, namely:

a) Three-window counting with linear background.
This procedure uses the gross counts acquired in the peak window and in two background windows on both sides of the peak. The peak window selected was between 184.26 and 187.14 KeV, while the background windows were chosen between 179.6 and 180 and 188.95 and 189.36 KeV, respectively. The background subctraction was carried out by approximating the background with a stright line going through the midpoints of the two windows.

b) 2.5 FWHM with dispersed step-like background.
Evaluation of peak area is obtained by a best fit procedure, of a gaussian function, carried out on the net counts of a number of channels equal to 2.5 times the full width at half maximum for the peak. The net counts are determined by subtracting a backgraund which appears as a smoothed step below the peak, with a step height proportional to the integral net counts.

c) FWHM+2Ch. with dispersed step-like background.
This procedure is based on the same method as the previous one, but the number of channels used for the fitting procedure are equal to the FWHM plus two channels on both sides.

INSTRUMENT CALIBRATION

The relation of the 186 KeV gamma counting rate and the U^{235} enrichment can be expressed by the following formulas:

$$enr = a \cdot N + b$$

where N is the observed 186 KeV net peak counting rate and offset b takes into account gamma interferences due to gamma emissions from radioactive nuclei other than U^{235}, which overlap the 186 Kev region or affect the determination of the back ground continuum below the peak.

Calibration of the measuring system was carried out by measuring three out of five samples of the reference material set NRM 171.

Tables II,III, and IV report the net peak area evaluations, along with the uncertainties, as determined by the three methods, for the calibration samples and for the UF_6 unknown sample.

The parameters a and b were estimated from weighed least squares fitting, with errors in both directions, of experimental data.

FINAL RESULTS

The U^{235} atomic abundance of the UF_6 sample has been obtained by the aforementioned formulas. A matrix correction factor (1.0228) was introduced in order to take into account the different attenuation of photons in the two matrix material. Table V reports the final values of the unknown sample as determined according to the different net peak area evaluations.

Certified values of both test materials for DA and NDA interlaboratory measurements are reported in fig. 5.

Fig. 6 is a copy of the data sheet sent to CBNM/Geel for transmission of the final result.

Fig. 7 represents a graphical display of the results of the UF6 measurement round 1986 as distributed by CBNM/Geel.

REFERENCE

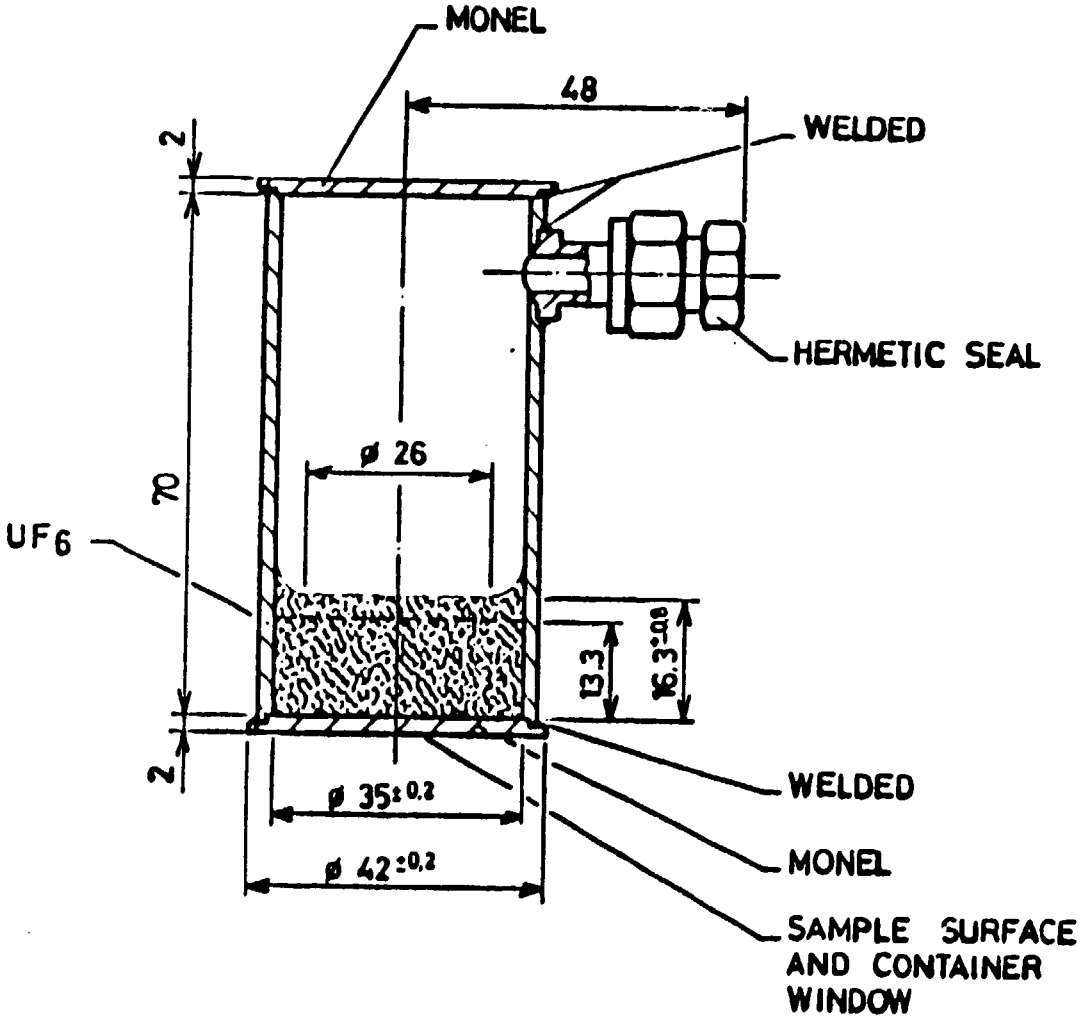
1. P. De Bievre et al.
"235 *uranium isotope abundance certified reference* material for gamma spectrometry - EC nuclear material 171 certification report"
COM 4153 - 1985

