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GEEL-BELGIUM

Progress report 1978

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I N T R O D U C T I O N

The annual progress report of CBNM reappears after a lapse of a year. As before, its purpose is to summarize the laboratory's scientific and technical activity during the year so that interested scientists elsewhere may be kept informed of work in progress and correspond, if they so wish, directly with these engaged on work which interests them.

In this way, the report stands alongside annual reports from other research laboratories which are one part of regular scientific exchange, supplementing publication in the open literature and communications to conferences. For the first time we are including some information on many of the supporting activities without which a laboratory would not be able to function.

The modernized accelerators for neutron production have been operating at their design specifications. On the linac the new mercury-cooled uranium target has operated very satisfactorily, whilst on the large Van de Graaff, ion source developments have improved mean life times, by about a factor of 3, to 450 hours, and an extra beam line has been installed to minimize change-over problems between different experiments. The small Van de Graaff has been used mainly for Rutherford backscattering experiments related to analytical work.

The neutron programme has concentrated mainly on scattering and capture of structural materials and on fission cross sections, the work on the fission of ^{241}Am being particularly interesting because of the first application of a new concept for the suppression of α pile-up, and that on ^{230}Th and ^{232}Th because of the stimulation it provides to the theory of fission. Another new development has been the direct detection of α particles in the measurement of (n,α) cross sections of structural materials for fast neutrons.

The radio-nuclide programme has been distinguished by the completion of work on the half life of ^{239}Pu (rounding off several years' work, and surely providing the final confirmation of the marked change in its accepted value), and on the decay schemes of ^{134}Cs and ^{137}Cs. Work on the decay of $^{93}Nb^{m}$, of importance for long-term neutron dosimetry, has shown an interesting discrepancy with the previous most precise measurements.

Developments in the application of the accelerators to chemical analysis have continued. They have been applied to the analysis of environmental samples of air particulate matter and will also be used in the analysis of solutions of $MnSO_4$ baths (used in an international comparison of neutron source strengths) which started this year.

A good start has been made in the programme to provide certified actinide reference materials for chemical analysis (U and Pu, metal and oxides) and for non-destructive assay of ^{235}U enrichment by gamma ray counting.

Important activities in support of safeguards have been the provision of a ^{244}Pu spike solution and of the first solid spike (mixed $^{235}U/^{244}Pu$) and its use for measurements on undiluted input samples of reprocessing plants. The inter-laboratory comparison of α-spectrometry AS-76, related to the determination of ^{238}Pu in reprocessed material, has been completed.

The development of a new method of preparing very thin polyimide substrates for the deposition of nuclear material promises to be extremely valuable, as will other new developments

for the deposition of such material by spray-painting and by vacuum evaporation. These developments have been accompanied by the production of a large series of reference fission foils of ^{235}U, ^{239}Pu and ^{240}Pu.

It is important to recall that a large fraction of our work is done either for or in active collaboration with laboratories outside the JRC, in the EC, USA and elsewhere and with international organizations like IAEA and NEA. Although they have not been listed in detail, it is clear that they are one of the main raisons d'êtres of a laboratory like ours and provide a great stimulus to our work, some of the highlights of which have been summarized in this brief introduction.

B. ROSE
Head, Research Division

W. MÜLLER
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N U C L E A R D A T A

1. FAST NEUTRON DATA

Cross Sections for the Reactions $^{54}Fe(n,\alpha)^{51}Cr$ and $^{54}Fe(n,p)^{54}Mn$

A. Paulsen, R. Widera, F. Arnotte, H. Liskien

Cross sections for the reaction $^{54}Fe(n,\alpha)$ are of interest in the context of neutron-induced embrittlement of fast reactor structural materials. The half-life of 27.75d and the emitted gamma-radiation of 320 keV in 9.815 % of the decaying ^{51}Cr product nuclei allowed the application of the activation technique. Measurements and data evaluation have been finished and a paper submitted to Nuclear Science and Engineering for publication which contains the following abstract :

"Cross sections for the reactions $^{54}Fe(n,\alpha)^{51}Cr$ and $^{54}Fe(n,p)^{54}Mn$ were measured by the activation technique in the 6 to 10 MeV energy range using quasi-monoenergetic neutrons produced by the 7 MV electrostatic accelerator of the CBNM and the D(d,n) source reaction. In addition cross sections for the reaction $^{54}Fe(n,\alpha)^{51}Cr$ were obtained between 12 and 17 MeV using the T(d,n) source reaction. The cross sections were determined relative to the $^{56}Fe(n,p)^{56}Mn$ cross section with estimated overall uncertainties of about $\pm$ 7 %."

Results are presented in Fig. 1.1. together with earlier data.

Measurement of Ni(n,α) Cross Sections

A. Paulsen, H. Liskien, F. Arnotte, R. Widera

A reaction chamber is used containing five charged particle detector telescopes at 14, 52, 79, 109 and 140° relative to the 0° neutron direction. Each detector telescope originally consisted of one scintillation detector and two proportional counters acting as dE/dx counters, with all three detectors linked to a triple coincidence circuit. The differential (n,α) cross sections are measured relative to the differential n-p scattering cross section as observed by the 14° detector in a separate run.

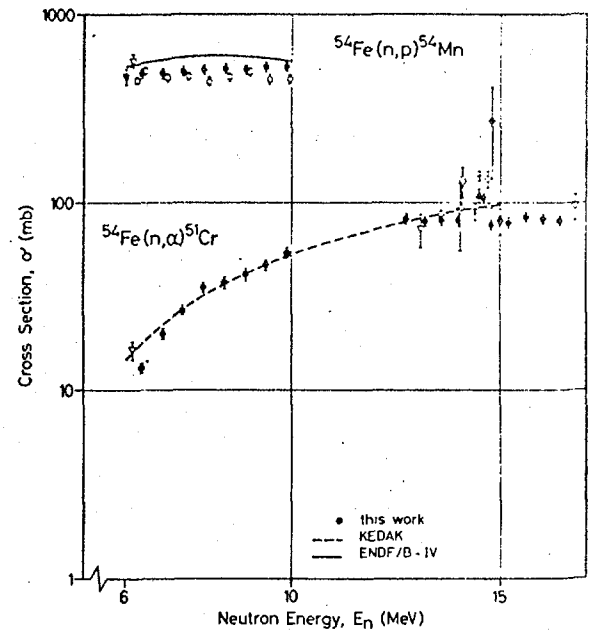

Figure 1.1

Some improvements have been realized concerning the reaction chamber. The neutron collimator was experimentally optimized by using brass instead of lead and by minimizing the volume. Simultaneously the collimator has become a fixed part of the chamber. The scintillation detectors have been replaced by silicon surface barrier detectors.

A bi-parametric data acquisition system of sufficient capacity became available in November 78.

Definite Ni(n,α) measurements were immediately started in steps of 0.5 MeV between 5 and 10 MeV neutron energy. These measurements and the associated data evaluation should be finished soon. The results are in rather good agreement with the KEDAK data file.

$^{103}Rh(n,n')^{103}Rh^{m}$ excitation function in the 0.2 to 4.1 MeV neutron energy range

A. Paulsen, R. Widera, F. Arnotte, H. Liskien

Our data so far accumulated for this reaction have been evaluated for inclusion in a general data evaluation * for the IAEA Advisory Group Meeting on Nuclear Data for Reactor Dosimetry. Corrections were calculated for neutron scattering in the backing of the neutron producing target and within the Rh samples. Further calculations determined the relative shape of the irradiation neutron spectrum and the average neutron energy for each sample, taking into account the shape of the measured $^{103}Rh(n,n')^{103}Rh$ excitation function. The results are shown in Table 1.1. and are still preliminary; they will be extended to higher neutron energies and they will be normalized to an absolute cross section measurement.

Cross Section Measurement for the Reaction $^{115}In(n,n')^{115}In^{m}$

H. Liskien, F. Arnotte, R. Widera, A. Paulsen

The data have been analysed and a paper written, which meanwhile appeared in Nuclear Science and Engineering 67 (1978) 334, with the following abstract :

"The excitation function for the reaction $^{115}In(n,n')^{115}In^{m}$ was measured by the activation technique from threshold up to 4.1 MeV in steps of 0.1 MeV. The absolute normalization of this excitation function has been performed at 2.1 MeV and is based on n-p scattering. The total uncertainty of the results is typically ± 6 % above 1 MeV. The results are in good agreement with recently published data".

In Fig. 1.2. our final data are presented and compared with other recent results.

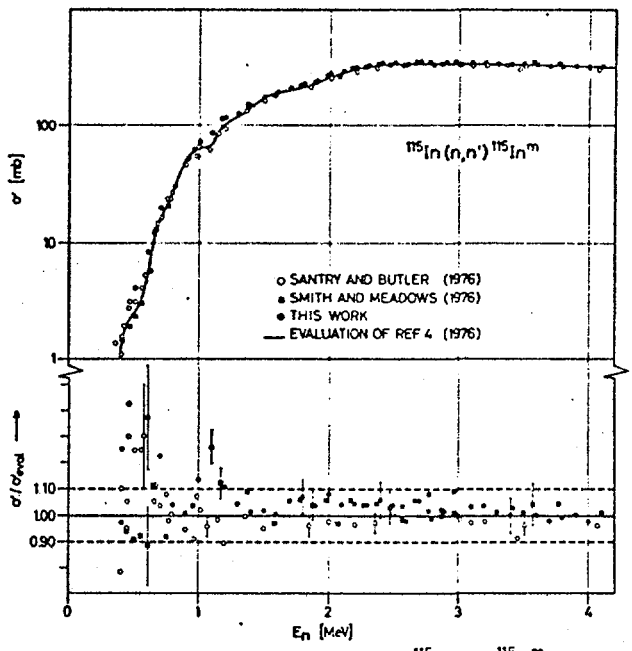

Figure 1.2 Cross sections for the reaction $^{115}In(n,n')^{115}In^{m}$ resulting from this work together with recent results from Chalk River (2) and Argonne (3). The degree of consistency may be judged in the lower part, where ratios against a recent evaluation (4) are plotted.

High Energy Resolution Measurement and Channel Analysis of the ^{230}Th Fission Cross Section

J. Blons **, C. Mazur **, D. Paya **, M. Ribrag **, J. Wartena, H. Weigmann

The $^{230}Th(n,f)$ cross section has been measured up to 5 MeV neutron energy using GELINA as a neutron time of flight spectrometer with a nominal resolution of 84 psec/m. At 720 keV,

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TABLE 1.1.

Relative excitation function for the reaction $^{103}Rh(n,n')^{103}Rh^{m}$ normalized to 1000 mb at 3.00 MeV. δE_n is the uncertainty of the average neutron energy E_n. The given total width $\pm \Delta E_n$ covers the full range of the neutron energy spectrum seen by the sample.

E_n(MeV)	δE_n(MeV)	ΔE_n(MeV)	σ(mb)	$\delta\sigma$(%)
0,199	± 0,003	+ 0,023 - 0,024	62,1	6,7
0,250	± 0,003	+ 0,024 - 0,028	82,7	5,9
0,300	± 0,004	+ 0,025 - 0,024	103,3	5.5
0.350	± 0.003	+ 0.021 - 0.020	84.1	6.2
0.399	± 0.004	+ 0.029 - 0.025	118.1	5.0
0.500	± 0.005	+ 0.063 - 0.053	146	4.8
0.601	± 0.005	+ 0.042 - 0.037	230	3.7
0.703	± 0.005	+ 0.070 - 0.067	356	3.0
0.802	± 0.007	+ 0.058 - 0.049	508	3.0
0.903	± 0.008	+ 0.095 - 0.100	573	3.0
1.006	± 0.008	+ 0.114 - 0.097	593	3.0
1.10	± 0.01	± 0.11	557	3.4
1.20	± 0.01	± 0.11	610	3.4
1.30	± 0.01	± 0.11	599	3.1
1.40	± 0.01	± 0.14	702	3.0
1.50	± 0.01	± 0.09	705	3.1
1.60	± 0.01	± 0.08	694	3.0
1.70	± 0.01	± 0.17	778	3.0
1.80	± 0.01	± 0.18	790	3.0
1.90	± 0.01	± 0.12	774	3.2
2.00	± 0.01	± 0.10	816	3.1
2.10	± 0.01	± 0.18	838	3.0
2.20	± 0.01	± 0.10	856	3.0
2.30	± 0.01	± 0.14	886	3.2
2.40	± 0.01	± 0.17	896	2.9
2.50	± 0.01	± 0.16	868	3.0
2.60	± 0.01	± 0.15	961	2.9
2.70	± 0.01	± 0.13	942	2.9
2.80	± 0.01	± 0.17	919	2.9
2.90	± 0.01	± 0.18	1024	2.9
3.00	± 0.02	± 0.19	1000	3.5
3.10	± 0.02	± 0.20	1005	3.3
3.20	± 0.02	± 0.21	1017	3.2
3.30	± 0.02	± 0.21	967	3.5
3.40	± 0.02	± 0.22	1004	3.3
3.50	± 0.02	± 0.23	1014	3.3
3.60	± 0.02	± 0.24	988	3.2
3.70	± 0.03	± 0.25	997	3.3
3.80	± 0.03	± 0.26	1039	3.2
3.90	± 0.03	± 0.27	1010	3.1
4.00	± 0.03	± 0.28	1041	3.0
4.10	± 0.03	± 0.29	1058	3.0

the effective neutron energy resolution was 1.7 keV (FWHM). The fission fragment detector was a 6 cell gas scintillator filled with xenon at NTP and containing 699.3 mg of ^{230}Th. A separate cell was loaded with 452.4 mg of ^{237}Np in order to allow the normalization of the ^{230}Th relative to the ^{237}Np(n,f) cross section (ENDF/B-IV).

An analysis of the ^{230}Th(n,f) cross section has been made in the $0.68 \leq E_n \leq 2$ MeV range. In the neigbourhood of 720 keV, the cross section shows an isolated structure which must be interpreted as a pure vibrational resonance. If however, this resonance can be resolved into its J components, as shown in Fig. 1.3, then a simultaneous interpretation of this fission cross section and the associated angular distribution of the fission fragments [1,2] can be made.

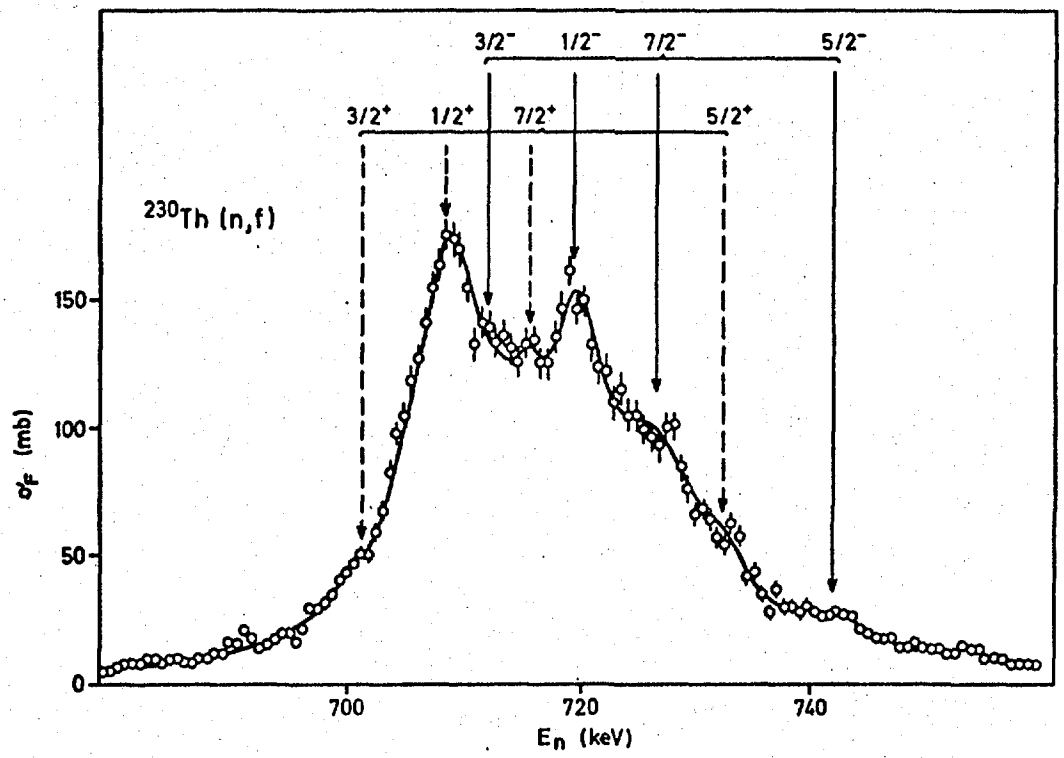

Figure 1.3

This vibrational resonance (k = 1/2) has indeed been resolved into eight components [3]. It was interpreted as beeing the result of an asymmetrically deformed third minimum with positive and negative rotational bands (see Fig. 1.3.).

The rotational parameters are $\frac{h^2}{2J} = 1.9 \pm 0.06$ keV, $a = -2.28 \pm 0.1$, the separation of positive and negative parity levels is constant $\Delta E^{\pm} = 10.8$ keV.

At the present stage, it seems reasonable to describe the fission cross section up to 2 MeV by successive opening of fission channels with K values of 1/2, 5/2, 7/2, 5/2, 1/2 and 5/2. The fission cross section has been calculated in a similar manner as for ^{232}Th [4]. This calculation gives the contribution of each spin value to the total fission cross section and is in good agreement with the experimental data.

The angular distributions are calculated for each energy where the angular distribution have been measured [1,2]. The agreement is fairly good although the spin assignment was originally made on the ground of the level spacings only.

It is worth noticing that similar results are found for the two thorium isotopes ^{230}Th and ^{232}Th : same moment of inertia, same decoupling parameter and same energy interval between positive and negative parity states.

1) JAMES G.D., LYNN J.E., EARWAKER L.G., Nucl. Phys. A189 (1972) 225.

2) YUEN G., RIZZO G.T., BEHKAMI A.N., HUIZENGA J.R., Nucl. Phys. A171 (1971) 614.

3) BLONS J., MAZUR C., PAYA D., RIBRAG M., WEIGMANN H., Phys. Rev. Lett. 41 (1978) 1289.

4) BLONS J., MAZUR C., PAYA D., RIBRAG M., WEIGMANN H., "Vibrational and rotational states in the $^{232}Th(n,f)$ cross section" to be presented at the International Symposium on Physics and Chemistry of Fission, Jülich, May 1979.

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Measurement of the Neutron Induced Fission Cross Section and of the Spontaneous Fission Half Life of ^{240}Pu

C. Budtz-Jørgensen, H.-H. Knitter

The fission cross section of ^{240}Pu was measured in a back to back geometry relative to the fission cross section of ^{235}U in the energy range from 140 keV to 9.75 MeV. The fission events were registered by fragment detection with the help of parallel plate ionization chambers. The fission layers of ^{235}U and ^{240}Pu have a diameter of 2.8 cm and a thickness of 200 $\mu g/cm^2$ respectively, and they are positioned at a distance of 6 cm from the neutron producing target and at zero degree with respect to the incident ion beam of the Van de Graaff accelerator. In order to avoid alpha pulse pile-up problems from the ^{240}Pu sample a special chamber described elsewhere in this report was used. The Van de Graaff was operated in pulsed mode and time-of-flight spectra were recorded at each incident neutron energy for the fission of ^{235}U and of ^{240}Pu respectively. A neutron time-of-flight detector was used to check the monoenergetic neutron beam. The time-of-flight spectra allowed to obtain the count rate ratio corrected for time-uncorrelated background and for the events from spontaneous fission. At energies below 1.10 MeV measurements were made using the $^{7}Li(p,n)^{7}Be$ reaction, employing LiF-targets of about 20 keV thickness. In this energy range experimental data were taken in steps of about 20 keV. In the neutron energy range from 1.0 MeV to 4.0 MeV the $T(p,n)^{3}He$ reaction was used to produce the neutrons. Between 1.0 MeV and 2.0 MeV and between 2.0 MeV and 4.0 MeV measurements were made in 50 keV and 100 keV intervals using TiT-targets of 0.4 and 2.0 mg/cm^2 respectively. Above 4.0 MeV the $D(d,n)^{3}He$ reaction was used in conjunction with a deuterium gas target. The energy spread of the neutrons varied with neutron energy from about 120 keV at 4 MeV to 45 keV at 9.75 MeV. Measurements were made with and without deuterium gas in the cell, in order to correct for fission events which were not induced by neutrons from the $D(d,n)^{3}He$ reaction. In this energy range from 4 MeV to 9.75 MeV experimental data were taken in incident neutron energy steps of 200 keV. The results of these measurements are shown in Fig. 1.4. The overall uncertainty of the data is 3 %. The agreement with data of Behrens et al., N.S.E. 66, 433 (1978), and Cierjacks et al., Conf. Neutron Physics and Nuclear Data, Harwell, Sept. (1978) p. 905, is good.