2. P. Matussek
"Accurate determination of the U^{235} isotope abundance by gamma spectrometry - A User's Manual for the Certified Reference Material EC-NRM171/NBS-SRM-969"
Kernforshungszentrum Kalsruhe, KfK 3752.

COLLIMATOR CHOICE

Dc= 5	Hc= 5	Ds=31.81
Dc= 5	Hc=10	Ds=20.90
Dc= 5	Hc=15	Ds=16.08
Dc= 5	Hc=20	Ds=13.45
Dc= 5	Hc=25	Ds=11.82
Dc= 5	Hc=30	Ds=10.71
Dc= 5	Hc=35	Ds= 9.90
Dc= 5	Hc=40	Ds= 9.30
Dc=10	Hc=15	Ds=30.09
Dc=10	Hc=20	Ds=25.90
Dc=10	Hc=25	Ds=23.08
Dc=10	Hc=30	Ds=21.08
Dc=10	Hc=35	Ds=19.59
Dc=10	Hc=40	Ds=18.45
Dc=15	Hc=25	Ds=33.49
DC=15	Hc=30	Ds=30.90
Dc=15	Hc=35	Ds=28.91
Dc=15	Hc=40	Ds=27.34

Tab.1



UF6 SAMPLE

FIG. 1

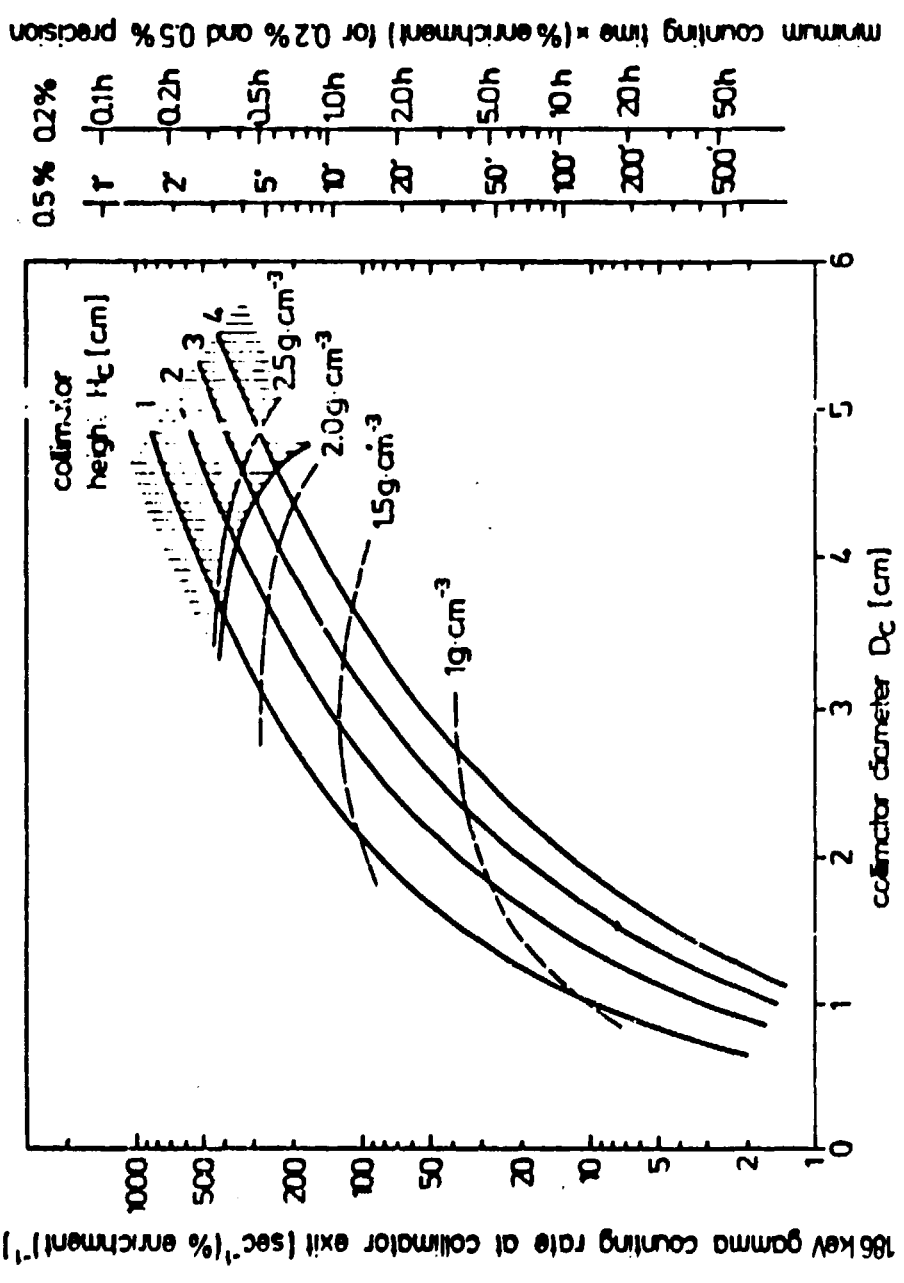
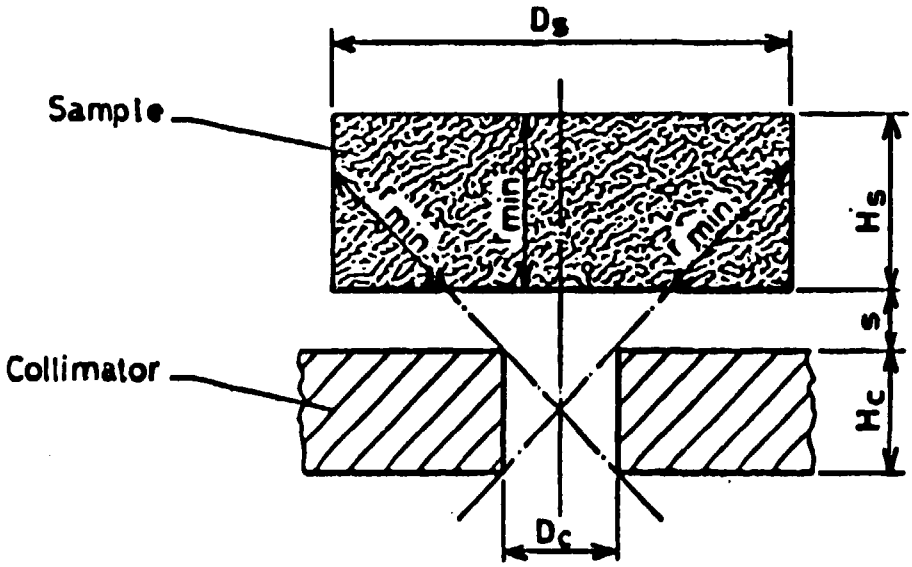
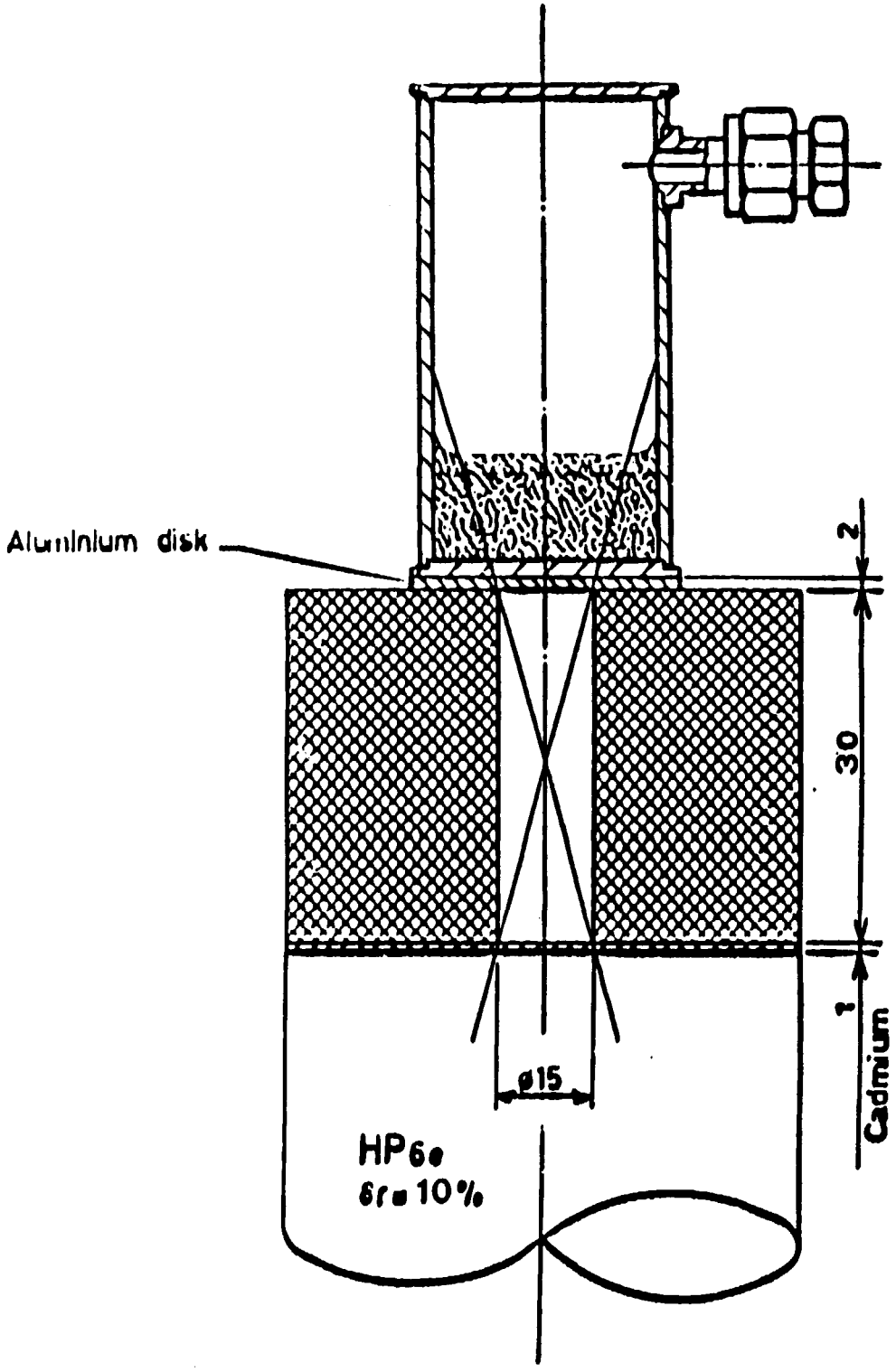