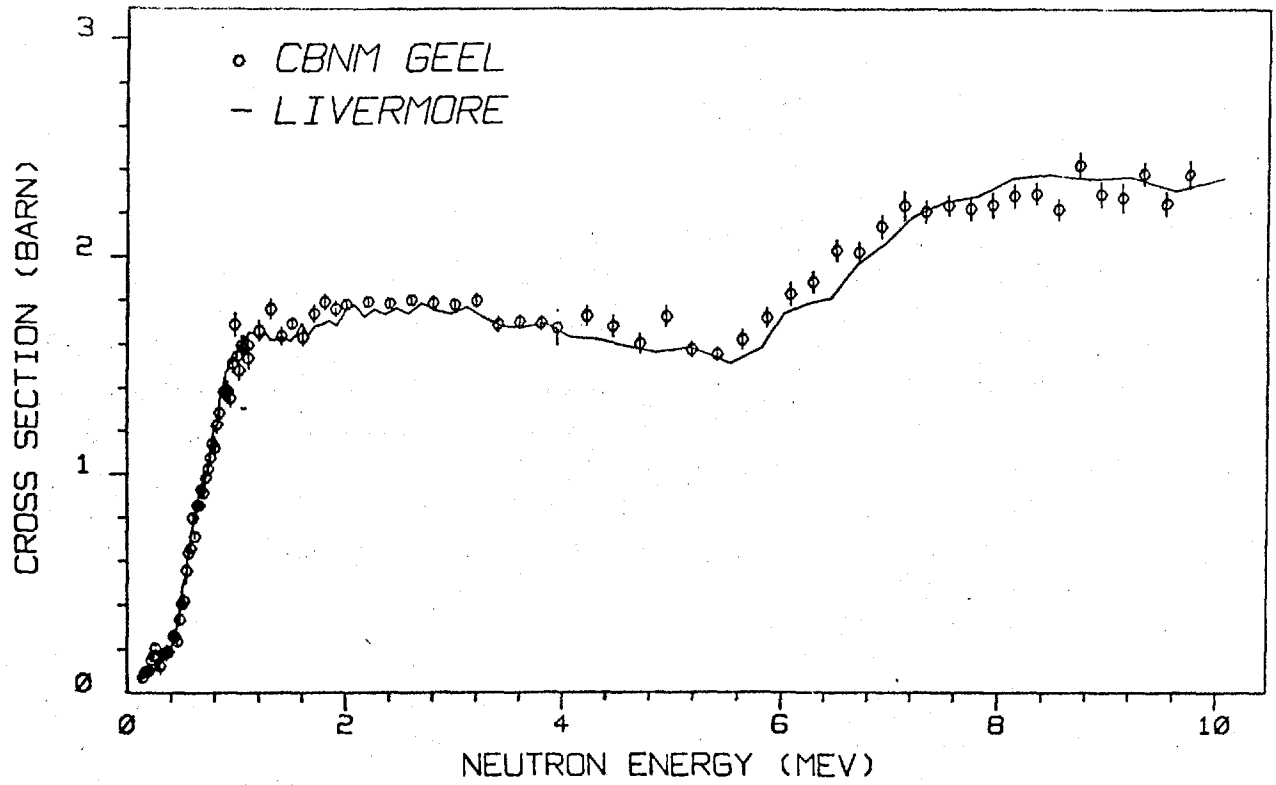

Figure 1.4

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Neutron Induced Fission Cross Sections of ^{241}Am from 100 eV to 5.3 MeV

H.-H. Knitter, C. Budtz-Jørgensen

The measurements of the neutron induced fission cross section of ^{241}Am were made using both the Van de Graaff and the Linac neutron facilities at CBNM. A neutron energy range from 1 eV to 5.3 MeV was covered. The fission cross sections were measured by fission fragment detection and relative to the fission cross section of ^{235}U. Two ^{241}Am samples, with diameters of 2.8 cm and masses of 0.8 mg and 4 mg respectively, were used above and below the fission threshold. These samples were mounted back to back with ^{235}U samples of 3 cm diameter and masses of 1.2 mg. The main experimental problem was to discriminate against the huge alpha background stemming from the Am samples. In the sub-threshold region the fission-to-alpha-rate ratio was 1 : 10^9. This problem was overcome by employing fission ionization chambers of a novel design developed at CBNM. The detectors allowed a reduction of the alpha pulse height by more than a factor 7 compared to a normal parallel plate chamber, whereby the alpha pulse pile up was reduced to have a negligible influence.

At the Van de Graaff 80 fission cross section data points were measured between 150 keV and 5.3 MeV. The reactions $^{7}Li(p,n)^{7}Be$, $T(p,n)^{3}He$ and $D(d,n)^{3}He$ were used to produce monoenergetic neutrons. The measurements at the Linac were made with à chamber positioned at a distance of 8 m from the neutron producing target. The Linac was operated with a pulse width of 4.4 ns and a repetition frequency of 800 Hz. The ^{235}U chamber part was again used as neutron flux detector. Below 2 keV where the ^{235}U fission cross section is dominated by resonances, the $^{6}Li(n,\alpha)T$ reaction, was used to determine the energy dependence of the neutron flux, which was measured with an ionization chamber loaded with a sample of ^{6}LiF. The time spectrum

obtained from the ^{241}Am half of the chamber reproduces, besides the ^{241}Am resonances, all the prominent resonances which are found in the fission cross section of ^{239}Pu. The relative amount of ^{239}Pu in the ^{241}Am sample was found to be (0.69 ± 0.10) % from the number of fission events in the ^{239}Pu 7.87 eV resonance. Within the statistics no other resonances from non-threshold fissile material could be observed. A detailed description of the measurements and the analysis of the experimental data will be given in Atomkernenergie ATKE 33 (1979).
Fig. 1.5. shows the obtained ^{241}Am fission cross sections compared with the data of other authors. There are only small discrepancies among the different data sets above threshold, although the present data are significantly 6 % lower than the preliminary data of Behrens and Browne, Report UCID-17324 (1976). Below the threshold the present data are in good agreement with the few points of Shpak et al., JETP Lett. 10, 175 (1969), whereas disagreement by more than one order of magnitude is found with the data of Seeger et al., Nucl. Phys. A96, 605(1967)
With respect to the data of Gayther and Thomas, Proc. of the Neutron Physics Conf. KIEV, Vol.3, page 3, a difference up to a factor of two is found in the energy range between 2 keV and 10 keV, whereas the two cross section results approach each other towards lower neutron energies. The data of Bowman et al., Phys. Rev. 137B, 326 (1965), not shown, are higher by almost a factor four than the present data in the range from 1 keV to 5 keV. The agreement becomes better towards lower energies.
Due to the relative high amount of ^{239}Pu in the ^{241}Am sample only a limited number of ^{241}Am resonances could be identified and their fission widths were obtained by area analysis. The derived fission widths are presented in Table 1.2. and are compared with those of Derrien and Lucas and those of Gaycher and Thomas. In the two latter experiments prompt fission neutrons were used for fission event detection.
The agreement between the different measurements is very satifactory remembering that the present work is based on fission fragment detection.

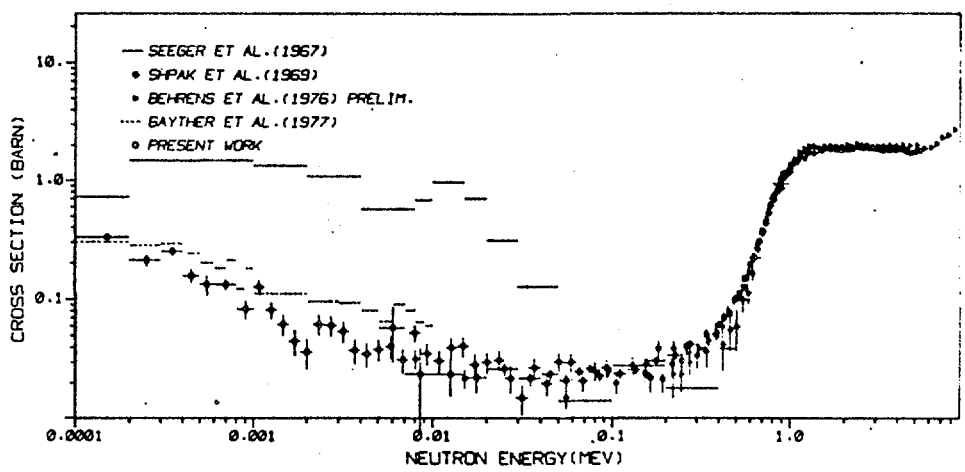

Figure 1.5

Table 1.2 : Comparison of ^{241}Am fission widths: Derien et al.[8], Gayther et al.[6] and the present authors.

Resonance Energy (eV)	Fission Width (meV)		
	Ref.[8]	Ref.[6]	This work
1.28	0.37	0.40	0.37 ± 0.02
1.93	0.08	0.06	---
2.37	0.18	0.16	0.19 ± 0.03
2.60	0.17	0.14	0.15 ± 0.03
3.97	0.16	0.13	0.13 ± 0.03
4.97	0.44	0.38	0.35 ± 0.05
5.42	0.63	0.55	0.64 ± 0.03
6.12	0.42	0.34	0.34 ± 0.11
6.74	0.22	0.08	---
7.66	0.10	--	---
8.17	0.12	0.19	---
9.12	0.18	0.17	0.17 ± 0.03
9.85	0.95	0.85	0.75 ± 0.08
14.68	0.27	—	0.29 ± 0.04

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The Original Neutron Spectrum to be Expected from a d-t Fusion Plasma

H. Liskien

The usual assumption of emission of monoenergetic particles (14.1 MeV neutrons, 3.5 MeV α-particles) from a d-t fusion plasma, is very rough. Using Monte Carlo methods the neutron- and α-particle spectrum to be expected from a plasma of temperature T = 10,20 and 30 keV have been calculated assuming a pure two-body reaction and isotropic velocity distribution for triton and deuteron. Reaction products from d-d and t-t reactions were not considered. Results are shown in Fig. 1.6. for the neutron spectrum obtained. Because every individual event was weighted with the relevant fusion cross section, the integral over each spectrum is proportional to the average fusion cross section at the given temperature. As may be seen, the usual assumption of emission of 'monoenergetic' neutrons is inaccurate. For inclusion of particles with at least 5 % of the maximum intensity one obtains for T = 30 keV an energy band of about 2 MeV width.

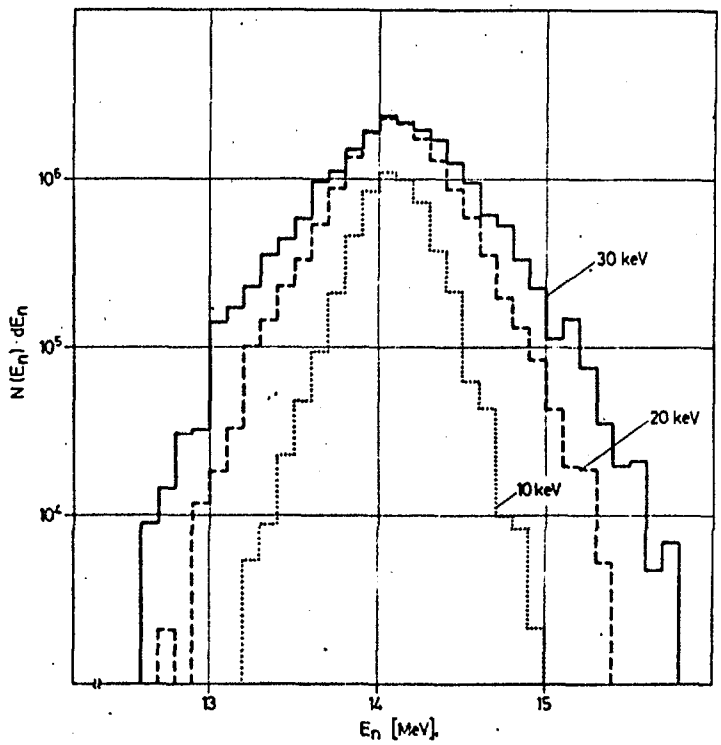

Figure 1.6

As a by-product one obtains also the distribution of the energies F_d (the deuteron laboratory energy with the triton at rest, which corresponds to the starting condition E_d, E_t and θ_t). If again the lower level of interest is set at 5 % of the maximum then - for T = 30 keV - the region of interest for the $T(d,n)^4He$ cross section is limited to $F_d < 250$ keV.

Contribution to IAEA Advisory Group Meeting on Nuclear Data for Fusion Reactor Technology

H. Liskien

On request of IAEA a summary on "Neutron Reaction Data in the MeV Range" has been presented at the above meeting held at VIENNA, Dec. 1979. The abstract reads as follows :
"The energy range and reaction types of relevance for neutron reaction data for fusion reactors are shortly discussed followed by a summary of the presently available methods to determine such data. Principal deficiencies are underlined and examples, typical for the data status are given."

2. RESONANCE NEUTRON DATA

Neutron Resonance Parameters of ^{91}Zr

A. Brusegan, F. Corvi, G. Rohr, C. Coceva *, P. Giacobbe *, M. Magnani *

The analysis of ^{91}Zr neutron resonance data carried out in collaboration with CNEN-Bologna was completed. Transmission spectra were shape analysed by means of a least-square computer program using single level formalism. Main characteristics of this analysis were : i) simultaneous fit of the transmission spectra of all sample thicknesses, ii) use of an asymmetrical resolution function whose detailed shape was calculated from a Monte Carlo simulation, iii) introduction of constraints to take into account *a priori* information on parameters to be fitted. Resonance parameters deduced from this analysis were $2g\Gamma_n$, and in some cases also spin and parity.

Taking into account such $2g\Gamma_n$ values, a modified version of the TACASI code was employed to deduce $2g\Gamma_\gamma$ from capture areas. Doppler, resolution and multiple scattering corrections were applied, the last one by means of a Monte Carlo routine. The resonance parameters of 77 resonances were determined up to 14.57 keV. In the range 14.57 to 19.80 keV, where only capture measurements were performed, $2g\Gamma_\gamma$ values for 20 additional resonances were given assuming the $2g\Gamma_n$ values of ref. [1]. Average parameters were calculated from this sample of resonance widths : their values are compared in Table 2.1. with those of Musgrove et al [1].

Table 2.1. : Average Resonance Parameters of ^{91}Zr in the range 0.15 keV

	Present Work	Musgrove [1]
$10^4 \times S_o$	0.49 ± 0.17	0.42 ± 0.12
$10^4 \times S_1$	7.2 ± 1.6	5.7 ± 1.0
$\langle \Gamma_\gamma \rangle_{l=0}$	138 ± 10 meV	140 ± 8 meV
S.D.of $\Gamma_\gamma(l=0)$	12 meV	30 meV
$\langle 2g\Gamma_\gamma \rangle_{l=1}$	250 meV	220 meV
S.D.of $2g\Gamma_\gamma(l=1)$	104 meV	110 meV
Potential scattering radius a = (7.5 ± 0.4) fm		

The points to be noted are :

i) the capture widths of p-waves are widely scattered around an average value which is almost twice that for s-waves. This is expected because of the low level-density of the compound nucleus, and the fact that the parity of the excited states up to 3 MeV is predominantly positive.

ii) While the agreement on Γ_γ values is excellent, our neutron widths are systematically larger than those of ref. [1]. The relative discrepancy tends to become more important for small Γ_n values.

Finally, by comparing the transmission measurements of the enriched oxide and of the natural metallic samples, it was possible to deduce a value for the effective temperature of zirconium oxide. It was found T_{eff} = (317 ± 13)°K at 20°C as compared with a calculated T_{eff} of 302°K for the metal at the same temperature.

1) A.R. de L. MUSGROVE et al., Aust. J. Phys. 30 (1977) 391.

* CNEN, Bologna, Italy

Resonance Parameters of ^{93}Nb

J. Winter, E. Cornelis *, L. Mewissen **, F. Poortmans **, G. Rohr, R. Shelley, T.van der Veen

We have performed a complete set of neutron time-of-flight experiments on ^{93}Nb in the resolved energy range including total, capture and elastic scattering cross section experiments, self-indication ratio measurements and low energy capture gamma-ray spectrum measurements. A special effort was made to perform spin and parity assignments of the neutron resonances.
The transmission experiments were performed on samples cooled at liquid nitrogen temperature.
The scattering cross section was measured using lead as a standard scattering sample.
The capture cross section was measured using two C_6F_6 liquid scintillators for the capture gamma-ray detector system. Time-of-flight and pulse-height were recorded simultaneously on magnetic tape and a weighting was applied to the measured pulse-height to achieve a detector response proportional to the total energy released in the capture process.
The energy spectrum of the neutrons at the detector station was measured using the same C_6F_6 detector system but by replacing the Nb sample with a ^{10}B slab. The absolute calibration of the capture experiment was based on the knowledge of the neutron widths of several weak resonances below 400 eV obtained from our transmission experiments. Indeed, for these cases the neutron width Γ_n is directly obtained from the area analysis of the capture data. The systematic error due to this normalisation is assumed to be less than 5 %.
Initially, the transmission data were analysed using a modified version of the Atta-Harvey area analysis programme. Afterwards, we have repeated the analysis using the multi-level Breit-Wigner code SIOB. The agreement between the results of both analysis procedures is very good for well-isolated and weak resonances but in cases where strong resonances are near to one another, the multi-level analysis yields a much better fit to the data and the results obtained for the neutron widths are sometimes as much as 20 % different from the results of the Atta-Harvey area analysis. For this reason we have used the results from the multi-level Breit-Wigner shape fitting.
The scattering data were corrected for multiple interaction effects using a Monte Carlo computer code. An area analysis of the corrected data yields Γ_n as a function of Γ_γ.
The neutron widths for resonances up to 7 keV were obtained from the analysis of the transmission data and, for very weak resonances, from the area analysis of the capture data. The total capture widths were deduced from the combination of the results of transmission, scattering, capture and self-indication ratio measurements. The results have yielded the following average properties of the resonance parameters in the energy range up to 7 keV :

s-wave strength function :

$$S_o = \frac{\Sigma\, g\, \Gamma_n^{\,o}}{\Delta E} = (0.40 \pm 0.06) \times 10^{-4}$$

p-wave strength function :

$$S_1 = \frac{\Sigma\, g\, \Gamma_n^{\,l}}{(2\ell+1)\, \Delta E} = (5.1 \pm 1.0) \times 10^{-4}$$

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mean capture widths :

$(\overline{\Gamma}_\gamma)_{\ell=0} = 145 \pm 10$ meV $\qquad (\overline{\Gamma}_\gamma)_{\ell=1} = 168 \pm 14$ meV

The results for the strength functions are in good agreement with those from previous work.

Resonance Parameters of Pd Isotopes

P. Staveloz *, E. Cornelis **, L. Mewissen ***, F. Poortmans ***, G. Rohr, R. Shelley, T. van der Veen

We have started a systematic study of resonance parameters of the stable Pd isotopes (enriched isotopes on loan from Oak Ridge National Laboratory). Up to now, the following experiments have been performed at the 150 MeV Linac of CBNM (GELINA):

^{105}Pd : $\sigma_{n,n}$(10 eV - 2.05 keV)
$\sigma_{n,\gamma}$(10 eV - 2.05 keV)
$\sigma_{n,T}$(10 eV - 2.05 keV)
The analysis of the transmission and capture data have been completed; the analysis of the scattering data is in progress.

^{106}Pd : $\sigma_{n,T}$(45 eV - 30 keV)
The analysis has been started.

^{108}Pd : $\sigma_{n,n}$, $\sigma_{n,\gamma}$ and $\sigma_{n,T}$(30 eV - 5 keV)
The analysis of all data has been completed.

^{110}Pd : $\sigma_{n,T}$ experiments in the energy range 45 eV - 30 keV have been started.

Capture cross section experiments on $^{104, 106, 110}$Pd and transmission experiments on ^{104}Pd are in preparation.

From the analysis of the data on ^{105}Pd and ^{108}Pd we have obtained the following results :

^{105}Pd :
1) mean s-wave level spacing : $D_o = 10.6 \pm 0.4$ eV
2) s-wave strength function $S_o = 0.59 \,^{+\,0.08}_{-\,0.07} \times 10^{-4}$
3) the capture widths below 200 eV were obtained from a shape analysis of the transmission data yielding a mean value : $\langle \Gamma_\gamma \rangle = 166.7 \pm 1.6$ meV (stat. error). The capture widths of resonances above 200 eV will be deduced later after completion of the analysis of the scattering data.

^{108}Pd :
1) s-wave strength function $S_o = 0.78 \,^{+\,0.19}_{-\,0.15} \times 10^{-4}$
2) The capture widths were obtained from an area analysis of scattering, capture and transmission data, yielding a mean value : $\langle \Gamma_\gamma \rangle = 76.8$ meV + 2.8 meV (stat.) $\pm$ 4 meV (syst.error)

These results were presented at the "International Conference on Neutron Physics and Nuclear Data for Reactors and other Applied Purposes", Harwell, Sept. 1978.

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p-Wave Assignment of ^{232}Th Neutron Resonances

F. Corvi, G. Pasquariello ★, T. van der Veen

The p-wave assigment technique already used for ^{238}U neutron resonances [1] was also applied to the ^{232}Th case. The preliminary measurement performed with the old linac at a 50 m station was repeated at a 30 m flight path in order to improve the statistics and the signal to background ratio. The yield of high-energy capture γ-rays above 4.4 MeV from a 6 mm thick metallic Th sample was measured in the neutron energy range 20 - 2200 eV. This run was then compared to a normalisation run in which γ-rays in the range 3.7 to 4.4 MeV were dedected : such a high threshold was chosen in order to limit the γ-activity from the decay of the daughter product ^{208}Tl. A total of 58 resonances showing an enhancement of the high energy γ-ray yield were assigned as p-waves.

The integral distribution of the reduced neutron widths $g\Gamma_n^l$ for the assigned p-wave resonances is plotted in Fig. 2.1. together with the fitted Porter-Thomas function (continuous curve). The best estimate of the p-wave strength function derived from such a fit is :

$$S_l = (2.0 \,^{+\,0.5}_{-\,0.4}) \times 10^{-4} \qquad \text{for } R = 8.30 \text{ fm}$$

1) F. CORVI, G. ROHR, H. WEIGMANN, Proc. Conf. Nuclear Cross Sections and Technology, Washington, D.C., NBS SP 425 (1975) 733.