Fig.2a Recommended collimator dimensions, expected counting rates and approximate counting times.



COLLIMATOR CHOICE

FIG. 2b



GEOMETRICAL POSITION

FIG. 3

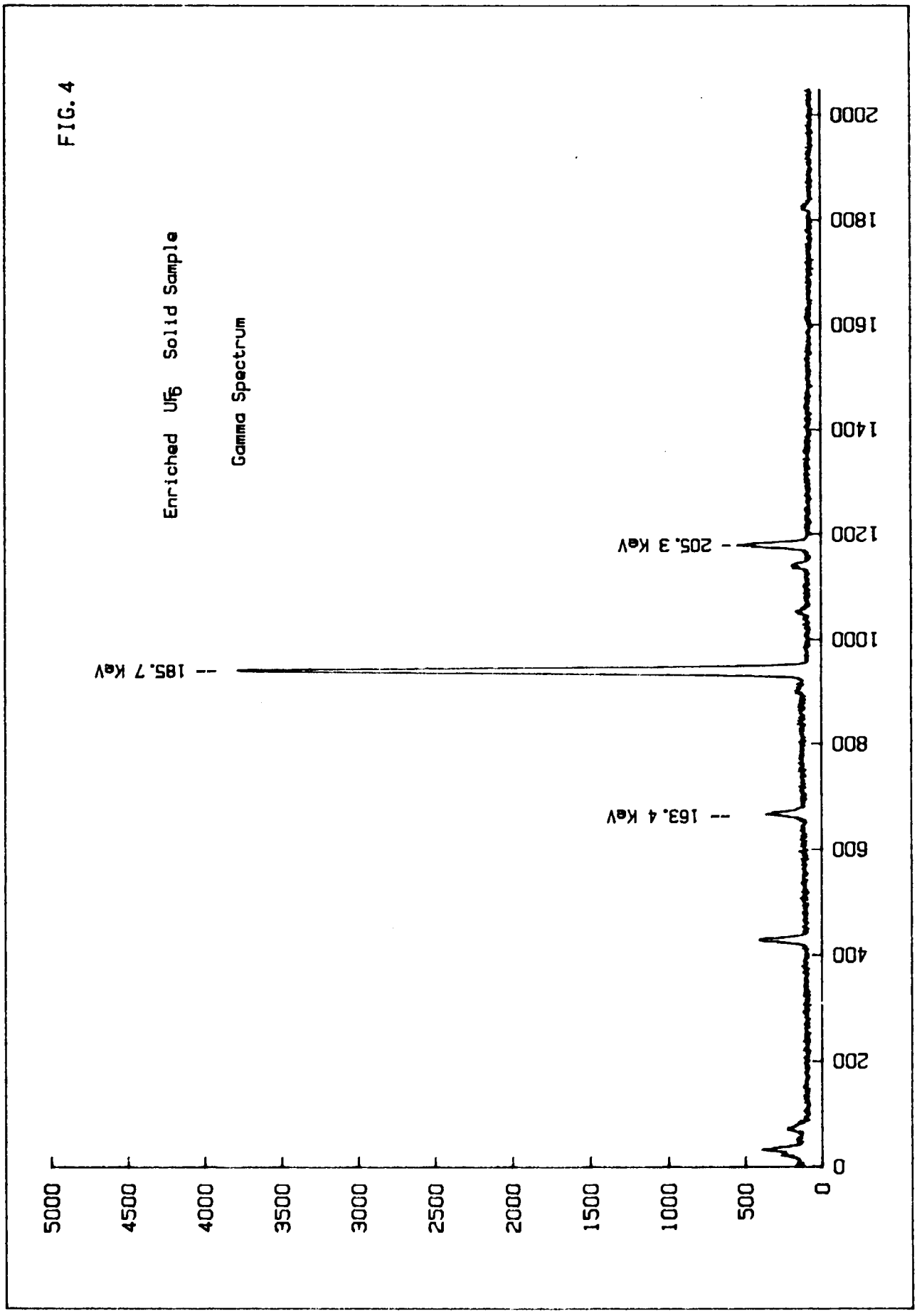


FIG. 4
Enriched UF_6 Solid Sample
Gamma Spectrum
-- 185.7 KeV
-- 205.3 KeV
-- 163.4 KeV
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
0
200
400
600
800
1000
1200
1400
1600
1800
2000

TABLE 2 - Measurement with CICERO M.C.A.
Counting time 36000 sec.
Three-window counting with linear background

Sample	Atom %	Net area at 185.72 KeV	Error % (2s)
U-Stand. + monel disk	4.5168	557447	.32
		557355	.32
		558013	.32
		$\bar{x}$ 557605	$\bar{x}$ .18
U-Stand. + monel disk	2.9857	370093	.42
		369035	.42
		$\bar{x}$ 369564	$\bar{x}$ .30
U-Stand. + monel disk	1.9664	244199	.56
		244180	.56
		$\bar{x}$ 244189	$\bar{x}$ .40
UF6 + alumin. disk	unknown	423096	.36
		424173	.36
		423894	.36
		$\bar{x}$ 423721	$\bar{x}$ .20