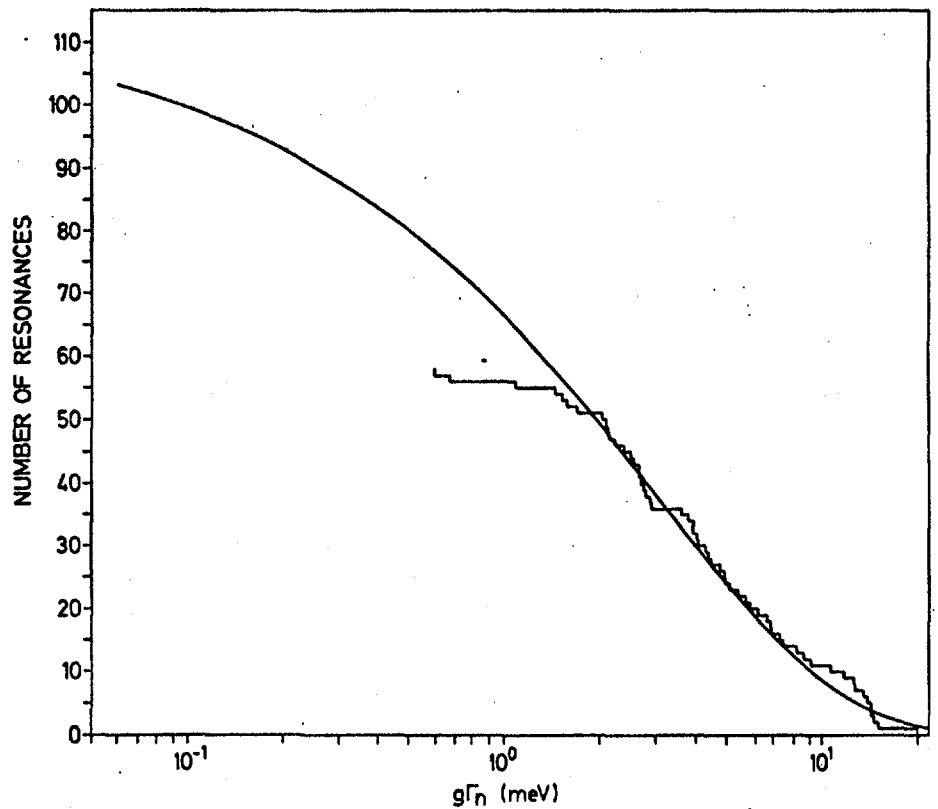

Figure 2.1 Plot of the integral distribution of $g\Gamma_n^l$ (histogram) and of the Porter-Thomas fit (continuous curve).

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Measurement of ^{231}Pa Neutron Induced Fission Cross Section

C. Budtz-Jørgensen, H.-H. Knitter

A fission ionisation chamber was loaded with a sample of 360 $\mu g/cm^2$ thickness and with a diameter of 3.2 cm. The alpha activity of this sample was about 5×10^6 s^{-1}. The detector was tested at the Van de Graaff accelerator and electronic adjustments were made. The chamber was then mounted at a 8 m station at the linac and a measurement in the neutron energy range from 50 keV to 2 MeV with a resolution of 0.5 ns/m was started. However, the time-of-flight spectra show structure from electronic pick-up in the first microsecond after the linac burst and "common mode rejection" is being tried to get rid of these pick-up signals.

Neutron Resonance Parameters of ^{237}Np

L. Mewissen *, F. Poortmans *, E. Cornelis **, G. Van Praet **, A. Angeletti ***, G. Rohr, H. Weigmann

The resonance analysis of capture, elastic scattering and total cross section data on ^{237}Np has been completed. The neutron widths Γ_n were obtained for 213 resonances and the radioactive widths Γ_γ for 25 resonances. The mean capture width $\langle \Gamma_\gamma \rangle = 41.2 \pm 2.9$ meV and the mean s-wave resonance spacing $D_o = 0.740 \pm 0.061$ eV. The s-wave strength-function was found to be $S_o = (1.2 \pm 0.14) \times 10^{-4}$.
A final report on the work on ^{237}Np has been written and accepted for publication in Nuclear Science and Engineering.

On the Number of Prompt Neutrons emitted in ^{239}Pu Fission

H. Weigmann

The liquid scintillator detector used for measurements of the fission cross section of ^{239}Pu by fission neutron detection, is subdivided into four individual detector cells. Thus, it is possible to measure the relative numbers of two-fold and three-fold coincidences due to the simultaneous detection of two or three prompt neutrons, respectively. Their ratio depends on the average number of prompt neutrons emitted per fission event, $\bar{\nu}$, and on the distribution of individual ν-values around $\bar{\nu}$.
The ratio, R, of three-fold to two-fold coincidences has been measured per individual resonances in the neutron energy range from 12 eV to 460 eV. It is observed that :

1. R-values of very weak fission resonances ($\Gamma_f < 10$ meV) are systematically lower than average;
2. excluding weak fission resonances ($\Gamma_f > 20$ meV), the average R-values for resonances of spin 0^+ and 1^+ are the same to within 1 %.

Representing, as usual, the distribution of ν by a Gaussian distribution with average $\bar{\nu} = 2.9$ and $\sigma = 1.08$, the sensitivity of the observable R to changes in $\bar{\nu}$ and σ is obtained from a Monte Carlo calculation. With this information the following conclusions may be drawn from the data :

1. If σ is assumed constant, the above-mentioned dependence of R on Γ_f for weak fission resonances implies a corresponding dependence of $\bar{\nu}$ on Γ_f which is found to agree with

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the results of Shackleton et al. [1] obtained by a different method. For the stronger fission resonances ($\Gamma_f > 20$ meV), an upper limit for the spin-dependence of $\bar{\nu}$ is obtained as $|\ \bar{\nu}\ (0^+) - \bar{\nu}\ (1^+)\ | < 0.03$ which again is in agreement with, but less accurate than, the corresponding result of Shackleton [2].

2. Adopting the value of $\bar{\nu}\ (0^+) - \bar{\nu}\ (1^+) = 0.013 \pm 0.0055$ as given by Shackleton, the above limit for the difference in R-values for the two spin-families implies that $|\ \sigma\ (0^+) - \sigma\ (1^+)\ | < 0.02$.

1) D. SHACKLETON, J. TROCHEN, J. FREHAUT, M. LE BASS, Phys. Lett. 42B (1972) 344.

2) D. SHACKLETON, D. Sc. Thesis, Paris 1974.

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Measurement of the Fission Cross Section of ^{239}Pu

R. Barthélémy, C. Wagemans *, J. Wartena, H. Weigmann

Measurements of the ^{239}Pu fission cross section in the energy range from 5 eV to 30 keV have been made using two methods :

1. Surface barrier detectors were used to measure fission fragments and α-particles from back-to-back foils of ^{239}Pu and ^{10}B, respectively, where the latter is used for determination of the n-flux;
2. The shape of the cross section has been measured with comparatively higher resolution and statistical precision with a liquid scintillator tank for detection of the prompt fission neutrons.

The analysis of the data is in progress.

Kinetic Energy- and Mass-distribution for the Fissioning System ^{240}Pu

C. Wagemans *, G. Wegener-Penning ** H. Weigmann, R. Barthélémy

During the annual shut down of GELINA, the kinetic energy- and mass-distribution of the fission fragments emitted during the spontaneous fission of ^{240}Pu were measured. The resolution of these measurements was better than in the case of Deruytter and Wegener-Penning (Conf. on Phys. and Chem. of Fission, IAEA, Rochester 1973, p. 51). Comparison of the present results with our previously obtained ^{239}Pu(n_{th},f) data indicate that the mean total kinetic energy $\bar{E}_K$ is roughly 1 MeV smaller for spontaneous fission than for thermal neutron induced fission, which confirms the conclusion of Deruytter and Wegener-Penning.
Furthermore, a new series of measurements of the kinetic energy- and mass-distributions for the resonance neutron induced fission of ^{239}Pu was started recently after the receipt of a new ^{239}Pu sample.

Neutron Induced Fission Cross Sections of ^{241}Am from 100 eV to 5.3 MeV

H.-H. Knitter, C. Budtz-Jørgensen

See chapter 1 "Fast Neutron Data" page 10.

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A New Interpretation of the Level Density Systematics

G. Rohr, R. Shelley

The intermediate- or weak- coupling compound model has been successfully used to explain the fluctuation of the strength function for adjacent nuclei by means of the doorway density calculated at neutron separation energy. Based on this model we have started to study the level densities of states of specified hierarchy corresponding to intermediate steps of the compound process in order to check their influence on the level density systematics. This information would provide important insights into the mechanism involved in generating a compound nuclear state.

In doing this we have calculated the level density of doorway states at neutron separation energy for more than 200 nuclei spread over the whole atomic weight range. The density of these states has been calculated in the framework of the single particle shell model using Nilson single particle states (without deformation). The pairing force has been taken into account according to the quasi-particle formalism. Finally the level density has been determined in a straightforward way using the combinatorial method.

The results for the doorway level density and the experimentally determined level density of the compound states (newly evaluated up to A = 60) have been reduced by means of the Fermi gas level density expression to the level density parameters a_D and a_C respectively. The level density parameter a_D can be described by a linear function without structure at closed shell nuclei. The compound state level density parameter a_C as a function of A seems to have the same slope as a_D except where it is interrupted at closed shell nuclei (decrease of a_C) and by steps (increase of a_C). The steps can be explained by an increase of the degree of freedom for the excitation process by two additional quasi particles. Based on this model the following aspects are being studied :

- prediction of compound level densities
- energy dependence of level densities
- possibility of determining the effective excitation energy at neutron separation energy.

Finally, as an example, the intermediate structure observed in ^{56}Fe and ^{206}Pb will be interpreted with this model.

Statistical Model Calculation of the Total Radiative Width and the Degree of Freedom in the Capture Process

G. Rohr

The total neutron radiative width and the degree of freedom in the capture process have been calculated with the statistical model, using as far as possible experimental data obtained mainly from γ-ray spectra measurements. Comparing these results with data obtained in total capture cross section measurements, laws for the level density and in particular for gamma-ray strength functions (E1 and M1) have been studied. The method has the advantage of providing complete gamma-ray strength functions down to low transition energies. Results for ^{56}Fe, ^{98}Mo and ^{238}U were presented at the 3rd Int. Symp. on Neutron Capture Gamma Ray Spectroscopy and Related Topics at Brookhaven.

High Resolution Cross Section Measurement of Structural Materials

A. Brusegan, F. Corvi, G. Rohr, R. Shelley, T. van der Veen, C. Jungmann *, L. Mewissen **, F. Poortmans **

Transmission measurements have been performed for the separated isotope ^{54}Fe at a 200 m flight path, using a plastic scintillator NE 110 (15 x 15 x 2.5 cm) viewed side-on by four photomultipliers (RCA 4516). The experiment was done at GELINA with 4 n sec burst and channel widths in the neutron energy range from 20 keV up to 2.5 MeV. The analysis of the data has been started.

Capture cross section measurements for the separated isotopes ^{54}Fe and ^{56}Fe have been performed using a pair of cylindrical deuterated liquid scintillators (C_6D_6) each with a diameter of 4 inches and a length of 3 inches, the faces of which were in optical contact with EMI photomultipliers (2823 QKB). The measurements were done at a flight path length of 58 m and a burst width of 4 n sec in the energy range 500 eV - 600 keV. A weighing over 256 amplitude channels was applied to achieve a detector response proportional to the total energy released in the capture process. The neutron flux was measured both with a 0.5 mm thick Li-glass detector as well as with the same capture detector system with the capture sample replaced by a sintered 3 mm thick B_4C slab.

Absolute calibration was obtained by the black resonance technique using resonances in Ag below 72 eV, and will be compared with a calibration based on the 1.15 keV resonance of ^{56}Fe. The analysis has been started but is delayed due to differences which have been obtained in these two calibration procedures.

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3. STANDARD NEUTRON CROSS SECTION DATA

Interlaboratory Comparison of Neutron Fluence Measurements

H. Liskien, A. Paulsen

A comparison of fast neutron fluence measurement between "Institut für Radiumforschung und Kernphysik" at Vienna and CBNM was carried out at 13.7 MeV neutron energy. An aluminium sample was irradiated at Geel with a fluence determined by proton recoil counting, and the induced activity was determined the day after at Vienna. The ratio of H(n,n) to Al(n,α) based neutron fluence is 0.973 $\pm$ 0.029. The comparison uses a high-accuracy Al(n,α) cross section determined earlier in Vienna by fluence determination employing associated particle counting.

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The $^{10}B(n,\alpha\gamma)^{7}Li$ Cross Section between 0.1 and 2.2 MeV

G. Viesti, H. Liskien

The measurements are described in an article which appeared in Ann. of Nucl. Energy 6 (1979) 13 with the following abstract :

"The shape of the excitation function for the standard neutron reaction $^{10}B(n,\alpha\gamma)^{7}Li$ in the energy range from 0.1 to 2.2 MeV has been determined relative to the angular distribution of the neutron source reaction $T(p,n)^{3}He$ at 1.6, 2.3, and 3.0 MeV proton energy. The 478 keV γ's produced in a 3 mm thick boron carbide sample were observed in a Ge(Li)-detector. The time-of-flight method was used to discriminate against events from neutrons of degraded energy". Results are shown in Fig. 3.1.

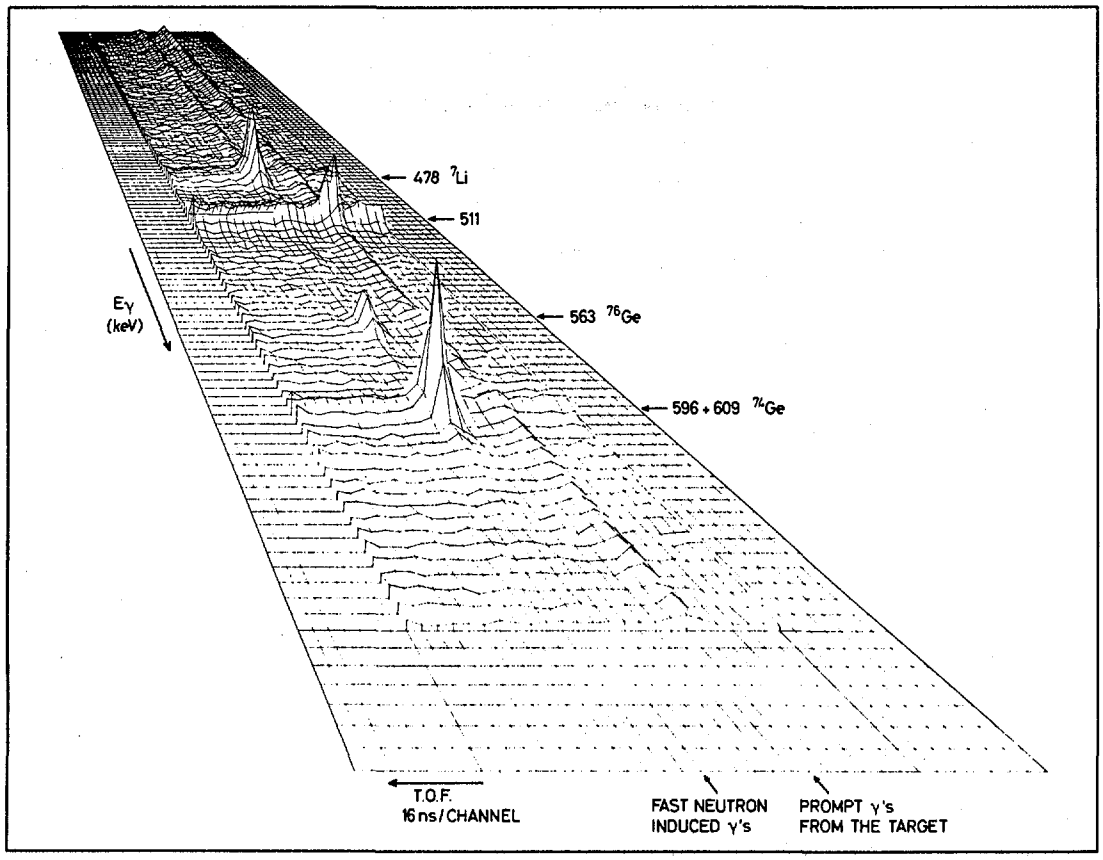

Figure 3.1 Three-dimensional perspective plot of obtained data. The shown spectrum was obtained at 713 keV neutron energy (2.3 MeV proton energy, 77.5° emission angle) in about six hours

Tentative Cross Section Measurements at GELINA below 10 meV

R. Barthélémy, C. Wagemans ★, E. Wattecamps

Experimental neutron cross section data below 25 meV suffer from statistical and systematic uncertainties. For g-factor calculations it is essential to know the shape of the cross section below 25 meV also since half of the integral stems from the energy range below 25 meV. The g-factor is defined by

$$g = \frac{\int_o^\infty \sigma(E).\sqrt{E}.M(E)dE}{\sigma_o.\sqrt{E_o}} \qquad \text{with}$$

$$M(E) = \frac{2}{\sqrt{\pi}} . \sqrt{E} . (kT)^{-3/2} . e^{-E/kT} \qquad \text{and}$$

$$\int_o^\infty M(E)dE = 1$$

and E_o the neutron energy corresponding to the velocity of 2200 m/s.
Preliminary measurements have been performed at GELINA with a flight path of 9.45 m to determine the capabilities and/or limitations for measuring below 10 meV. The detector is a large surface barrier semi-conductor of 20 cm^2 in the neutron beam, with a sample at 0.4 cm distance in front of it. Three samples have been used :

^{235}Uranium	:	150 μg UF_4 per cm^2,	35 mm diameter
^{239}Plutonium	:	51.4 μg PuF_3 per cm^2,	20 mm diameter
^{6}Lithium	:	176 μg LiF per cm^2,	80 mm diameter

The linear accelerator was operated at 25 pps with a pulse width of 2 μs and an electron energy of 100 MeV, thus producing 1.0 KW beam power on the neutron target, with the moderator geometry which is commonly used for fast neutron cross section measurements.
Measurements with uranium and lithium have been made also with Cd in the beam, thus allowing to some extent an estimation of the size of the background. The measurements have reasonable counting rates down to 1 meV with a signal to background ratio at 2 meV, as determined by Cd-transmission, of 60 to 1 and 5 to 1 for ^{235}U and LiF respectively.
A preliminary analysis gives a measured neutron energy spectrum which fits rather well with a Maxwellian of $\frac{kT}{2}$ = 35 meV.
See Fig. 3.2.
Investigations on systematic errors such as the reliability of the background determination are in progress.

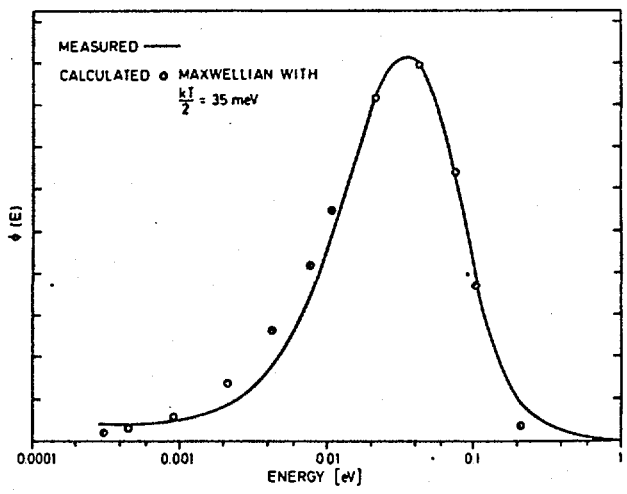

Figure 3.2 Measured and calculated thermal neutron spectra of GELINA.

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Accurate Measurement of the Fission Cross Section of ^{235}U

C. Wagemans *, R. Barthélémy

An additional measurement of the ^{235}U fission cross section was performed at an 8 m fligth-path, covering the neutron energy region from 30 keV down to thermal energy. Back-to-back foils of ^{235}U were used with surface barrier detectors.
The analysis of the data is in progress.

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4. NON-NEUTRON NUCLEAR AND ATOMIC DATA

Studies on the Decay of $^{93}Nb^{m}$

W. Bambynek, G. Grosse, W. Oldenhof, D. Reher, R. Vaninbroukx

The reaction $^{93}Nb(n,n')^{93}Nb^{m}$ is very useful for the determination of fast neutron fluences especially for radiation damage studies of reactor pressure vessels. However, the decay scheme parameters of $^{93}Nb^{m}$ have to be known more accurately. A detailed study of this decay is going on at CBNM using radioactive source material of various origins.

The study to get pure, carrier-free $^{93}Nb^{m}$ has been continued. At the National Accelerator Centre of the South African Council for Scientific and Industrial Research, Pretoria, a strip of Nb metal was bombarded with 16 MeV deuterons. The produced ^{93}Mo separated from the Nb will be used to obtain $^{93}Nb^{m}$ via the electron capture decay of ^{93}Mo. The material has arrived at CBNM and has been checked for γ-ray impurities with a pure Ge detector for energies below 250 keV and with a Ge(Li) detector for higher energies.

From a first examination the following radionuclides could be identified : ^{54}Mn, ^{56}Co, ^{57}Co, ^{58}Co, $^{92}Nb^{m}$, ^{94}Nb, ^{95}Nb, ^{181}W, and ^{182}Ta. These measurements are continuing in order to determine the intensities of the impurities.

Thin and homogeneous sources of low self absorption are essential for an accurate determination of the conversion electrons of $^{93}Nb^{m}$. Consequently, an intensive study has been started on the preparation of such sources by various techniques such as freeze drying of droplets, evaporation of $^{93}Nb^{m}$ fluoride or oxalate from a filament and ion bombardment of $^{93}Nb^{m}$ metal or its compounds. Freeze drying gave promising results and is used for sources from which the concentration of the radioactive material in the original solution has to be deduced.

For the preparation of sources by ion bombardment and by flash-evaporation from a filament an evaporation device has been installed. It consists mainly of a diffusion pump (1000 ls^{-1}), a base plate and a bell jar. In the flash-evaporation technique only a very small part of the filament is heated for a short time (a fraction of a second) to produce as little heat as possible and only as much as necessary to evaporate submicrogram quantities of the radioactive material on to VYNS foils of less than 20 $\mu g\ cm^{-2}$ thickness.

A schematic view is shown in Fig. 4.1.

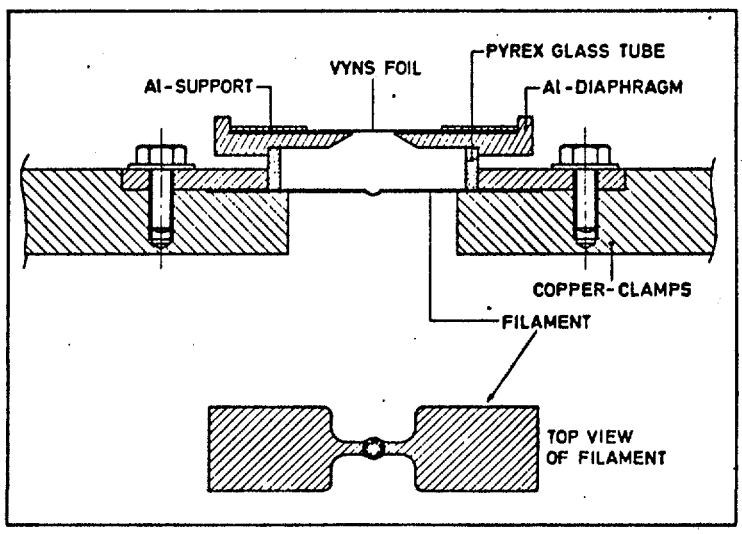

Figure 4.1 Source preparation by flash evaporation.

This technique makes it possible to evaporate Nb compounds. Evaporation of Nb metal by filament heating seems not to be possible due to its high melting point (2740 K). To overcome this problem a small ion gun producing a beam of about 1 to 2 mm diameter has been installed. A small piece of the Nb metal (about 5mg) is mounted on a graphite support and bombarded with Ar ions under an angle of about 45°. Due to the evaporation yield ratio of niobium and carbon the evaporation of niobium is favoured. The set-up is shown in Fig. 4.2.

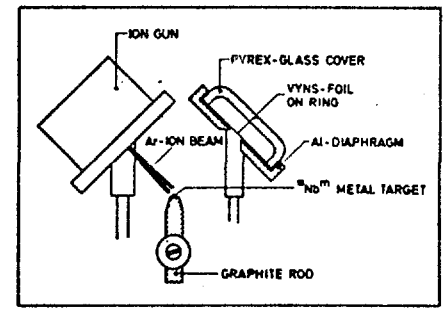

Figure 4.2 Source preparation with an ion gun.

Test evaporation and sputtering with inactive Nb and Nb compounds gave promising results. Thus, preparation of sources with radioactive material has been started.
The measurements for the determination of the half-life of $^{93}Nb^{m}$ have been continued using Si(Li) detectors. Four sources prepared from two different samples have been measured 10 times over a period of 1.5 year. During this period only 7 % of the $^{93}Nb^{m}$ decayed. Consequently the uncertainty is still rather high. The mean value of the preliminary result is $T_{1/2}$ = (14.3 ± 1.3)years which is definitely lower than the value of (16.4 ± 0.4)years published recently by R. Lloret [1].
The results achieved in the $^{93}Nb^{m}$ project have been presented at the "International Conference on Neutron Physics and Nuclear Data for Reactor and other Applied Purposes" at Harwell, September 1978.

1) R. LLORET, Mesure de la période de décroissance radioactive de $^{93}Nb^{m}$, Radiochem. Radioanal. Letters 29 (1977) 165.

Studies on the Decay of ^{141}Ce

H.H. Hansen, E. Celen, G. Grosse, D. Mouchel, A. Nylandsted-Larsen, R. Vaninbroukx

The results of this investigation were published with the following abstract :
"The decay of ^{141}Ce has been reinvestigated using different experimental methods. The efficiency extrapolation technique applied to $4\pi\beta$-γ coincidence measurements has been employed for the determination of the disintegration rate of the sources and of the total internal conversion coefficient. The K X-ray and γ-ray emission rates have been measured with a calibrated Si(Li) and an intrinsic germanium detector. Additionally the photon intensities have been determined from the internal conversion coefficients. Electron experiments with a magnetic β-spectrometer yielded the relative intensities, the shape factors and the maximum energies of the two β^--transitions and the internal conversion ratios K/L and K/(L+M+...). The K-shell internal conversion probability has been determined with an electron K X-ray coincidence technique using the magnetic β-spectrometer together with a high energy resolution Si(Li) detector. New values for the following decay properties have been deduced: β^--intensities(ρ_1=0.306±0.006, ρ_2=0.694±0.006), shape factors C=const(1+Aϵ) (A_1=-(0.24±0.03), A_2=-(0.22±0.04)), maximum β^--energies(E_{01}=(582.2±2.6)keV, E_{02}=(436.7±4.6)keV), K-shell internal conversion coefficient (α_K=0.376±0.008), total internal conversion coefficient (α=0.438±0.010), internal conversion ratios (K/L=7.29±0.24, K/(L+M+...)=5.78±0.18), γ-ray intensity (I_γ=0.482±0.003), K X-ray intensity (I_{KX}/I_γ=0.349±0.005). The procedure of uncertainty quotation is presented in some detail. The results are discussed with respect to existing experimental data".

Studies of the Decay of ^{239}Pu

R. Vaninbroukx, B. Denecke, M. Gallet, G. Grosse, F. Hendrickx, W. Zehner

The determination of the half-life has been finished. The α-emission rates of samples of a Pu material containing originally 99.98 atom % ^{239}Pu but spiked with ^{242}Pu for the determination of the Pu content by mass-spectometric isotope dilution techniques, were determined by liquid scintillation counting. Corrections for the contribution of ^{238}Pu, ^{240}Pu, and the spike material ^{242}Pu to the count rates were applied and the specific α-emission rate and half-life of ^{239}Pu were deduced.

The results of these measurements and those deduced from counting α particles in a defined solid angle of low geometry, which were reported earlier [1], are given in Table 4.1. The uncertainties quoted correspond to a 1 σ level taking into account random and systematic effects.

Table 4.1 : Specific α-emission Rate and Half-life of ^{239}Pu

Method	Specific α-emission $s^{-1}/\mu g$ ^{239}Pu	Half-life years
Low geometry	2298 ± 3	$(2.4085 \pm 0.0030)10^4$
Liquid Scintillation	2295 ± 3	$(2.4114 \pm 0.0030)10^4$
Mean	2296 ± 3	$(2.4100 \pm 0.0030)10^4$

The mean value of $(2.410 \pm 0.003)10^4$ years is in close agreement with the value of $(2.411 \pm 0.003)10^4$ years recommended at the "First Coordinated Research Meeting on the Measurement of Transactinium Isotope Nuclear Data", I.A.E.A., Vienna, 20-21 April 1978 [2].

1) Central Bureau for Nuclear Measurements, E.D. WATTECAMPS (ed.), Geel, Belgium, Nuclear Data Progress Report 1977, NEANDC(E)-192"U" (1978).

2) A. LORENZ, First Coordinated Research Meeting on the Measurement of Transactinium Isotope Nuclear Data, Vienna, 20-21 April 1978, INDC(NDS)-96/N (1978).

Studies on the Decay of ^{241}Pu

R. Vaninbroukx, G. Bortels, P. De Bièvre, B. Denecke, M. Gallet, G. Grosse

The determination of the half-life of ^{241}Pu has been continued using the following methods :

1. Mass-spectrometric determination of the ^{241}Pu decay by measurements of the change in the $^{241}Pu/^{240}Pu$ ratio and the $(^{241}Pu/^{240}Pu)/(^{240}Pu/^{239}Pu)$ ratio of ratios as a function of time;
2. Measurement of the ^{241}Am ingrowth by α counting in a defined low geometry solid angle, and by γ counting using Si(Li) detectors, calibrated for the 60 keV line of ^{241}Am.

The ^{241}Pu half-life results obtained till to the middle of 1978 (final results for the method based on the ^{241}Am ingrowth measurements and preliminary ones for the direct decay measurements using mass-spectrometric techniques, MS) have been compared with other preliminary results obtained at the National Bureau of Standards (NBS), Washington, and the Atomic Energy Research

Establishments (AERE), Harwell. All these results are listed in Table 4.2.
A paper on this subject was presented at the "International Conference on Neutron Physics and Nuclear Data for Reactor and other Applied Purposes" at Harwell, September 1978. Due to the discrepancies which still exist new measurements on well defined material will be necessary.

Table 4.2. : Recent Preliminary Values for the Half-life of ^{241}Pu

Laboratory	Sample	Period of observation in years	Half-life in years
AERE - MS Ref. (1)	1	24	14.25 ± 0.10 14.31 ± 0.10
- MS Ref. (2)	2-A	6	14.24 ± 0.12 14.53 ± 0.12
	2-B	7	14.53 ± 0.08 14.33 ± 0.11
- Ingrowth Ref. (3)	2	-	14.56 ± 0.15
NBS - All MS Ref. (4)	SRM 946 SRM 947 SRM 948 UK-131 5d5a-2e 5d5a-7e	6 6 12 10 8 8	14.406) 14.402) 14.387) 14.368) 14.322) 14.383) 14.38 ± 0.07
CBNM - MS	ORNL	2	14.30 ± 0.14
- Ingrowth	ORNL	1.5	14.60 ± 0.10

1) M. LOUNSBURY, G.R. HALL, E.A. CROUCH, U.K. Nuclear Data Progress Report, UKNDC (78) P 88 (1978) 97.
2) E.A. CROUCH, U.K. Nuclear Data Progress Report, UKNDC (78) P 88 (1978) 97.
3) C. WHITEHEAD, U.K. Nuclear Data Progress Report, UKNDC(76) P 86 (1977) 41.
4) E.L. GARNER, Private communication (July 1978).

Studies on the Decay of ^{241}Am

W. Bambynek, B. Denecke

The measurements to determine the intensity of the 60 keV γ rays in the ^{241}Am decay are continued. The source mount has been improved to avoid damage of the backing foils when changing the sources. Experiments to prepare thinner source backings will go on. New photomultipliers are used which are much better adapted to the wave length of the light emitted by the CsI crystals.
The growing dead layers on the surface of the crystals deteriorate the resolution and give rise to additional absorption which can hardly be corrected for. We will remove these dead layers and measure their residual thickness with a scanning device, which is built into a vacuum chamber and controlled by a microprocessor system. This scanning device is now finished and the microprocessor software is under test.

Studies on the Decay of ^{133}Ba

H.H. Hansen, D. Mouchel

The experiments for the half-life determination have been pursued. The Ge(Li) measurements performed so far during nine months indicates a half-life of 10.5 years. Some similar experiments made with a NaI(Tl) γ-ray spectrometer during the last 4 years yielded values between 10.46 and 10.51 years for the half-life. However, due to the short observation period (Ge(Li) experiments) or due to the restricted number of observations (NaI(Tl) experiments) rather important uncertainties have to be assumed. Thus, it will be necessary to continue these measurements.

For the determination of the internal conversion electron intensities after being energetically selected an improvement of the electron detection can be stated. Serious difficulties arose due to the unfavourable signal to noise ratio and/or due to perturbations entering the detection system probably via the mains supply. Such effects have been found if using a plastic scintillator (NE 102) together with a phototube or a surface barrier detector operated at room temperature. Only if a Si(Li) detector cooled by liquid nitrogen was used the signal to noise ratio did improve considerably and no additional disturbing phenomena could be observed.

In Fig. 4.3. three examples of pulse height spectra for 197.8 keV electrons are shown for a plastic scintillator (NE 102) plus phototube, for a silicon surface barrier detector at room temperature and for a Si(Li) probe cooled with liquid nitrogen, respectively.

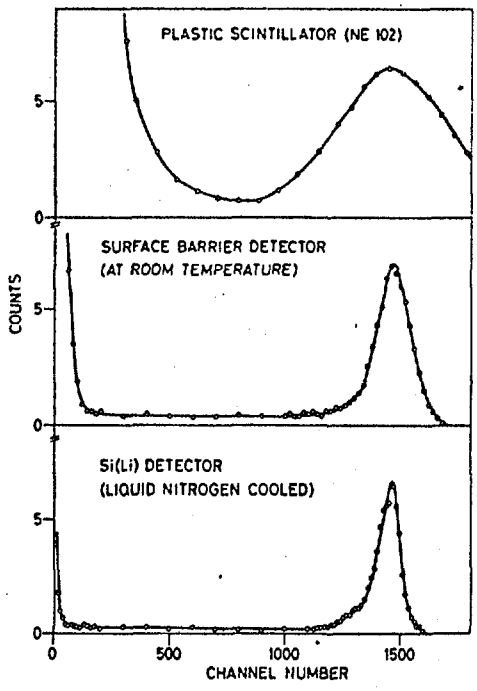

Figure 4.3 Pulse height spectra for electrons of energy E_e = 197.8 keV selected with the magnetic spectrometer and detected as indicated.

Studies on the Decay of $^{103}Rh^m$

R. Vaninbroukx, G. Grosse, W. Zehner

For the study of the reaction $^{103}Rh(n,n')$ at the Van de Graaff accelerator the $^{103}Rh^m$ disintegration rates of the irradiated Rh foils have to be determined. The two main difficulties for such determinations are : (1) the high degree of self-absorption of the emitted radiations, and (2) the lack of knowledge with sufficient accuracy of the decay parameters of $^{103}Rh^m$. As a first step, an investigation of the most important decay parameters has been started. Solutions of $^{103}Rh^m$ have been prepared by extracting the $^{103}Rh^m$ from a ^{103}Pd solution obtained from TRC, Amersham, using ion exchange techniques. The radionuclidic purity of the ^{103}Pd solution was checked with an intrinsic Ge detector. No γ lines except those belonging to ^{103}Pd and its daughter $^{103}Rh^m$ could be observed. The measurement of the radioactive concentration of the extracted $^{103}Rh^m$ solutions is one of the most important parts of the investigation. Because of the low decay energy of this nuclide, the accuracy of counting techniques using solid sources, even thin ones, is strongly limited by self-absorption effects. For this reason we applied the liquid scintillation technique, where corrections for self-absorption are not necessary. A device

equipped with a RCA 8850 photomultiplier of a high multiplication factor at the first dynode was used. A typical energy spectrum obtained with this device is shown in Fig. 4.4. Integration and extrapolation to energy zero yield the disintegration rate of the sample. From the position of the peak relative to that of the one-photoelectron peak one can deduce that the mean number of photoelectrons hitting the first dynode of the photomultiplier is about 10. Assuming that the distribution of these photoelectrons is a Poissonian $P_n(m)$, where m is the mean number per event, the probability that n electrons hit the first dynode is $P_n(m) = \frac{m^n}{n!} e^{-m}$. The zero probability is $P_o = 5.10^{-5}$ and consequently the efficiency attainable by extrapolation to zero energy is 100 %. A further advantage of the liquid scintillation technique is the very simple and quick source preparation procedure, without any time-consuming drying. This allows measurements within the first hour after the separation, which is important in view of the short half-life of $^{103}Rh^m$ (56 min).

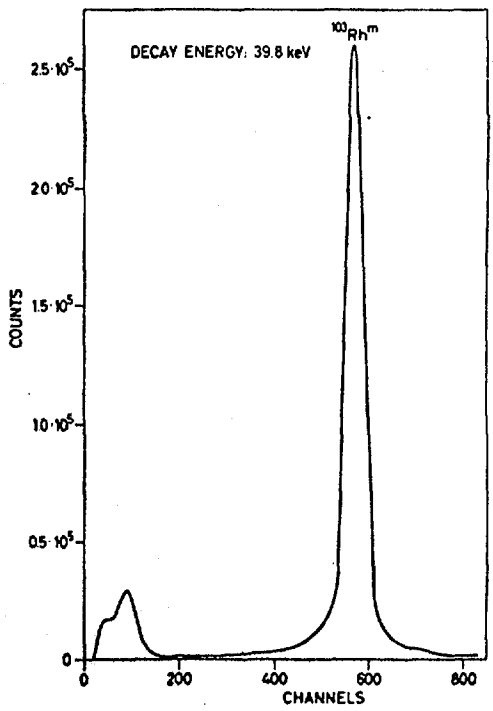

Figure 4.4 Measurement of the disintegration of a $^{103}Rh^m$ solution with the liquid scintillation technique.

Six extractions have been made and each time two sources for liquid scintillation and two sources for photon-emission rate measurements were prepared. The amount of solution used for each source was determined accurately by weighing. The KX- and γ-ray emission rates measurements could only be started one hour after the separation for allowing the deposited drops to dry. A calibrated Si(Li) detector was used for the measurements. A typical photon spectrum is shown in Fig. 4.5.

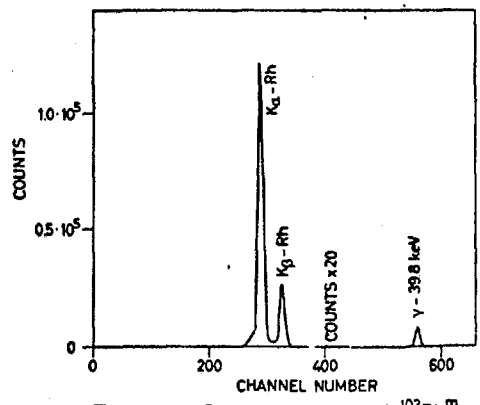

Figure 4.5 Photon spectrum of $^{103}Rh^m$.

The preliminary results of these measurements are :

I_{KX} = 0.084 ± 0.005,

$I_\gamma(39.8)$ = 0.00070 ± 0.00004,

Total conversion coefficient

α = 1430 ± 89.

Combining the above values of I_{KX} with the K-shell fluorescence yield of Rh, $\omega_K = (0.807 \pm 0.031)$ [1] the K-conversion electron emission rate can be calculated as $I_{eK} = (0.104 \pm 0.009)$ yielding a K-conversion coefficient of $\alpha_K = (148 \pm 18)$. The values of I_{eK} and I_γ are in good agreement

with those of Grunditz et al. [2] being respectively (0.104 ± 0.009) and (0.00072 ± 0.00006). However, our I_{KX} value is definitely higher than the value of I_{KX} = (0.676 ± 0.005) published more recently by Czock et al. [3].

The conversion coefficient values α_K and α agree within the quoted uncertainties with the theoretical values α_K = 139.4 and α = 1429 obtained by interpolation from the tables of Hager and Seltzer [4] and Dragoun et al. [5].

The measured KX-ray emission rate is also in good agreement with the value deduced from the theoretical α_K and the ω_K values.

A further decay parameter that was measured during this investigation is the half-life of $^{103}Rh^m$. The preliminary result is : $T_{1/2}$ = (56.1 ± 0.1)min, in good agreement with the values of (56.116 ± 0.009)min published by Günther et al. [6].

1) W. BAMBYNEK, B. CRASEMANN, R.W. FINK, H.-U. FREUND, HANS MARK, C.D. SWIFT, R.E. PRICE, P. VENUGOPALA RAO, X-ray fluorescence yields, Auger and Coster-Kronig transition probabilities, Rev. Mod. Phys. 44 (1972) 716.

2) Y. GRUNDITZ, S. ANTMAN, H. PETTERSON, M. SARACENO, Studies in the decay of ^{103}Pd, Nucl. Phys. A 133 (1969) 369.

3) K.H. CZOCK, N. HASELBERGER, F. REICHEL, The disintegration of $^{103}Rh^m$. Intern. J. Appl. Radiation Isotopes 26 (1975) 417.

4) R.S. HAGER, E.C. SELTZER, Internal conversion tables, Nucl. Data Tables A 4 (1968) 1.

5) O. DRAGOUN, Z. PLAJNER, F. SCHMUTZLER, Contribution of outer atomic shells to total internal conversion coefficients, Nucl. Data Tables A 9 (1971) 118.

6) E. GUNTHER, K. KNAUF, K.F. WALZ, The half-life of $^{103}Rh^m$, Intern. J. Appl. Radiation Isotopes 24 (1973) 87.

Determination of Half-lives of ^{57}Co, ^{103}Pd and ^{109}Cd

R. Vaninbroukx, G. Grosse

Several sources of ^{57}Co, ^{103}Pd and ^{109}Cd were used for checking the long-term stability of the detection efficiency of Si(Li) detectors in the energy range between 10 keV and 25 keV e.g. for the half-life measurements on $^{93}Nb^m$ and for the investigation on the Rh KX emission rates. As a check of the sources they were remeasured at regular intervals over the whole period of observation with a 7.5 cm x 7.5 cm NaI(Tl) detector for which the long-term reproducibility is known to be better than ± 0.05 %. As a by-product, these stability measurements were used for the calculation of the half-lives of these radionuclides. The results obtained till now are summarized in Table 4.3. where the uncertainties quoted are at the 1 σ level taking into account random and systematic effects.

Table 4.3. : Preliminary Results of the Half-lives of ^{57}Co, ^{103}Pd and ^{109}Cd

Nuclide	Number of sources	Number of solutions of different origins	Period of observation in half-lives	Half-life in days
^{57}Co	8	4	2.0	271.8 ± 0.3
^{103}Pd	4	2	1.8	17.0 ± 0.1
^{109}Cd	6	3	1.3	461.7 ± 0.5

Half-life Measurements of Excited Nuclear Levels

A. Nylandsted-Larsen, H.H. Hansen, D. Mouchel

The half-life of the first excited nuclear level of ^{133}Cs at 81 keV has been measured by the delayed coincidence method. The electron capture decay of ^{133}Ba was utilized to populate the 81 keV level. The delayed time distribution was obtained by using the 356 keV γ ray detected in a 3.8 cm x 3.8 cm NaI(Tl) crystal as start pulse and the 81 keV γ ray detected in either a 3 mm x 3.8 cm or a 8 mm x 3.8 cm NaI(Tl) crystal as stop pulse. The NaI(Tl) crystals were mounted on RCA 8575 photomultiplier tubes. A preliminary value of (6.23 $\pm$ 0.03) ns has been obtained for the half-life, where the quoted uncertainty is the standard error of the mean. A detailed examination of the different contributions to the systematic uncertainty is in progress. The value of the half-life compares well with recent published values ranging from 6.25 ns to 6.36 ns. Further measurements will be performed using other combinations of radiation detectors.
Measurements on ^{119}Sn, which were postponed due to detector problems, are now going on.