Measured E% of UF6 unknown sample: 3.5067 +/-.2059%

TABLE 3 - Measurement with CICERO M.C.A.
Counting time 36000 sec.
2.5 FWHM with dispersed step-like background

Sample	Atom %	Net area at 185.72 KeV	Error % (2s)
U-Stand. + monel disk	4.5168	556037 556150 558343 --- $\bar{X}$ 556843	.56 .56 .56 --- $\bar{X}$ .32
U-Stand. + monel disk	2.9857	369631 367960 --- $\bar{X}$ 368795	.70 .70 --- $\bar{X}$ .50
U-Stand. + monel disk	1.9664	242940 242238 --- $\bar{X}$ 242589	.88 .88 --- $\bar{X}$ .64
UF6 + alumin. disk	unknown	423512 422370 422829 --- $\bar{X}$ 422903	.64 .64 .64 --- $\bar{X}$ .36

Measured E% of UF6 unknown sample: 3.5074 +/-.2937%

TABLE 4 - Measurement with CICERO M.C.A.
Counting time 36000 sec.
FWHM+2 Ch. with dispersed step-like background

Sample	Atom %	Net area at 185.72 KeV	Error % (2s)
U-Stand. + monel disk	4.5168	559529	.34
		558944	.34
		564087	.34
		$\bar{X}$ 560853	$\bar{X}$ .20
U-Stand. + monel disk	2.9857	368981	.42
		367551	.42
		$\bar{X}$ 368266	$\bar{X}$ .30
U-Stand. + monel disk	1.9664	244425	.50
		244916	.50
		$\bar{X}$ 244670	$\bar{X}$ .36
UF6 + alumin. disk	unknown	421545	.38
		428511	.38
		428792	.38
		$\bar{X}$ 426282	$\bar{X}$ .22

Measured E% of UF6 unknown sample: 3.5114 +/-.2378%

TABLE 5

Peak area evaluation	UF6 (atom%)	Error% (2s)
Three-window counting with linear background	3.5067	.2059
2.5 FWHM with dispersed step like background	3.5074	.2937
FWHM + 2 Ch. with dispersed step like background	3.5114	.2378

Regular European Interlaboratory Measurement Evaluation Programme

REIMEP, UF_6 1986

DATA SHEET

Laboratory : ENEA - COMB-MEPIS Laboratory Code* : 15

Sample identification
number : 8

MS	Atomic isotope abundance or ratio	Accuracy (±2s)	Accuracy (±2s) in % relative to value	**
$^{235}U/U$		±		
$^{235}U/^{238}U$		±		
$^{234}U/^{238}U$		±		
$^{236}U/^{238}U$		±		

MS	Abundance in Mass %	Accuracy (±2s)	Accuracy (±2s) in % relative to value	**
^{234}U		±		
^{235}U		±		
^{236}U		±		
^{238}U		±		

NDA	Atomic isotope abundance	Accuracy (±2s)	Accuracy (±2s) in % relative to value	**
$^{235}U/U$	3.5067	+/- .00722	.2059	

Time period of measurement : 2/3/87 - 6/3/87

Signature : Date : 13/3/1987

Fig. 5

Regular European Interlaboratory Measurement Evaluation Programme

REIMEP, UF_6 1986

DATA SHEET

Laboratory : ENEA Laboratory Code : 15

CERTIFIED VALUES :

MS	Atomic isotope abundance or ratio	Accuracy (±2s)
$^{235}U/U$	0. 035 001	± 0. 000 010
$^{235}U/^{238}U$	0. 036 295	± 0. 000 011
$^{234}U/^{238}U$	0. 000 203 3	± 0. 000 004 4
$^{236}U/^{238}U$	0. 000 326 3	± 0. 000 003 6

MS	Abundance in Mass %	Accuracy (±2s)
^{234}U	0. 019 28	± 0. 000 42
^{235}U	3. 457 96	± 0. 001 01
^{236}U	0. 031 22	± 0. 000 34
^{238}U	96. 491 54	± 0. 001 15

NDA	Atomic isotope abundance	Accuracy (±2s)
$^{235}U/U$	0. 035 001	+/- 0. 000 010