Compilation of Internal Conversion Data

H.H. Hansen

A series of additional experimental data on internal conversion which came lately to the compiler's knowledge has been incorporated into the compilation of internal conversion coefficients and ratios. A list of input data as atomic number, transition energy and multipolarity necessary to calculate the theoretical conversion coefficients has been prepared. The evaluation of nuclides often used for detector calibration has been started.

Evaluation of Fluorescence Yields

W. Bambynek

A reevaluation of X-ray fluorescence yields has been started in order to include new results into the list of recommended data which was published six years ago [1]. The main effort has been spent to evaluate K-shell data.

1) W. BAMBYNEK, B. CRASEMANN, R.W. FINK, H.-U. FREUND, HANS MARK, C.D. SWIFT, R.E. PRICE, P. VENUGOPALA RAO, X-ray fluorescence yields, Auger and Coster-Kronig transition probabilities, Rev. Mod. Phys. 44 (1972) 716.

REFERENCE MATERIALS AND TECHNIQUES

5. PREPARATION AND CHARACTERIZATION OF SAMPLES

Sample Preparation Service

R. Eyckens, E. Freistedt, H. Mast, J. Mast, M. Parengh, J. Pauwels, F. Peetermans, J. Tjoonk, J. Triffaux, J. Van Audenhove, J. Van Gestel, A. Verbruggen*

The objective is to prepare well characterized samples and targets to be used in nuclear measurements, to meet special requirements of both CBNM and other institutions, as well as to supply either by purchase or on loan special nuclear materials to CBNM - and the Community neutron scientists.
Samples in the form of thin films or foils were prepared by electrospraying, including suspension spraying, vacuum deposition, spraypainting or rolling. Alloys with certified composition or bulk samples were produced by melting, including high frequency levitation melting, rolling, powder metallurgy and conventional machining techniques.
The degree and quality of their characterization depended on the application and varied from crude film thickness measurements by an oscillating quartz device to the extremely accurate mass assay by α-counting and isotopic, including dilution analysis.
The total number of samples delivered during 1978 amounted to about 470, covering 77 different applications (Table 5.1.).

The Preparation and Characterisation of Reference Fission Foils (1)

J. Pauwels, F. Peetermans, J. Van Audenhove, A. Verbruggen*

As a typical example of accurately defined nuclear reference samples made available by CBNM we could mention this year the ^{239}Pu fission foils as needed for absolute fission rate measurements. This work took place in the frame of an international collaboration with National Bureau of Standards (Washington), Hanford Engineering Development Laboratory (Westinghouse) and the Belgian Nuclear Research Centre (SCK/CEN) at Mol.
Important numbers of fission foils in nine thicknesses ranging from 0.07 to 120 $\mu g/cm^2$ were prepared via vacuum deposition onto stainless steel backings of PuF_3, obtained by dry hydrofluorination.
To achieve optimum uniformity and increase productivity an evaporation system was devised with multi planetary rotating substrate holders. Deposits prepared in subsequent evaporation runs are measured relatively by low-geometry alpha-counting. Subsequently, some of the deposits are assayed for the absolute number of ^{239}Pu atoms by isotope dilution mass spectrometry (IDMS). The thus defined calibration factor allows absolute characterization of the remaining deposits with an accuracy depending on :

- the precision of the low geometry alpha countings of each target and of the determined calibration factors;
- the accuracy of IDMS (typically ≤ 0.4 % for Pu amounts > 1 μg).

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* SCK/CEN, Mol, Belgium

TABLE 5.I.

Samples delivered in 1978

Materials	Applicants See list(a)	Numbers of Samples	Definition Method See list(b)	Preparation Method See list(c)
^{233}U	(4)(10)	20	α-count.-IDMS	VD
^{234}U	(1)	4	α-count.-IA	ES
^{235}U	(1)(2)(3)(10)(11)(12)	133	α-count.-IA	ES-VD-P
^{235}U	(13)	24	MD-DC-IA	Pr-C-Mach.
^{236}U	(1)	1	MD	
^{238}U	(3)(2)(12)	10	MD-IDMS	VD
^{239}Pu	(4)(3)	39	α-count.-IDMS	VD
^{232}Th	(1)(3)	9	MD-XRF	ES
^{237}Np	(1)(3)(5)	7	MD	ES
^{240}Pu	(3)(5)(10)(11)	38	α-count.-IDMS	VD
Al-^{239}Pu	(9)	6	QA-IA	LM-R-C
^{235}U-0.5 % ^{240}Pu Nb-0.5%	(3)	16	QA	LM-R
^{235}U	(2)	1	QA	LM-R
Al-16 % U	(14)	6	QA	LM
^{104}Pd	(3)	1	MD	Pr-C
^{105}Pd	(3)	1	MD	Pr-C
^{106}Pd	(3)	1	MD	Pr-C
^{110}Pd	(3)	1	MD	Pr-C
Gd	(6)	1	MD	P
^{6}LiF	(3)	29	MD	VD
^{6}Li	(6)	1	MD	C
V	(6)	30	MD	R-Mach.
nat B.	(7)(8)	15	MD	VD
ZrO_2	(3)	1	MD-DC	Pr
Nb	(6)	30	DC	R-Mach.
Fe	(6)	10	DC	R-Mach.
Current Services : Al,Fe,S,Cu, Ti,Ta,In,Au, Ni,Cr,Vyns, Polyimide	(3)	50	DC-MD	VD,R or Mach.

(a) List of applications

(1)	Univ. Bonn	(D)
(2)	Max von Laue-P.Langevin	(F)(D)
(3)	CBNM	(EUR)
(4)	CEN Saclay	(F)
(5)	Rijksuniversiteit Gent	(B)
(6)	S.C.K.-C.E.N. Mol	(B)
(7)	Univ. Louvain-la-Neuve	(B)
(8)	Univ. Liège	(B)
(9)	Univ. Kiel	(D)
(10)	NBS Washington	(USA)
(11)	HEDL Westinghouse Hanford	(USA)
(12)	CEA Bruyères-le-Châtel	(F)
(13)	ESARDA NDA	(EUR)
(14)	KfK-Jülich	(D)

(b) List of definition methods

MD	:	Mass Definition
α-count	:	Counting of α-particles
IDMS	:	Isotope Dilution Mass Spectrometry
IA	:	Isotope Analysis
DC	:	Dimensional Control
QA	:	Quantitative Alloying
XRF	:	X-ray fluorescence

(c) List of preparation methods

ES	:	Electrospraying
VD	:	Vacuum Deposition
Mach.	:	Machining
R	:	Rolling
LM	:	Levitation Melting
Pr	:	Pressing
C	:	Canning
P	:	Painting

This means that independently from the knowledge of half-lives, activity contribution of minor isotopes, uncertainty on absolute geometry factor etc ..., deposits are characterized at ± 0.5 % provided sufficiently good statistics can be obtained on the relative alpha-countings. Independently from the good homogeneity (± 0.5 % for rotating substrates) and the fact that sets of quasi-identical (± 2 %) deposits can be obtained, the adherence of the fluoride deposits seems to be excellent.

The adhesion of 100 $\mu g/cm^2$ PuF_3 deposits on stainless steel has been determined by the following simple tests :

- "Macrofol" plastic foils, as used for fission track measurements, pressed firmly against the entire deposit and then taken off.
- A cellophane tape pressed firmly over the entire deposit and then stripped off.

Resistance tests to different mediums have been conducted :

- Immersion in tap water at room temperature during 4 days.
- Immersion in boiling water during 1 min.
- Immersion in CH_3OH, CH_3COCH_3 and rubbing of the wet deposit with Kleenex.

Activity losses remain below detection limit (≤ 0.3 %) after these different adhesion and corrosion tests.

The transfer of activity to the "Macrofol" foil is of the order of 0.03 %.

For the near future similar programmes for ^{233}U, ^{235}U, ^{238}U, ^{240}Pu, ^{242}Pu and ^{232}Th are in preparation.

1) J. VAN AUDENHOVE, P. DE BIEVRE, J. PAUWELS, F. PEETERMANS, M. GALLET, A. VERBRUGGEN, The Preparation and Characterization of Reference Fission Foils. World Conference of the International Nuclear Target Development Society, München, 11-14 September 1978.

Reference Materials for Non-metals in Non-ferrous Metals

J. Pauwels, F. Peetermans, J. Triffaux, J. Van Audenhove

The influence of non-metallic impurities on the technological properties of non-ferrous metals makes their accurate control of foremost significance. The availability of suitable reference materials is therefore a primary requirement of industrial laboratories [1].

About 50 laboratories participate in 19 ongoing projects covering the analysis of oxygen, nitrogen, carbon, boron, phosphorus or sulphur in one or different metals, alloys or compounds. Their work is coordinated by four specialists groups for "surface related problems", non-metals in Al, Cu, Pb and Ni, non-metals in refractory metals and non-metals in sodium.

JRC Geel is responsible for the scientific secretariat of the BCR working- and specialists groups.

During 1978 four meetings were organized, three certificates and reports approved by ACPM-BCR [2][3][4] and four reports published as BCR information [5][6][7][8]. The presently available certified reference materials are summarized in Table 5.2. Furthermore draft reports were prepared on intercomparisons for oxygen in lead and alloys [9] and oxygen and nitrogen in nickel [10].

Furthermore JRC Geel has been asked to ensure a centralized sample preparation service in the frame of this activity. In 1978 several different non-ferrous metal samples have been prepared for interlaboratory tests, and methods have been investigated for their mechanical shaping in order to reduce residual surface contaminations of O, N, C, P and B. Metallographical examination and measurements of residual surface contamination have been continued in collaboration

with the University of Liège, the University of Compiègne and CNRS Orléans [11].
Recommended mechanical and chemical treatments have been updated (Table 5.3.) and will be published as BCR-information.
Particular attention has been paid to present tool and machining parameters for turning in a standardized and easily understandable form (Fig. 5.1.)

Table 5.2. : BCR-CRM's for Non-metals in NF-metals

BCR No.	Certified Reference Materials	Certified Value (µg/g)
18	Oxygen in phosphorous-deoxide copper	70 ± 7
21	Oxygen in zirconium	1182 ± 35
22	Oxygen in E.T.P. copper	138 ± 7
23	Oxygen in molybdenum	14.7 ± 2.1
24	Oxygen in titanium	608 ± 23
25	Oxygen in primary ingot aluminium	< 0.1
54	Low oxygen in copper	1.2 ± 0.3
55	Oxygen in refined pure lead	1.0 ± 0.5
56	Oxygen in zirconium	477 ± 30
57	Oxygen in zirconium	498 ± 25
58	Oxygen in continuous-cast copper rod	390 ± 12
59	Oxygen in titanium - 6 % aluminium - 4 % vanadium alloy	1750 ± 70

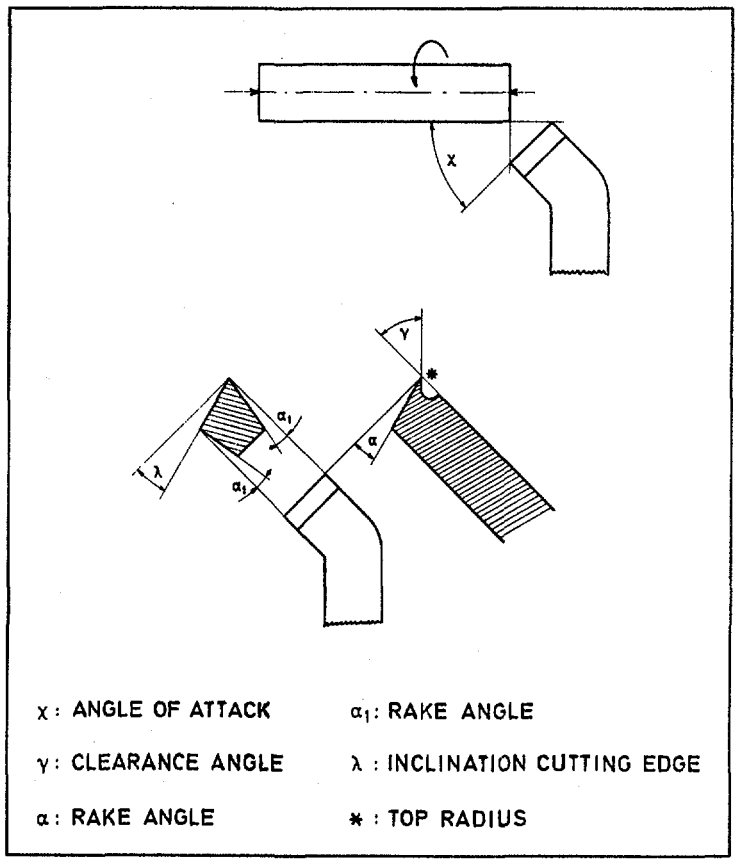

Figure 5.1 Tool parameter for turning.

1) VAN AUDENHOVE J. - Euroanalysis III, Dublin (Aug. 1978).
2) PAUWELS J. - BCR/91(5)/77(1978).
3) PAUWELS J. - BCR/92(5)/77(1978).
4) PAUWELS J. - BCR/197(4)/77(1978).
5) PAUWELS J. - EUR 5932 EN (1978).
6) PAUWELS J. - EUR 5517e (1976).
7) PAUWELS J. - EUR 5934 EN (1978).
8) PAUWELS J. - EUR 5935 EN (1978).
9) PAUWELS J. - BCR/61(2)/78(1978).
10) PAUWELS J. - BCR/85(2)/78(1978).
11) QUAGLIA L. et al., ITE-Report N° 90, Eurisotop Office, Brussels (1976).

Table 5.3. : Recommended Surface Treatments and Residual Surface Contaminations

METAL	Surface treatment		Residual Surface contents ($\mu g/cm^2$)			
	Mech. Shaping(*)	Subsequent Etching(**)	Oxygen	Carbon	Nitrogen	Fluor
Al - 99.5	T, R	1 HF + 1 HNO_3 (20°C, 2 min)	0.3 - 0.4	0.1 - 0.2		Without etch < 0.01
Al - Si	T, R	Idem	0.2 - 0.4			
Cu	T	a) HCl (20°C, 2 - 3 min) b) 1HNO_3 + 1CH_3COOH(20°C, 1 min)	0.2 - 0.4			
Pb PbSnCd PbCuTe	T, R, P	3 CH_3COOH + 1 H_2O_2 (20°C, 45 s)	0.2 - 0.4	0.15 - 0.2		
Ni	T, SiC	75 CH_3OOH + 25 HNO_3 + 1.5 HF (60°C, 60 s, ultrasonics)	0.1 - 0.2	0.15 - 0.2	< 0.03	
Ti	T, S, HSS	4 HNO_3 + 1 HF (20°C, 1 min)	0.4 - 0.6	0.1 - 0.3	< 0.1	
TiAl6V4	T, HSS	Idem	0.3 - 0.6	0.1 - 0.3	< 0.05	
Zr	T, S	5 HNO_3 + 0.5 HF + 5 H_2O (20°C, 50 s)	0.4 - 0.6	0.1 - 0.2	< 0.05	
W	S	1 HF + 4 HNO_3 (20°C, 2 min)	0.1 - 0.2		< 0.05	
Mo	S, HSS	a) 4 HF + 1 HNO_3 (20°C, 10 s) b) HCl (20°C)	0.2 - 0.4	0.1	< 0.05	

(*) Mech. Shaping : T : turning on a lathe; S : sawing with a diamond saw; SiC : sawing with a SiC saw; R : rolling; P : punching; HSS : high speed sawing

(**) Chem. Etching : Bath is given in volumes of each component (concentrated acids)
Arrest is carried out in water (when not stated otherwise) and rinsing by dipping in two more water vessels and three methanol vessels.
Drying carried out in warm air stream (≈ 60°C).

Reference Materials for Traces of Noble Metals

J. Pauwels, F. Peetermans, J. Triffaux, J. Van Audenhove

Minning and refining noble metals industry have solicitated the preparation of suitable reference materials for analytical purposes. JRC Geel is reponsible for the scientific secretariat of, and the sample preparation for the BCR-sub working group in charge of this activity. A report on the determination of Pt and Pd in Cu has been published (1).
Preliminary reference batches of Cu samples doped with known amounts of Ag and Au, have been analyzed by the participating laboratories and results discussed in February 1978. The preparation of three reference materials of Ag and Au/Cu (≈ 100 kg) is planned for 1979.
Technical annexes for contracts have been prepared and submitted to BCR.

1) PAUWELS J. - EUR 5916 EN (1978).

6. ANALYTICAL TECHNIQUES

6.1. ELEMENTAL MEASUREMENTS

Actinide Reference Materials Programme

H.L. Eschbach, Y. Le Duigou, V. Verdingh

This programme started on the advice of ACPM-METRE Nucl. and a study of the needs of actinide reference materials (ARM) in the European Communities has been carried out. As one of the results a document (CBNM AS/15/78) was produced containing the most urgent needs for ARM in the EC as compiled from the different experts' reports received at CBNM. A number of ARM with highest priority was indicated. In order to deal with these first, the experts' group asked that special working parties be established for the following topics :
RMs for elemental analyses : U metal, U oxide, Pu metal and Pu oxide;
RMs for isotopic analyses : ^{230}Th, ^{233}U, ^{238}U, ^{242}Pu, ^{244}Pu and Pu isotopic mixtures.
Meanwhile, two working parties, U elemental and Pu elemental, composed of specialists in the field from different countries, have been created. During its second meeting the working party for the elemental analysis of uranium acknowledged the high quality of the work performed on the existing French U metal RM and decided that some verification measurements should be performed before an EC certification could be recommended. Regarding the characterization of the UO_2 reference material of UK origin, a more elaborate measurement campaign was worked out. The following analyses will be performed by different laboratories : panoramic assay, selected impurities, oxygen to uranium ratio and direct uranium measurements.
The working party for the elemental analysis of plutonium came to similar conclusions concerning the existing French Pu metal standard. Regarding the characterization of the PuO_2 reference material of U.K. origin an elaborated measurement campaign was worked out. The following analyses will be performed by different laboratories : selected impurities, oxygen to plutonium ratio, isotopic composition and direct Pu measurements.

Development and Application of Reference Techniques

M. Aerts, Ch. Berthelot, H. Herrmann, Y. Le Duigou, W. Leidert, A. Michiels, L. Van Hengel, V. Verdingh

Chemical Determination of Main Constituents

Coulometric methods are widely used for the determination of uranium or plutonium contents in nuclear materials. In order to improve working conditions a new cell, as shown in Fig. 6.1.1., has been designed for electrolysis at a platinum electrode. By this new cell configuration it is intended to decrease the time of titration and to obtain a more efficient deoxygenation of the solution because of the reduced volume and the correspondingly smaller quantities of electrolyte in the cell.
The cell has been tested for the reduction or oxidation of plutonium at a platinum electrode. The volume of solution in the cell has been decreased from 20 to 10 ml. The quantity of plutonium used is 25 mg per determination. The magnitude of the correction due to the integration of the background current during an electrolysis is about 0.05 %. The mechanism of a secondary photochemical reaction during the coulometric determination of uranium in a mixture with iron has been described and appropriate working conditions to obtain reliable uranium

measurements have been established. See also LE DUIGOU Y., LEIDERT W., Fresenius A. Anal. Chem. 289 (1978), 279.

Chemical Determination of Trace Elements

For different requestors from CBNM a service is maintained to determine the impurities of materials that are to be used for nuclear measurements. According to the individual requests one or more of the following methods is applied : emission spectroscopy, spectrophotometry, atomic absorption spectroscopy, oscillo- and pulse-polarography and gas extraction for gas analysis by heating. Two additional methods have recently been added : photon activation and ion induced x-ray analysis. They are particularly suited for multielemental determination of trace constituents.

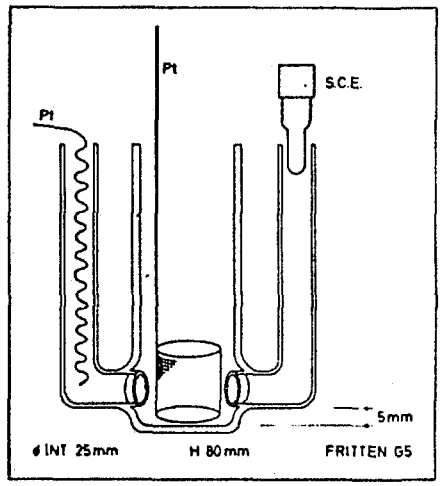

Figure 6.1.1 Cell for coulometric analysis

The calibration of the installation for the determination of oxygen by reducing fusion has been extended to the range 400 ppm down to 1 ppm. The calibration is performed by direct injection of known volumes of carbon monoxide. Pure carbon monoxide and calibrated CO-Argon mixtures (50 % CO, 50 % Ar and 2 % CO, 98 % Ar) were used as absolute oxygen standards. With this calibrated instrument a series of steel-oxygen standards were measured and good agreement was achieved with the standards in the concentration range between 10 and 100 ppm oxygen with standard deviations (σ) from 0.6 to 2.1 ppm. These results were achieved for series of 10 measurements and at extraction temperatures of 2750 K.

The calibration of the nitrogen analysator was also performed by direct gas injection. In this case pure nitrogen and calibrated N_2-He mixtures were used as absolute nitrogen standards. Applied to nitrogen-containing steel RMs excellent agreement in the concentration range 10 ppm to 350 ppm with σ ranging from 0.7 ppm to 3.4 ppm could be obtained.

An independent calibration method suited for the determination of nitrogen in refractory metals was tested. Standards were prepared by filling small gold cups with 2-3 mg TiN under helium atmosphere. For the heat extraction 1 g of pure nickel was added as flux. Using this method less than 0.7 ppm equivalent was observed from the Au cups or from the flux.

Apart from routine spectographic analyses of traces in various matrixes a method to determine ppm amounts of thorium in uranium had to be developed. Traces of thorium were separated by means of anion exchange in HCl medium. Th is not absorbed on Dowex I resin and the eluted solution is evaporated onto graphite electrodes. Using the 3351 Å, 3392 Å and the 4019 Å Th lines, the limit of detection by C-spark technique is 3 ppm for a sample of 2 g.

The determination of impurities in uranium has been performed by emission spectroscopy.

Traces of rare earth elements in U-metal or -compounds were analysed using a chromatographic concentration method. The concentration is made on a cellulose powder column in ether-nitric acid medium. The analysis is performed on metallic uranium samples of 10 to 20 g. It was carefully checked that the cellulose did not contain rare earth elements. After preconcentration, ashing and mixing with lithium carbonate, the sample is analysed by emission spectrography. The detection limits that can be obtained with this method are 0.5 ppb(Yb) to 5 ppb(Gd).

The main dispersion for a determination of Europium and Yttrium at concentration level of 1.5 ppm and 0.5 ppm respectively with three independent measurements is 10 %.

Reference Materials (other than ARM) for Use in the Nuclear Field

Ch. Berthelot, R. Besenthal, W. De Spiegeleer, H. Herrmann, Y. Le Duigou, H. Ruts, L. Van Hengel, V. Verdingh

An extensive programme of neutron source calibration using an improved manganese bath technique is being organized on a world-wide international level by Section III of the Comité Consultatif pour les Etalons de Mesure des Rayonnements Ionisants.

The induced ^{56}Mn activity is monitored as a measure of source strength. The main problems connected with the manganese solution method are those of calibration of the counting efficiency, correction for thermal absorption, correction for parasitic reactions due to the presence of impurities in the bath, and the determination of the manganese concentration.

An important analytical effort is required to support this project; for the calibration of the manganese concentration, the overall impurity content of about twenty elements, amongst them elements with high neutron cross sections (B, Cd, rare earths at very low concentrations) has to be measured. A variety of analytical techniques will be applied : gravimetric, volumetric, potentiometric titration, emission spectrography, atomic absorption, spectrophotometry, polarography and photon activation.

Preliminary measurements have been performed on a $MnSO_4$ bath used by CBNM in a previous intercomparison. With gravimetry and EDTA titration for the manganese determination uncertainties of < 0.1 % could be obtained with concentrations of 330 mg(Mn)g^{-1} solution. The potentiometric titration of Mn^{++} has also been applied. The oxidation of manganous iron with permanganate in the presence of fluoride has been investigated earlier as a method for determination of Mn^{++} with colorimetric detection of the end-point. However, this method cannot be applied with high concentrations of Mn^{++}. In this case the net reaction

$$MnO_4^- + 10\ HF_2^- + 4\ Mn^{++} \rightarrow 5\ MnF_4^- + 2\ H^+ + 4\ H_2O$$

can be followed potentiometrically, considering the redox potentials MnO_4^-/Mn^{+++} and Mn^{+++}/Mn^{++}. The effects of acidity, fluoride concentration and concentration of manganese were studied.

All measurements are performed with the aid of an automatic potentiometric titrator (type Radiometer) using a platinum-calomel electrode system.

The determination of impurities in the $MnSO_4$ was performed by the application of suitable emission spectrography techniques. The boiler-cup method was used for the direct determination of B and Cd. The limits of detection for this method were B $\leq$ 0.2 ppm and Cd $\leq$ 0.4 ppm, respectively. Rare earth elements (Eu, Sm, Dy, Gd) were determined by preconcentration of their fluorides using Yttrium as a tracer to a limit of 0.05 ppm. Synthetic reference materials containing 0.05 - 0.1 - 0.3 and 0.1 ppm of rare earths were prepared.

For the boron and cadmium determination synthetic RMs containing 0.1 - 0.3 - 1.0 and 10.0 ppm of B or Cd were prepared. Pulse-polarography is used as a very sensitive method for the determination of Cd, $MnSO_4$ being taken as base electrolyte. The detection limit is about 2.10^{-3} μg Cd g^{-1} solution. A coefficient of variation of 1 % is achieved at levels of 100 ppb. One of the analyzed $MnSO_4$ solutions contained about 3 ppm of Cd and 1 ppm of B.

Support to BCR Projects

Y. Le Duigou, W. De Spiegeleer

In the frame of the BCR project of the DG XII to establish reference materials for the calibration of methods used for the elemental analysis of organometallic compounds the triphenyllead-dimidazole and the mercurisuccinide have been tested for stability and purity. Samples have been distributed to a group of laboratories for elemental composition measurements. CBNM is contributing to the lead and mercury measurements using coulometric titrations. Successive oxidation and reduction have been performed on lead in a 0.5 molar NaBr electrolyte after wet destruction of the organic compound in a nitric-sulfuric acid medium. The relative standard deviation of a single lead determination is $\pm$ 0.07 %. Five individual results have been reported.

The technical report describing the work performed on the bis diethyltinchloride oxide and the draft certificate have been written. See GRIEPINK B. et al., EUR 6103 (1978).

The material has been bottled, packed and four hundred bottles, containing 5 g material each, are stored at CBNM and are available for sale as BCR material n° 34.

Photon Activation Analysis

Ch. Berthelot, G. Carraro, T. Mencarelli, H.M. Silvester, V. Verdingh

With the installation of the receiving and recovery station for the activated samples, the set up for the photon activation analysis was finally completed. For the non-destructive analysis of Sm, Eu, Gd, and Dy in boron carbide irradiations have been performed at the Linac, using bremsstrahlung produced by the impact of an electron beam (32 MeV, 33 μA) on a water-cooled platinum target. Photon activation has been chosen because the determination of rare earths in boron carbide by neutron activation analysis meets with difficulties due to the large capture cross section of boron 10 for thermal neutrons. On the other hand, the absorption of high energy photons by boron and carbon is very small and the whole sample can be activated homogeneously.

The boron carbide, synthetic standards prepared from graphite, doped with 10 to 4000 ppm of each of the four lanthanons (Sm, Eu, Gd, Dy) and pure graphite (blank sample) are encapsulated in aluminium cans. For the irradiation, the standards are placed before and behind the sample; the whole assembly is placed in an aluminium sample holder which in itself is mounted in a rabbit (52 mm Ø). This can be rotated (120 rpm) during γ-irradiation in front of the conversion target at a distance of about 10 mm. The rabbit is transported by pneumatic transfer to the recovery station where it arrives 4 seconds after the irradiation has been stopped.

The induced activities are measured using a Philips single open-ended coaxial Ge(Li) detector (54.6 cm^3) and the γ-ray spectra are recorded with a 8180 Canberra Multichannel Analyser.

The Fig. 6.1.2. shows the gamma-ray spectrum of a boron carbide sample.

The location and the intensity of each photopeak are calculated using the GAFIT programme. The values for the γ-ray intensities agree well with those determined by the total peak area and Wasson methods. The Table 6.1.3. gives the limits of detection for an irradiation time of 7.5 h and a decay time of 10 h before starting of the counting. The detection limits corresponds to three times the background standard deviation at the appropriate energy in a boron carbide sample.

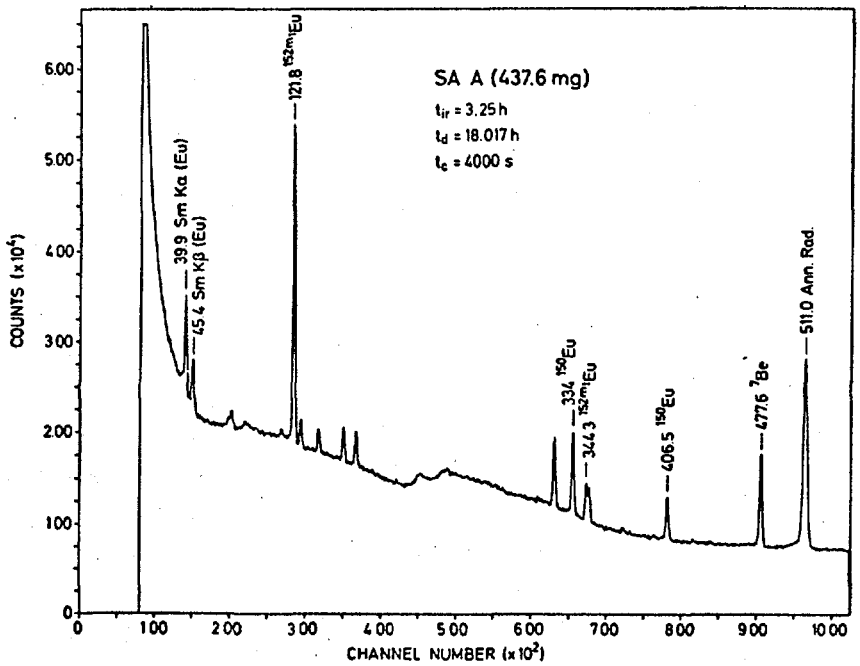

Figure 6.1.2 Spectrum of a boron carbide sample.

Table 6.1.3. : Detection Limits of Rare Earths

Element	Sm	Eu	Gd	Dy
Detection limit (μg)	0.082	0.23	0.28	3.5
γ-line (keV)	103.180	121.783	363.508	326.2
Radionuclide	^{153}Sm	^{152}Eu^{m}	^{159}Gd	^{157}Dy

The irradiation and rotating device has been used to check the accuracy and the homogeneity of the standards. Four samples encapsulated in Al cans, are placed in an cylindrical Al block at equal distances around the rotation axis so that each sample is alternately irradiated by high-energy photons. The following samples were used :

1) 2.11 mg Eu_2O_3 - 2) 4000 ppm.w Eu - 3) 1000 ppm.w Sm, Eu, Gd, Dy (each) - 4) empty Al can.

A known amount of europium oxide is weighed directly in the aluminium can, mixed in the container with graphite and finally encapsulated. The irradiation conditions were : t_{ir} = 3,50 h at 40 MeV, 26 μA. The ^{152m}Eu activities in the standards compared to the activity of Eu_2O_3, agree well with the nominal composition.

In another experiment, two portions of the same standard - 4000 ppm.w Sm - are alternately irradiated using the rotating device for the control of homogeneity.

The use of photon activation for the non-destructive analysis of Sm, Eu, Gd and Dy in boron carbide, has been presented at the 5th Symposium on Recent Developments in Activation Analysis, Oxford (July 1978), and the paper is accepted for publication in the Journal of Radioanalytical Chemistry.

As a support to the $MnSO_4$ programme instrumental photon activation for the analysis of impurities was used.
To improve experimental conditions a decelerating system, to slow down the speed of the rabbit before it reaches the radiation station, is now set up.

Surface Analysis by Rutherford Backscattering

I.V. Mitchell, K. Barfoot *, W.D. Dobma, E. Louwerix, P. Rietveld, H.L. Eschbach

A paper reporting on CBNM measurements on bismuth implanted silicon samples to determine the absolute bismuth dose by Rutherford backscattering has been published in Nucl. Inst. and Meth. 149 (1978) 727. Its abstract reads as follows :
"The need for backscattering standards appears to be twofold and depends on the uses and requirements of the users. The first is as a calibrated reference by which samples of a similar nature to the standard may be absolutely compared. The second is as a means of intercomparing the relative results obtained by different laboratories using, as near as possible, identical samples. This type of comparison is of a relative nature and the absolute values are not necessarily required. In the present work we try to satisfy both needs by providing identical samples which have been absolutely calibrated to a high accuracy.
Very thin copper and vanadium layers were evaporated onto bismuth implanted silicon crystals and on glass plates under carefully controlled conditions. The mass of the deposits was determined in situ using a sensitive UHV microbalance. In addition, two quartz oscillator monitors were used.
The samples have been analysed by Rutherford backscattering and the absolute quantity of bismuth determined by a comparison with the known amounts of deposited material."
Differences between the results obtained here and the results obtained on the same samples by using current measurement to determine the ion dose have recently led to a reevaluation of the absolute value of the Rutherford cross section for the energies of interest in Backscattering analysis. Discrepancies of up to 4 % have thus far been found in the absolute values of Rutherford cross sections. (Ref. J. Davies, Chalk River, private communication).

Ion Implanted Reference Standard

I.V. Mitchell, W.D. Dobma, E. Louwerix, P. Rietveld, H.L. Eschbach

Work on the production of an ion implanted reference standard is continuing and a small batch of polished vitreous carbon discs have been sent to two outside laboratories to be ion implanted. A heavy and a light atomic weight ion has been chosen for implantation into each of the discs and their energy and dose chosen so that their range and concentration profiles should coincide. Calibration of the implantations will take place by the method described earlier.
Difficulties have been experienced in producing a large number (more than 300) of identically polished vitreous carbon discs. It appears that even from the same batch of base material differences in texture and pitting occur from disc to disc. Several different polishing techniques have been used to investigate this problem but to date the problem remains unsolved.

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Stochiometry of Si_3N_4 Thin Layers

I.V. Mitchell, W.D. Dobma

Interest has been shown in recent years on using silicon nitride as a passivating layer in the silicon microelectronics technology. The stochiometry of nominally Si_3N_4 plasma-deposited thin layers produced under a variety of different conditions on Si and SiO_2 substrates have been investigated using 2 MeV $^4_2He^+$ Rutherford backscattering. Whilst it appears from preliminary results that the Si to N ratio varies from the stochiometric value the analysis was hampered by the spectral interference of oxygen in an underlying silicon oxide layer.

Ion Beam Intensity Monitor

I.V. Mitchell, W.D. Dobma, K. Barfoot *, P. Rietveld, H.L. Eschbach

A novel method to monitor the intensity of an ion beam has been developed. A thin self-supported gold foil whose thickness has been determined very accurately is posed in the incident ion beam. A fraction of the ions backscattered from the gold atoms is measured by an annular surface barrier detector. This arrangement is very sensitive to small fluctuations of the primary ion beam. It is very useful in normalising experimental parameters especially in cases where a Faraday cup measurement of the ion beam intensity is not feasible. A paper describing this method has been presented at the Oxford Conference on Recent Developments in Activation Analysis. Continuing this work ion beams of protons and deuterons as well as $^4_2He^+$ of energies between 1 and 3 MeV have been used to test the stability of the self-supporting foil and the consistency of the values of ion current obtained using different ion species.

Trace Analysis by the Particle Induced X-Ray Emission

I.V. Mitchell, K. Barfoot *, W.D. Dobma, P. Mason **, W. Gilboy **, H.L. Eschbach

Particle induced X-ray emission (PIXE) studies have been carried out a number of environmental samples using the CBNM 3 MeV Van de Graaff accelerator as the source of X-ray exciting projectiles. This investigation was carried out in collaboration with a research team working at the University of Surrey (UK). Air dust streakers, giving a time resolution of two hours and taken near Guildford (UK) were analysed both by the Surrey group and by CBNM. The thin foil ion beam monitor mentioned earlier was successfully used with the PIXE analysis. Results were communicated in two presentations at the Oxford Conference on Recent Developments in Activation Analysis.

K X-Ray Production Cross Sections

K. Barfoot *, I.V. Mitchell, W.D. Dobma, E. Louwerix

A series of thin electron beam evaporated layers have been deposited on polished vitreous carbon discs and used in a series of experiments the aim of which was to determine with high precision the K X-ray production cross section for a variety of different metals of $Z \leq 32$. These experiments were performed for proton, deuteron and helium beams and for energies spanning the useful PIXE analytical range from 1-3 MeV. The results are compared with other available experimental data and existing theories. The thin film results will be compared with experimentally produced thick film, solid target, data.

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6.2. ISOTOPIC MEASUREMENTS

Support to the Safeguards Inspectorate of the European Community

R. Damen, W. De Bolle, P. De Bièvre, A. Loopmans, G. Müschenborn, E. Sattler, W. Wolters

Support was given to the Safeguards Inspectorate of the European Community in the framework of U and Pu verification measurements. The start of the verification of the EC Safeguards system by the IAEA is resulting in a considerable increase of the number of verification measurements. At the request of Safeguards Inspectorate Luxemburg further collection of samples at inspection site and organizing transports to JRC laboratories for verification measurements have been performed.

Results of "verification" measurements have continued to be compared to "declaration" values (supplied by DCS) and evaluated for agreement or discrepancy.

Tentative threshold values for discrepancies in measurement results of U and Pu content and ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{235}U abundances are further used on a test-basis. Final statements such as "acceptable", "suspicious" or "unsatisfactory" were communicated to DCS for possible further action.

Some quality controls of the verification laboratories were performed.

The experience over the years 1977 and 1978 has made clear that a shorter response time for the evaluation system is required. This needs a highly reliable data transmission system for verification and declaration values between CBNM Geel and the Safeguards Directorate in Luxemburg.

Increasing problems have shown up with the shipment of Inspection samples, namely

- The German Government (PTB-Braunschweig) put new special and more stringent requirements on vehicles for the transport of radioactive materials in Germany.
- The Belgian Government required a new study of the Type B (U) container before granting a re-validation.

This resulted in the performance of a very small number of transports and indeed all radioactive transports had to be stopped on November 1st. 1978.

Interlaboratory Measurement Evaluation Programmes of Feed Solutions of Reprocessing Plants :

- "AS-76" On $^{238}Pu/^{239}Pu + ^{240}Pu$ Ratio Measurements by α-Spectrometry
- "IDA-78" On Mass Spectrometric Isotope Dilution Measurements of Uranium and Plutonium.

P. De Bièvre, G. Bortels, J. Broothaerts, W. Lycke, W. Wolters and collaboration with K.F.K. Karlsruhe, Safeguards Project

The AS-76 Interlaboratory Test Programme is essentially finished. Precise $^{239}Pu/^{240}Pu$ isotope ratio measurements (Relative Standard Deviation < 0.05 % relative) have contributed to the observation of a correlation with $^{238}Pu/(^{239}Pu + ^{240}Pu)$ activity ratio measurements (see 2.2 of Table 6.2.1.) in one of the test solutions. This could indicate a small but significant inhomogeneity in that particular solution caused by e.g. inhomogeneous distribution of Pu in the liquid phase relative to sludge in input solutions of reprocessing plants. It came to our knowledge that similar observations on sludges have been made elsewhere. This conclusion could reduce the usefulness of that particular solution in the Programme. Evaluation and end report

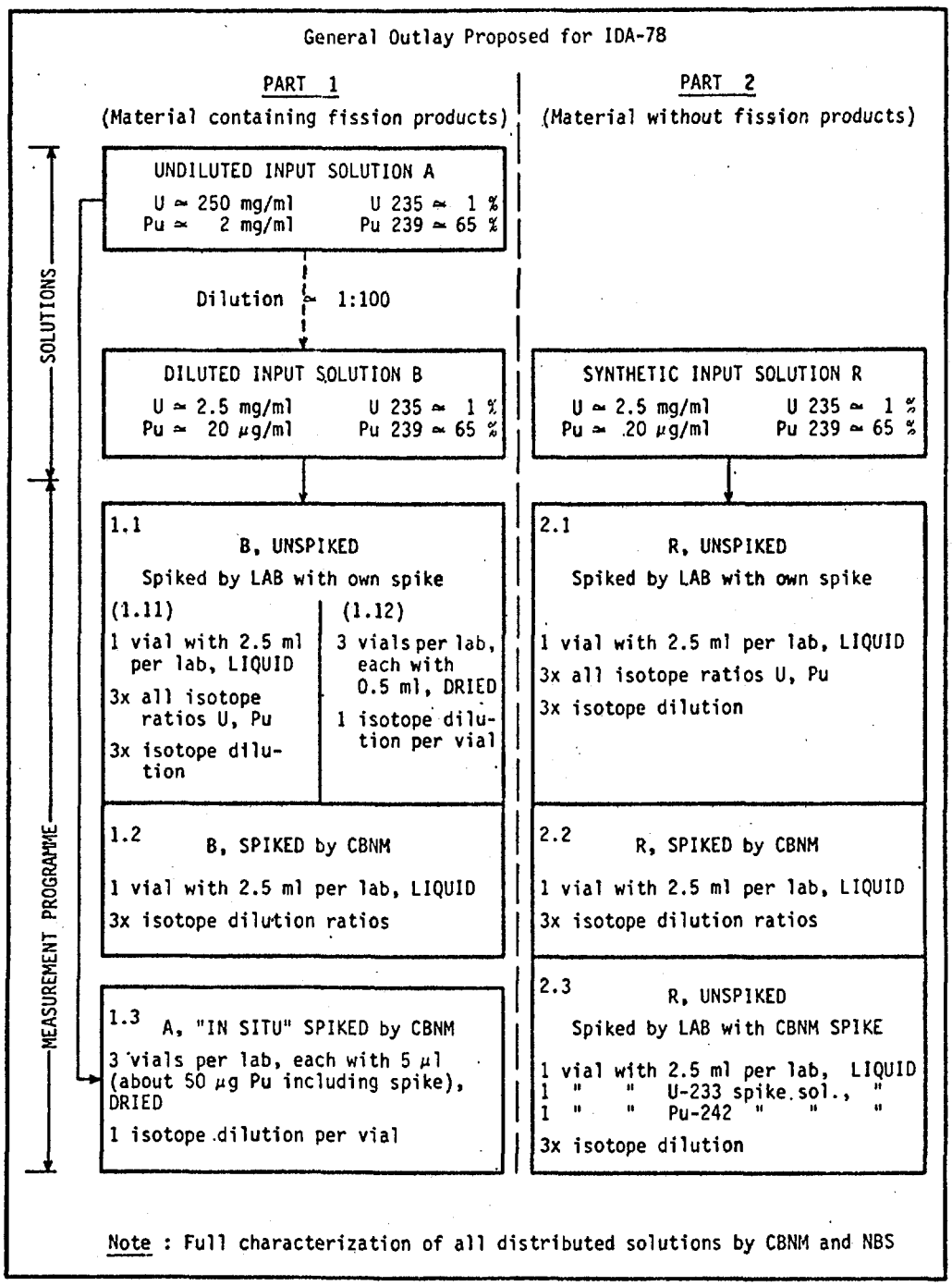
General Outlay Proposed for IDA-78
PART 1
(Material containing fission products)
PART 2
(Material without fission products)
SOLUTIONS
MEASUREMENT PROGRAMME
UNDILUTED INPUT SOLUTION A
U ≃ 250 mg/ml
U 235 ≃ 1 %
Pu ≃ 2 mg/ml
Pu 239 ≃ 65 %
Dilution ≃ 1:100
DILUTED INPUT SOLUTION B
U ≃ 2.5 mg/ml
U 235 ≃ 1 %
Pu ≃ 20 µg/ml
Pu 239 ≃ 65 %
SYNTHETIC INPUT SOLUTION R
U ≃ 2.5 mg/ml
U 235 ≃ 1 %
Pu ≃ 20 µg/ml
Pu 239 ≃ 65 %
1.1
B, UNSPIKED
Spiked by LAB with own spike
(1.11)
1 vial with 2.5 ml per lab, LIQUID
3x all isotope ratios U, Pu
3x isotope dilution
(1.12)
3 vials per lab, each with 0.5 ml, DRIED
1 isotope dilution per vial
1.2
B, SPIKED by CBNM
1 vial with 2.5 ml per lab, LIQUID
3x isotope dilution ratios
1.3
A, "IN SITU" SPIKED by CBNM
3 vials per lab, each with 5 µl (about 50 µg Pu including spike), DRIED
1 isotope dilution per vial
2.1
R, UNSPIKED
Spiked by LAB with own spike
1 vial with 2.5 ml per lab, LIQUID
3x all isotope ratios U, Pu
3x isotope dilution
2.2
R, SPIKED by CBNM
1 vial with 2.5 ml per lab, LIQUID
3x isotope dilution ratios
2.3
R, UNSPIKED
Spiked by LAB with CBNM SPIKE
1 vial with 2.5 ml per lab, LIQUID
1 " " U-233 spike.sol., "
1 " " Pu-242 " " "
3x isotope dilution
Note : Full characterization of all distributed solutions by CBNM and NBS