Fig. 6

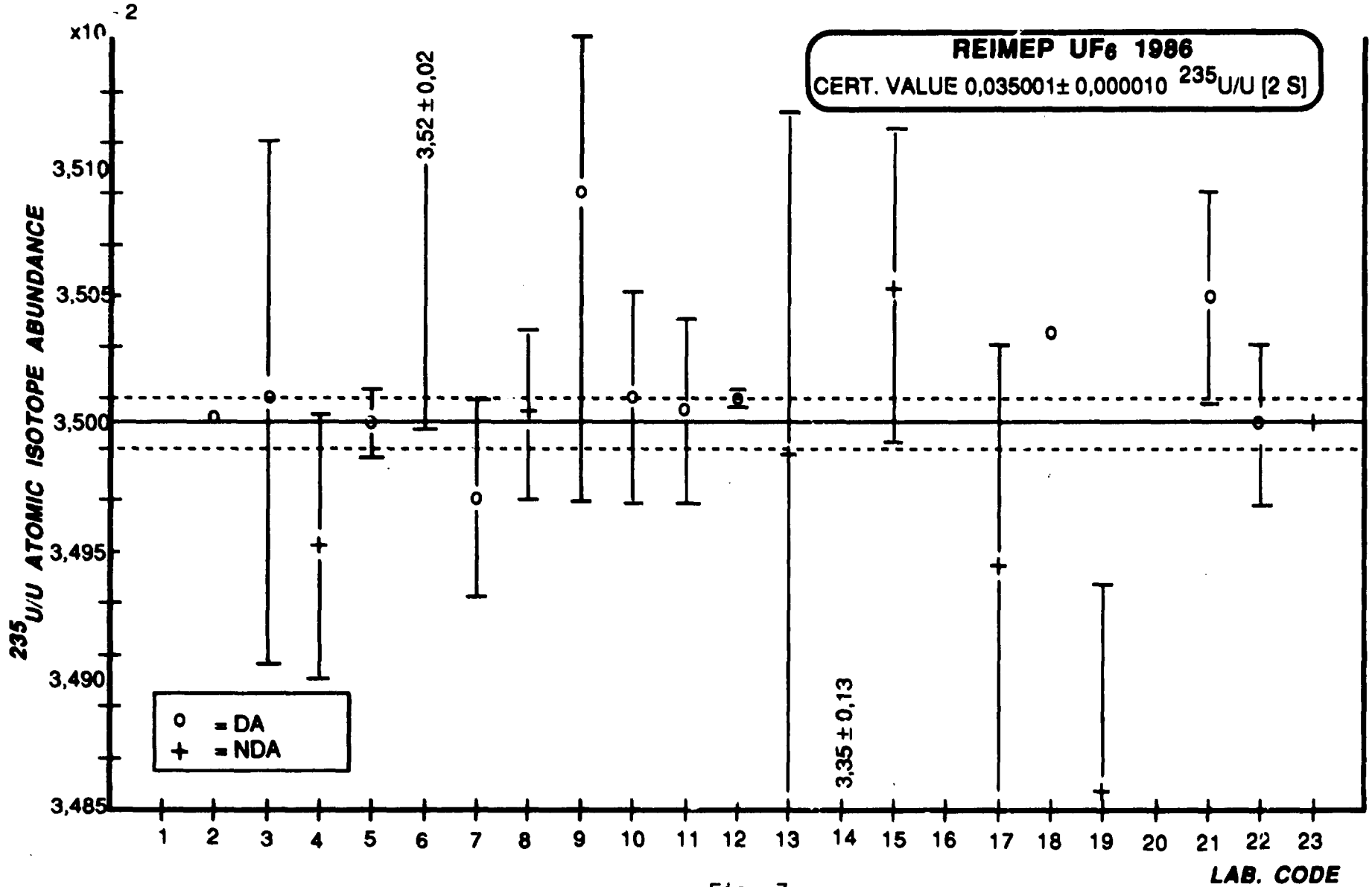

REIMEP UF6 1986
CERT. VALUE 0,035001± 0,000010 235U/U [2 S]
x10-2
3,510
3,505
3,500
3,495
3,490
3,485
235U/U ATOMIC ISOTOPE ABUNDANCE
3,52 ± 0,02
3,35 ± 0,13
o = DA
+ = NDA
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
LAB. CODE

Fig. 7

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