Table 6.2.1

of the Programme are now in a final stage (Evaluation Meeting in Karlsruhe, February 1978). The IDA-78 programme aims at determining the uncertainty of the analytical measurements of U and Pu isotopes and element contents by isotope dilution mass spectrometry under routine operation conditions. The general outlay is given in Table 6.2.1.
The solid spikes for part 1.3 of the Programme were characterized. They are the first $^{235}U/^{242}Pu$ solid (metal) spikes ever made. Independent measurements at CBNM by Mass Spectrometry of $^{239}Pu/^{240}Pu$ ratio and α-activity of $^{238}Pu/(^{239}Pu + ^{240}Pu)$ ratio and at KFK have however given quantitative evidence that sludges of input solutions could contain more Pu (1-2 %) than the solution (see above). This now puts a suspicion on the original material for IDA-78. The advice of the ESARDA-Working Group on Destructive Analyses was obtained and the use of new material is recommended. It is to be noted that large series of liquid and dried samples had already been prepared and were ready for distribution.

Independent Verification of Mass of Input Solutions at Reprocessing Plants

P. De Bièvre, W. Lycke

A verification method for the independent verification of masses of individual solutions in the accountability tanks of reprocessing plants has been laboratory-tested with very satisfactory results. Indications are that < 0.5 % accurate and independent verification is possible. Field tests should now follow. Combined with the long-time established CBNM procedure for independent verification of U and Pu concentrations in the undiluted input solutions, it may be possible to assay independently total U and Pu in the accountability tanks as well as mass of the total solutions to better than 0.5 %.

Characterization of Reference Samples for Non-Destructive Analysis

E. Bouwmeester, M. Gallet

Enriched as well as depleted uranium samples were characterized for use in CBNM's programme of ^{235}Uranium enrichment determination by non-destructive analysis. On request of the Bureau National de Metrologie, C.E.A., six UO_2 pellets with enrichment between natural and 3 % were characterized. They will serve as French Standards for Non-Destructive Assay.

Isotopic Reference Materials

W. De Bolle, P. De Bièvre, M. Gallet, W. Lycke, M. Müschenborn

- D_2O (99.7 %)

The standing task to intercompare on request D_2O working reference materials in the Community to the D_2O Primary Isotopic Reference Material and Technique of CBNM for accurate Deuterium content was continued (samples from Winfrith). An absolute determination of the oxygen isotopic composition was estimated to be necessary in our opinion and in that of an IAEA panel of experts. It would usefully complete the characterization of our D_2O IRM e.g. for use in density measurements of heavy water samples.

$\underline{UF_6}$

Work on the existing UF_6 Reference Materials for Safeguard measurements at JRC CBNM Geel is going on; delivery of highly enriched ^{235}U to prepare synthetic isotope mixtures, has been negotiated with USA Authorities. Glove-boxes and furnaces have been built and tested. Weight loss of Pt crucible at different temperatures is being determined. A 70 g depleted UF_6 ($^{235}U/U = 0.002$) batch has been hydrolyzed and converted to U_3O_8. Together with a 93 % enriched U_3O_8 it is being used to test out the preparation of synthetic isotope mixtures by weight. Extensive investigation of a 0.05 % systematic error in the $^{235}U/^{238}U$ ratio measurement of converted UF_6 (fluorination of Uranium oxide with CoF_3) led to the discovery that traces of water in the oxide probably cause the formation of traces of UOF_4 during the fluorination. The $^{238}UOF_4^+$ ion at mass 330 appears just above detection level and causes a positive bias on the $^{235}UF_5^+$ ion current measurement on the same mass position 330 in the mass spectrum thus biasing the $^{235}UF_5^+/^{238}UF_5^+$ ratio measurement. The fluorination procedure was modified in order to prevent the UOF_4 formation and hence the biasing influence of UOF_4^+ ions.. The validity of this modification is tested by a) measuring a given UF_6 batch b) hydrolyze the UF_6 and convert to oxide c) fluorinate the oxide back to UF_6 d) intercompare this UF_6 to the original UF_6 under a). Results look promising : the 0.05 % discrepancy between $^{235}U/^{238}U$ ratios in the UF_6 under a) and c) shrunk to < 0.02 %, i.e. to within measurement precision.
As part of the acceptance test of the Eurodif product control mass spectrometer, a programme of UF_6 measurements and preparation of reference samples is carried out.
The usual gas measurement service (130 measurements) was continued.
Requests for UF_6 RMs from Ispra and Almelo have been fulfilled.

- $\underline{\text{Spike Reference Materials } ^{233}U - ^{242}Pu - ^{244}Pu}$

A new stock of ^{233}U and of ^{242}Pu spike solution has been characterized, and provisional certificates are delivered (see Table 6.2.2.).
The delivery of characterized ^{233}U and ^{242}Pu spike solutions has been continued (GWK reprocessing plant Karlsruhe; Kraftwerk Union, Karlstein; Risø National Laboratory, Denmark, JRC Ispra : Reprocessing plant Saluggia, Italy; CNEN Casaccia, Italy).
Some of the deliveries cannot be made as more stringent official rules on containers for transport have caused a new container-approval process with the Belgian Government. A second ^{242}Pu Spike Reference Material at a higher concentration (75 μg ^{242}Pu/g solution) was prepared and provisional characterization is being finished (see Certificate in next Report). The preparation of the first ^{244}Pu Spike Reference Material has been achieved at 1 μg ^{244}Pu/g solution and is being provisionally characterized.
The delivery of highly enriched ^{239}Pu and ^{242}Pu isotopes for the preparation of synthetic isotope mixtures has been discussed for years with US Authorities and is now agreed. The mixtures will serve the characterization of Reference Materials to be distributed for calibration of Pu isotope ratio measurements.

PROVISIONAL

ISOTOPE REFERENCE MATERIAL
CBNM-IRM-041-1 ^{242}Pu

The IRM is supplied with a concentration certified as

(2.3089 ± 0.0070) . 10^{16} atoms ^{242}Pu per gram solution

The above concentration corresponds to

(5.834 ± 0.012) . 10^{-8} moles ^{242}Pu per gram solution.
(9.281 ± 0.028) . 10^{-6} gram ^{242}Pu per gram solution.

Other plutonium isotopes present are related to ^{242}Pu through the atom ratios:

238/242 : 0.011552 ± 0.000035
239/242 : 0.002759 ± 0.000020
240/242 : 0.097867 ± 0.000200
241/242 : 0.026850 ± 0.000080
244/242 : 0.000212 ± 0.000020

Consequently the element concentrations are computed to be:

(4.368 ± 0.013) . 10^{-8} moles Pu per gram solution.
(1.0562 ± 0.0030) . 10^{-5} gram Pu per gram solution.

The isotopic composition of the material is

	Atom %	Weight %
^{238}Pu	1.0140	0.9983
^{239}Pu	0.2421	0.2394
^{240}Pu	8.5906	8.5270
^{241}Pu	2.3568	2.3494
^{242}Pu	87.7779	87.8671
^{244}Pu	0.0186	0.0188

The atomic weight of the material is 241.8127.

PROVISIONAL

ISOTOPE REFERENCE MATERIAL
CBNM-IRM-040 ^{233}U

The IRM is supplied with a concentration certified as

(2.6163 ± 0.0062) . 10^{18} atoms ^{233}U per gram solution.

The above concentration corresponds to

(4.343 ± 0.010) . 10^{-6} moles ^{233}U per gram solution.
(1.0123 ± 0.0024) . 10^{-3} grams ^{233}U per gram solution.

Other uranium isotopes present are related to ^{233}U through the atom ratios:

234/233 = 0.002817 ± 0.000058
235/233 = 0.000024 ± 0.000012
236/233 < 0.000001
238/233 = 0.000555 ± 0.000030

Consequently the element concentrations are computed to be:

(4.359 ± 0.010) . 10^{-6} moles U per gram solution.
(1.0157 ± 0.0024) . 10^{-3} grams U per gram solution.

The isotopic composition of the material is

	Atom %	Weight %
^{233}U	99.6616	99.6592
^{234}U	0.2807	0.2819
^{235}U	0.0024	0.0024
^{238}U	0.0553	0.0565

The atomic weight of the material is 233.0451.

Figure 6.2.2

Establishment of Values of Differences in Measurements to be regarded as Discrepant

P. De Bièvre

A list of "1978" values for differences in U and Pu measurement results considered to be discrepant, has been submitted to and discussed with the European Laboratories measuring Fissionable Material (ESARDA-Working Group on Destructive Analyses) at their semi-annual meeting in November 1978.

It was agreed to try to establish aim-values for :

a) reproducibility (standard deviation for a single determination)

b) uncertainties from corrections for systematic errors

of a number of analytical methods used in Safeguard measurements in Europe.

A list will be submitted for approval at the Spring 1979 meeting of the ESARDA-Working Group.

6.3. RADIOISOTOPIC MEASUREMENTS

Support to Certified Actinide Reference Materials (CARM's) for Non-Destructive Analysis (NDA)

H. Meyer, W. Nagel, F. Quik

U_3O_8 samples in Al containers as common primary standards for the determination of the ^{235}U enrichment of low enriched uranium materials like oxides, metal and fluoride are in preparation in collaboration with NBS, JRC Ispra and KFK-Karlsruhe. In agreement with conclusions of IAEA on the development and identification of safeguards needs for NDA physical standards [1,2], the investigation of standards for the calibration of measurements on plutonium materials should have some priority in future.

A computer aided measuring system for high resolution γ spectrometry with intrinsic germanium counters has been set up and applied for the accurate and reliable determination of the ^{235}U content of uranium samples and for the investigation of parameters influencing the performance of standard samples. The influence of various system parameters on the countrate of the 185,7 keV line of ^{235}U from low enriched quasi-infinite thick U_3O_8, UO_2 and metal samples in Al containers was investigated. A reproducibility of results of 0.3 - 0.5 % for well chosen measurement conditions was obtained for U_3O_8 samples with 0.3 % and 3 % enrichment and for U nat. metal samples; γ countrate ratios of U_3O_8 samples with 0.3 % and 3 % ^{235}U were compared with ratios from mass spectrometer measurements and results were in agreement within the measured reproducibility limits. Transmission measurements on U_3O_8 samples with ^{137}Cs γ's for testing the homogeneity of mass distribution have shown a non-uniformity of $\leq$ 2-3 % which influences γ countrate measurements only to a negligible extent. Investigations of the influence of ^{232}U daughter products on possible measurement biases for low resolution γ counting have underlined that for the application of NaI counters ^{232}U impurities should be smaller than 0.01 ppm of the ^{235}U content of samples for CARM's and of product materials to be used for in-plant calibration with non-destructively characterized samples. One hundred sets of U samples are planned to be used as U enrichment CARM's in Europe and the US.

More thorough consideration will be devoted in future to the development and production of physical standards for Pu isotopic ratio measurements.

Investigations and negotiations for obtaining commonly accepted recommendations for certificates in connection with NDA-CARM's will be supported in collaboration with other bodies like ESARDA, NBS, taking U enrichment RM's as a pilot example.

The establishment of users oriented experts groups from EC countries is envisaged.

1) IAEA Advisory Group Meeting on the Use of Physical Standards in Inspection and Measurements of Nuclear Materials by Non-Destructive Techniques, AG-112, August 1977.

2) IAEA Advisory Group Meeting on the development of NDA Instrumentation and Techniques, AG-187, May 1978 (including standards).

Determination of ^{241}Pu in ^{241}Am Samples

R. Vaninbroukx

Samples of ^{241}Am are prepared by extracting the ^{241}Am grown into ^{241}Pu materials. For several purposes, e.g. for the preparation of high accuracy ^{241}Am reference sources or for cross-

section measurements on ^{241}Am, it may be necessary to know if any appreciable amount of ^{241}Pu has been extracted together with the ^{241}Am. Since accurate ^{241}Pu determinations via β-particle counting are very difficult, the possibilities of γ-ray spectrometry for the determination of ^{241}Pu in ^{241}Am samples were studied using a 7 cm^3 planar germanium detector. The γ rays of the 148.6 keV transition in ^{237}U, which follows the weak α-decay branches of ^{241}Pu, and those of the 208.0 keV transition in ^{237}Np, which follows both the ^{241}Am decay and the ^{241}Pu α branches, have been measured. As shown in Fig. 6.3.1. an amount of 1 wt % (weight percent) of ^{241}Pu in ^{241}Am, contributing less than 10^{-5} to the α-particle emission rate and the 60 keV γ-ray emission rate, can easily be detected. Even for counting times of a few days, which are still reasonable, and for samples of about 1 μg, the detection limit was found to be 0.2 wt %. This work has been published in Int. J. Appl. Radiat. Isotopes.

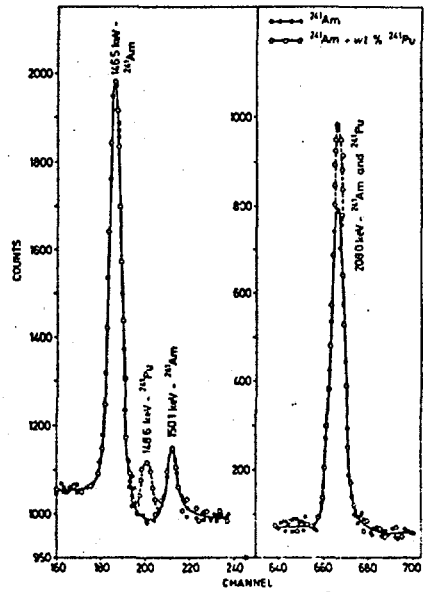

Figure 6.3.1 Observed γ-ray spectra for ^{241}Am and for ^{241}Am + 1wt % ^{241}Pu

International Comparison of Activity Measurements of a ^{134}Cs Solution

A. Nylandsted-Larsen, E. Celen, W. Oldenhof, W. Zehner

The radioactivity concentration of a ^{134}Cs solution distributed by BIPM for an international comparison of activity measurements of a solution of ^{134}Cs has been determined by the $4\pi\beta-\gamma$ coincidence counting method. Three different windows in the γ-ray channel were used :

- around the 605 keV photopeak;
- around the 796 keV photopeak;
- to include the energy range 700 - 1750 keV.

For the windows around the two photopeaks the efficiency functions could be well approximated by first order polynomials, for the integral gamma-gate setting only second order polynomials could be applied. The final value for the radioactivity concentration was found to be 828.4 Bq/mg having a standard error of the mean of 0.3 Bq/mg (0.04 %) with about 160 degrees of freedom; the total systematic uncertainty was estimated to be 1.2 Bq/mg (0.14 %).

International Comparison of Activity Measurements of a ^{137}Cs Solution

A. Nylandsted-Larsen, E. Celen, G. Grosse, R. Vaninbroukx, W. Zehner

The radioactivity concentration of a ^{137}Cs solution, which was distributed by BIPM for an international comparison of activity measurements, has been determined by the $4\pi\beta-\gamma$ efficiency tracer technique using ^{134}Cs as efficiency tracer and by an efficiency calibrated NaI(Tl) detector.

The final value for the radioactivity concentration as measured by the efficiency tracer technique was found to be 752.6 Bq/mg having a random uncertainty of 1.0 Bq/mg (0.13 %) with about 66 degrees of freedom; the total systematic uncertainty was estimated to be 0.42 %. The result

from the measurements using the NaI(Tl) detector was 754.1 Bq/mg with a random uncertainty of 0.8 Bq/mg (0.1 %) and an estimated total systematic uncertainty of 0.4 %.

Characterization of Plutonium Samples for AS-76

G. Bortels, J. Broothaerts, P. De Bièvre, M. Gallet, W. Wolters, I.L. Barnes *, K.M. Glover **

The AS-76 intercomparison was organised as a joint safeguards project of the Kernforschungszentrum Karslruhe, and CBNM, Geel. The objective was to determine the present spread in the α-spectrometric measurement of the activity ratio $^{238}Pu/(^{239}Pu + ^{240}Pu)$ under routine conditions. Four sample solutions have been used. Three of them were prepared from input solutions of a reprocessing plant containing about 0.2 %, 0.8 % and 1.6 % of ^{238}Pu. A fourth solution with about 0.9 % ^{238}Pu originated from reprocessed Pu solution.
The sample solutions have been characterized by α spectrometry at CBNM, Geel and AERE, Harwell, to certify the activity ratio mentioned above, and by mass spectrometry at NBS, Washington and at CBNM, Geel, to certify the isotopic composition. The characterization work was coordinated by CBNM. The results are given in Tables 6.3.1. and 6.3.2.

Table 6.3.1. : Results of the AS-76 Sample Characterization by α-Particle Spectrometry

Nominal ^{238}Pu abundance (%)	α-Activity Ratios $^{238}Pu/(^{239}Pu + ^{240}Pu)$		
	C B N M	A E R E	M e a n
0.8	1.4463 ± 0.0043(0.30%)	1.4455 ± 0.0043(0.30%)	1.446 ± 0.004(0.30%)
1.6	2.984 ± 0.010 (0.35%)	2.9866 ± 0.009 (0.30%)	2.985 ± 0.010(0.35%)
0.9	1.6361 ± 0.0057(0.35%)	1.6382 ± 0.0050(0.30%)	1.637 ± 0.006(0.35%)

Table 6.3.2. : Values and Accuracies of the Atom Ratios of $^{238}Pu/^{239}Pu$ in Samples from Different Characterization Laboratories and Methods

Nominal ^{238}Pu abundance (%)	Values and Accuracies of the Atom Ratios $^{238}Pu/^{239}Pu$			
	From α-Spectrometric Measurements of Activity Ratios $^{238}Pu/(^{239}Pu + ^{240}Pu)$		From Mass Spectrometric Measurements	
	C B N M	A E R E	N B S	C B N M
0.8	0.01224 ± 1.0 %	0.01223 ± 1.0 %	0.01212 ± 1.2 %	0.01221 ± 0.7 %
1.6	0.02596 ± 1.0 %	0.02598 ± 1.0 %	0.02598 ± 0.57%	0.02596 ± 0.7 %
0.9	0.01440 ± 1.0 %	0.01442 ± 1.0 %	0.01425 ± 1.1 %	0.01436 ± 0.7 %

* National Bureau of Standards, Washington, USA

** Atomic Energy Research Establishment, Harwell, UK.

The indicated accuracies corresponds to a 3 σ confidence level and include an estimate of the systematic uncertainties. For the calculation of the atom ratios from the measured activity ratios, as given in Table 6.3.1., the following half-lives have been used [1] : $T_{1/2}(^{238}Pu)=87.74 \pm 0.09$ years, $T_{1/2}(^{239}Pu)=24100 \pm 30$ years and $T_{1/2}(^{240}Pu)=6553 \pm 8$ years. The samples containing about 0.2 % of ^{238}Pu were found to be inhomogeneous. This solution had been prepared by spiking a given input solution successively with enriched ^{239}Pu and ^{240}Pu. Differences in the α-activity ratio up to about 3 % have been observed and a linear relationship was observed between the atom ratio of $^{239}Pu/^{240}Pu$ from mass spectrometry and the α-activity ratio of $^{238}Pu/(^{239}Pu + ^{240}Pu)$. It is assumed that the inhomogeneity is due to particulate matter present in the solution.

1) A. LORENZ, First Coordinated Research Meeting on the Measurement of Transactinium Isotope Nuclear Data, Vienna, 20-21 April 1978, INDC(NDS)-96/N (1978).

Mass Determination by α-particle Counting

B. Denecke

About 50 samples have been measured on request and relating certificates have been issued. The activity is determined by counting α particles in a defined solid angle. From the half-lives of the components and the isotopic composition of the sample the mass of the requested nuclide can be deduced.

Source Preparation

W. Oldenhof, D. Reher, W. Zehner

The preparation of thin, homogeneous radioactive sources is important for accurate measurements of the intensities of radiations. An improvement of quantitative source preparation is to utilize the freeze-drying process. For this purpose a micro-freeze dryer has been constructed and installed in a glove box. It is now used to prepare quantitative sources. Furthermore, studies to make thin sources with an Ar-ion gun are going on.

Measurement of Suspended Particulate Matter in Air

W. Bambynek, B. Denecke, Z. Diamantidis

An Environmental Programme of the European Community is going on. It concerns a "common procedure for the exchange of information between the surveillance and monitoring networks based on data relating to atmospheric pollution caused by certain compounds and suspended particulates" (Council Decision 75/441/EEC). Harmonization of these data, especially of suspended particulate matter in air is strongly requested.
In this context the Environmental and Consumers Protection Service (SEPC) is going to set up about 18 dust sampling stations at various places inside the Member States.
On request of SEPC techniques have been developed at JRC-Geel to identify automatically dust loaded filters and measure the amount of particulate matter by light reflectometry and by electron absorption. The measuring device has been set up and tested. The launching phase has been finished and sampling and measuring of the filters will start at the beginning of 1979.

6.4. CLASSICAL METROLOGY

B. Dyckmans-Van Hout, H. Eschbach, F. Hendrickx, P. Rietveld, J. Verdonck, F. Verheyen, R. Werz

The service of conventional metrology was called upon by different CBNM groups, mainly for mass and length measurements, for alignments of experimental arrangements (e.g. for time of flight measurements).

Thickness measurements on thin films and foils was performed by weighing as well as by x ray fluorescence analysis. For the determination of the uniformity and thickness of suspension sprayed ThO_2 layers on VYNS backing, conversion factors were determined in order to be able to use the calibration curve established previously for uranium. This was possible due to the new, very stable, triode x ray tube together with a highly stabilized power supply. Accurate thickness determinations by x ray fluorescence can be disturbed by the crystallinity of the backing material giving rise to diffraction spots. This effect seems to be very pronounced in molybdenum. Systematic experiments are being performed in order to find the best working conditions for the different material combinations.

The influence of the backing material on accurate layer thickness measurements by the x ray fluorescence technique has been further investigated. A number of carefully prepared layers with thicknesses between 200 and 300 $\mu g\ cm^{-2}$ were made by vacuum evaporation. Different metallic backings were used. The results of these measurements are shown in Fig. 6.4.1. This clearly demonstrates that in order to obtain highest accuracy only layers with the same backing material should be compared. For less accurate determinations diagrams like Fig. 6.4.1. can be used for interpolations.

The thickness measurement on thin self-supporting foils has been further improved. A set of high precision Au foils evaporated and weighed in ultra-high vacuum was fabricated for the final calibration. The thicknesses of the foils were between 128 and 1576 $\mu g\ cm^{-2}$. The energy loss of α particles from a thin 148 Gd-source traversing the foils was measured. The results are given in Fig. 6.4.2.

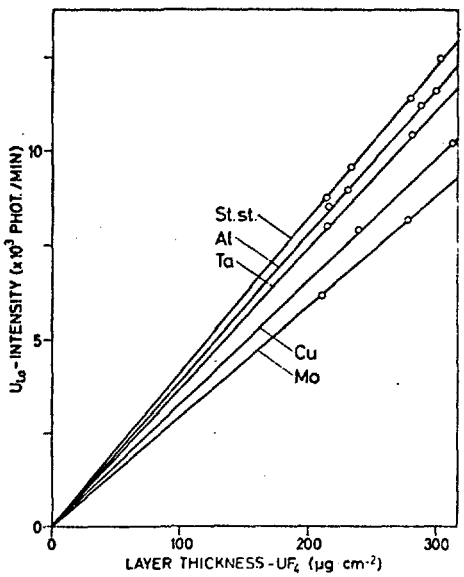

Figure 6.4.1 Calibration curves for layer thickness measurements by X-ray fluorescence

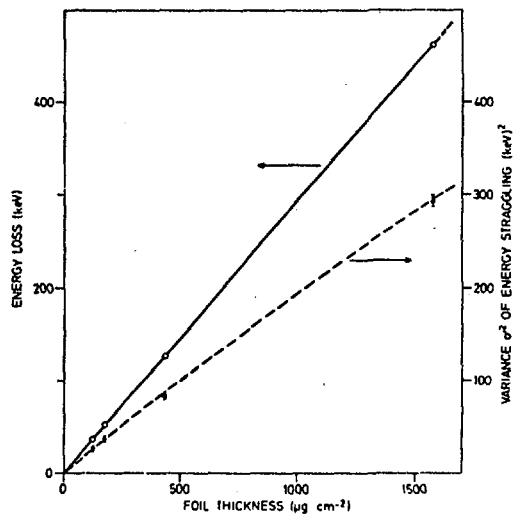

Figure 6.4.2 Measurements on gold foils, energy loss and straggling.

For thin foils the uncertainty of the measurements is about 1 $\mu g\ cm^{-2}$. At the other end of the curve the uncertainties amount to about 1 % of the energy loss.
From the peaks measured also the energy straggling can be obtained with rather high accuracy. The dashed line in Fig. 6.4.2. shows the energy straggling measured for the four gold foils.

MAJOR INSTALLATIONS AND TECHNICAL DEVELOPMENTS

7. ACCELERATORS

"GELINA" Geel Electron Linear Accelerator

R. Cools, R. Forni, F. Massardier, F. Menu, K. Meynants, J.M. Salomé, P. Siméone, F. Van Reeth, J. Waelbers, C. Waller

The Linac was operated regularly (Fig. 7.1.) with the parameters given in Table 7.1. The electron beam was available for 3171 hours being mainly used for neutron measurements by, on an average, five experimenters at the same time.

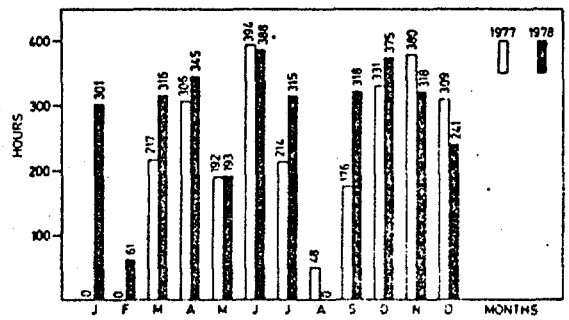

Figure 7.1 Linac operation Useful electron beam hours per month.

The neutron beams were always produced from a cylindrical mercury cooled uranium target (Ø 30 mm). No damage occured to this target which dissipated up to 5 KW. Development of the rotary target and beryllium canned moderators was proceeding on during the period covered, but not yet completed.

Table 7.1 : Electron Beam Parameters

Pulse length ns	Rep. rate Hz	Peak current A	Mean energy MeV	Time Hour	% of the time
4 - 5	800	9 - 10	110	1740	54.9
10	400 - 500	9 - 10	100	957	30.2
20	100	6.5	100	241	7.6
2000	250	0.12	32 - 45	42	1.3
Miscellaneous				191	6

In view of the preparation of the next programme, a firm was contacted to study the possibilities of increasing the electron beam intensity for short pulses. The first results show that two ways could be followed :

1. Enlarge the cathode diameter and improve the gun optics. A new triode gun has been developed and will be installed for measurements in our laboratory with high voltage of 100 kV and 10 kV - 100 ns - 100 Hz on the grid. The structure is not yet designed for short pulses but only to get high currents on a collector.
2. Develope a post acceleration buncher which utilizes the fall of electron energy during the pulse in the highly loaded sections. The feasibility of this proposal depends critically on the energy spectrum of the electron microbursts. A method has been developed to measure these spectra. The electron beam is magnetically deflected on a fast target.

A good quality sampling scope is used to display the signals which are resolved for each microbunch. Preliminary results show that the decrease in energy along the pulse is very linear, but the microburst spectra are dependent on many parameters. Measurements are going on to get more information.
Several irradiations have been performed for photon activation analysis. Until now it was not possible to deflect more than 40 μA at 45 MeV on the platinum converter. Electron beams with such parameters are very sensitive to the klystron power level as well as to the focusing magnetic fields on the sections, one of them in fact inducing very little energy to the electrons Work is in progress to improve the focusing conditions and increase the mean current available at this electron energy level.

Van de Graaff Accelerators

A. Crametz, P. Falque, J. Leonard, R. Smets

Due to the fact that both accelerators fired their particles into the same target hall, they cannot be used simultaneously. Accelerators were stopped during 200 hours day-time for repair of the building.

Operation and maintenance of the CN-7MV Van de Graaff accelerator

Out of 2410 working hours, 1710 hours were used for neutron physics experiments, including 120 hours for work in cooperation with T.H. Eindhoven. The remaining hours were used for maintenance and the conditioning of the accelerator up to 7MV.
The pressure vessel has been opened seven times, once to repair the HF oscillator and six times to exchange the ion sources.
At the beginning of the year, the life time of the ion sources was only around 150 hours, because of a leak on the glass-metal welding but with CBNM built ion sources the life time has been increased to 450 hours.
Orders have been placed for beam handling components, including a quadrupole lens, for a third beam extension on the 45° line after the switching magnet.
For the determination of the pulsed beam time performance, a 200 $\mu g/cm^2$ gold target has been installed on a special holder and introduced in the 0° beam extension near the target in order to scatter particles out of the beam. The signal on the visual readout of a multichannel analyser is such that a better adjustment of the voltages for the pulsed and bunched beam will be possible. Electronics are nearly completed and experiments will be made early next year.

Operation and maintenance of the KN-3.7MV Van de Graaff accelerator

This accelerator was operated for 510 hours of which 480 hours were used for Rutherford backscattering experiments. In addition, 75 hours were needed for maintenance and the exchange of the source. The reorganisation of the beam tubes is in progress and a switching magnet and two quadrupole lenses have been ordered for the installation of several experiments. The switcher is such that it will be possible to align the six beam tubes ($\pm$ 15°, $\pm$ 30° and $\pm$ 45°) with a bidirectional laser and to place the samples to be irradiated with high precision on the beam.

8. COMPUTER AND DATA ACQUISITION SYSTEMS

8.1. CENTRAL COMPUTER SYSTEM

Operation of the Central Computer

C. Cervini, J. Deckx-Van Baelen, D. De Pooter, G. Ehrenfreund, H. Horstmann

In order to increase the power for batch processing and time-sharing work the central processing unit IBM 370/135 has been replaced by an IBM 370/138 in January 1978. As before data acquisition stations (minicomputers and multichannel analysers) throughout the CBNM can transfer their data directly to the disk storage of the central computer.
A system of 6 display terminals (3 x IBM 3277/2 and 3 x ELBIT DS 377) has been connected to the IBM 370/138 computer ("local attachment") in order to increase the efficiency in the analysis of experimental data.
The computer time was used for the following activities :

- Neutron Data 83 %
- Non-Neutron Nuclear Data 6 %
- Reference Materials and Techniques 3 %
- System Software Development and Maintenance 6 %
- Administration 2 %

The total number of CPU running hours amounts to 3509.
The generation of the system software OS/VS1 Release 6 was prepared and executed. For the terminal system VSPC, VTAM and VSAM had to be studied in some detail before the application programmer could be made to work in VSPC. Up to now only VSPC FORTRAN is available for the terminal user, but the use of APL and TSIO is being prepared.

Support to Scientific Application Programmers

C. Cervini, G. Ehrenfreund

A series of routines for handling experimental data in the form of spectra on the central computer has been developed, in particular for transfers between various input/output units and for bookkeeping of the stored data.
An on-line programme for the analysis of unsorted capture cross section data on the central computer has been installed. ADC and TOF (time-of-flight) values are used to calculate in a weighting procedure several time-of-flight spectra from the measured TOF data.
In order to prepare the use of APL for the application programmer several general utility routines for the handling and storage of experimental data have been developed.

Support and Development Work for Administrative Problems

U. Meloni, C. Cervini, D. Gelly

The CBNM administration has been supplied with computer results for all kinds of personnel data. Statistics on careers and various other calculations have been performed. The financial service has been regularly supported in its accountancy and bookkeeping problems, including programme modifications where necessary.

Programme CORIG used at Ispra to solve the accountancy problems of the establishment has been modified for the use on the CBNM IBM 370/138 computer system. A test case has been run successfully.
Specifications for a programme for the control of fissile material at CBNM have been worked out. After similar work at the Ispra Establishment of the JRC the development of a common programme has been contracted out by CETIS to the firm IRCA.

8.2. DATA ACQUISITION SYSTEMS

Extension and Improvement of the CBNM Data Transmission System

A. De Keyser, H. Nerb, P. Van Roy, H. Mensch

The CBNM Data Transmission System consists of a parallel dataway interconnecting data acquisition stations in the different buildings of the centre and is linked by a supervising computer (SYCON) to the disk storage of the IBM 370/138 central computer. The system is now in operation for many years though giving problems whenever data from several sources need to be transferred at frequent intervals.
The connection of two more multichannel analysers (Canberra) has been studied and the necessary interface cards have been developed. Also special peripherical driver cards have been produced, which allow the use of faster printers and video terminals.
A fourth building crate (Mass Spectrometry) was connected to the system and all hardware supporting the CBNM alarm system (see below) has been installed.

Acceptance Tests and Improvements of the CORRSYS Multiparameter Data Acquisition Station at the Linear Accelerator

A. De Keyser, A. Idzerda

The CORRSYS data acquisition station is a stand-alone two-user multiparameter system controlled by a GA 16/460 minicomputer. A list mode multiparameter experiment and a two-parameter experiment with a special weighting procedure for capture cross section measurements can run simultaneously. The system is built by the firm CORRELATIVE SYSTEMS (Brussels) according to CBNM specifications and it is hoped to be fully operational by the first half of 1979.
Support has been given for the connection of a CAMAC branch to the system and several improvements of the CAMAC driver software have been made. The operating system of the minicomputer has been studied.

Modernization of Neutron Data Acquisition Stations

A. De Keyser, H. Horstmann

Specifications for two multiparameter data acquisition systems for measurements at the electron linear accelerator have been worked out in order to obtain price quotations. Each system is to control two experiments with a total event rate of about 6000 s^{-1}. One experiment is a list mode type measurement with one TOF (time-of-flight) and up to six ADC parameters. The other experiment can either be a single parameter measurement with sample changer control or a two parameter measurement, for which up to 128 windows on the TOF parameter or up to 32 windows on the ADC parameter can be used. A spectrum for which data are accumulated in one of these windows can have up to 4096 channels.

Finally an order was handed over to Nuclear Data for two modified versions of their ND 6660 system.

Data Storage System for Stand-Alone Multichannel Analysers
P. Klopf, W. Stüber

Specifications for a data storage system for stand-alone multichannel analysers have been sent to 25 firms. Incoming tenders have been analysed carefully. Finally the project had to be abandoned because of too high costs.

Installation of a Minicomputer and a Mass Storage for Multichannel Analysers
D. Reher

For the measurements of non-neutron nuclear data a system consisting of a minicomputer and a mass storage for several multichannel analysers has been conceived. The computer (PDP 11/34) runs under a foreground-background operating system, so that the process, i.e. the transfer of spectra to and from disk is not prohibited during off-line calculations. The first part of the system is installed. The connexions between the multichannel analysers and the computer will be completed in 1979.

Software Development for a Multiparameter Neutron Data Acquisition System
T. Babeliowsky

The system is based on a GA 18/30 minicomputer connected to the CBNM data transmission system and controlling a CAMAC branch with two CAMAC crates for two users. Input data from time coders and ADC's are derandomized in so-called double data buffers. The experiment is monitored by counters one of which can trigger a sample changer. In the MUP-2 system a multichannel analyser function with a total of 16K channels is programmed. For each sample changer position the accumulated spectrum is transferred to the disk storage of the central system. In-core spectra as well as spectra stored already on the disks of the central computer can be displayed. All system and application programming has been finished. The system is in a test phase. A list mode system (MUP-1) is also available. In this case the users have available more than 16K channels because the unsorted data are sent directly to the disks of the central computer and sorted by the GA 18/30 minicomputer in time-sharing with the data acquisition process.

Improvement of the Data Acquisition Software for Capture Cross Section Measurements
T. Babeliowksy

The data acquisition station for capture cross section measurements is based on a GA 18/30 minicomputer and connected to the CBNM data transmission system. Because of high data rates and limited capacity to the minicomputer memory, the ADC and time coder data are transferred to buffers on the disk storage of the central computer and analysed by this computer. Several extensions of the minicomputer software have been made, in particular to improve the monitoring of the experiment by scaler data.

Software Development for a Multi-User Single Parameter Data Acquisition System
A. Idzerda

The system is based on a GA 18/30 minicomputer, connected to the CBNM data transmission system and controlling a CAMAC branch with four CAMAC crates and two users per crate. Each user has an ADC and eight counters, one of which controls the experiment as a preset-counter. A dynamic overflow programme allows for storage of data in single precision with hundred channels in double precision, thus reducing the required space in core.
At the end of a preset interval all accumulated data are transferred to the disk of the central system and the next run is started. A retry-programme assures continuation of the runs after a period of breakdown of the CBNM data transmission system. The system became fully operational at the end of 1978. Some extensions are planned for the next year.

Installation of a Test Computer for CBNM Data Acquisition Systems
A. De Keyser, T. Babeliowsky, K. Hofmans

At CBNM four General Automation GA 18/30 computers are in use, one serving as the control computer of the data transmission systems and the three others connected to this system as satellite data acquisition stations. Furthermore a rather large amount of CAMAC circuitry is in use all over the centre.
In order to fulfill the demands for testing of new equipment and to solve maintenance and development problems a fifth machine has been installed as an independent test station, completed with the necessary interface units and a CAMAC branch. The connection of the test computer to the data transmission system has been prepared, as well as circuitry for the simulation of a branch of the data transmission system.

CBNM Alarm Control System
T. Babeliowsky, W. Stüber

An alarm system has been developed for the detection and the central display of alarm situations on the CBNM site, caused by fire, water, radioactivity, wrong pressure in glove boxes, etc. Various types of alarm detectors are connected to alarm comparators, which are scanned periodically by alarm scanners. If an alarm occurs, a minicomputer is informed by the corresponding scanner and can read detector number and status via the general CBNM data transmission system. The computer then sends an alarm message to the CBNM guards. Per building up to 512 alarm detectors can be connected to the system. The existing prototypes of alarm scanner and alarm comparator have been improved. The necessary software has been developed. First the mass spectrometry and the linear accelerator building were connected to the alarm system in order to gain experience before the connection of all alarm detectors.

8.3. DEVELOPMENT OF ELECTRONIC EQUIPMENT FOR COMPUTER SYSTEMS AND EXPERIMENTAL INSTALLATIONS

Two Nanosecond Time Coder
S. de Jonge

The first fast time coder which came out of production demonstrated bad reproducibility due

to high tolerances in the fast phasing and counter circuits which were not met by the supplier. The problem has been solved by modifications in the instrument and by the use of other integrated circuits (see also Internal Report CBNM/DE/17/76 by S. de Jonge).
For test and diagnostic purposes concerning single and multiparameter measurement equipment provided with this type of fast coder a time-of-flight test pulser has been developed. This instrument simulates linear accelerator signals as pre-trigger and time reference as well as timing and amplitude signals which are normally delivered by a detector. A prototype has been built which has to be improved because there is still some structure in the random time output.

Control Unit for α Source Scanning Device

B. Denecke, H. Nerb, Z. Diamantidis

The hardware and software for driving a scanner to measure α-sources of large sizes have been developed. Angular rotation and axial translation are provided to move every point of the source under the detector. Both movements are executed by stepping motors controlled manually or by a KIM-microprocessor. The programmes of the microprocessor allow any type of measuring track across the source area. Apart from the control unit a scaler is provided for the counting of the detected α-particles. Results are printed on a teletype. The equipment is in a test phase.

CAMAC Based Circuitry

A. De Keyser, P. Klopf, L. Peeters, W. Stüber, P. ter Meer

A series of units conform with the CAMAC standard have been developed for various purposes :

- An adaptor to interface a sample changer to a controller.
- An eight-filter sample changer controller to be driven by the CORRSYS multiparameter data acquisition system. On line tests of this unit still have to be carried out.
- Two CAMAC crates completely filled with plug-in units for measurements of neutron data and one crate for measurements on radionuclides have been installed and tested.
- A multiparameter multiplexer formatter combining the time-coicident binary output data words of a time-of-flight coder, a 4-bit routing unit and up to 6 pulse-height converters (ADC) into one multiparameter word with up to 104 bits. The word size can be reduced by omission of arbitrarily prescribed groups of bits and the reduced multiparameter word can be transmitted in groups of 16 bits to a double data buffer. The circuit design is almost completed.
- A CAMAC display buffer allowing the display of histograms and text on an X Y oscilloscope without periodic refreshing by a computer. It contains a 4K x 14 bit semiconductor memory, which will be loaded once by the computer via the CAMAC dataway and will then be scanned periodically for display. Circuit design and print design have been completed. A light-pen facility is being studied.

Miscellaneous Electronic Units

S. de Jonge, E. De Roost, K. Hofmans, P. Klopf, P. ter Meer, M. Van Aelten *, J. Van Gils **, L. Van Rhee

- A counting system for photon activation measurements at the linear accelerator has been developed. It measures and prints the irradiation duration, the start and duration of the counting time and the number of coincidences between two pairs of γ-spectrometers measuring the annihilation radiation of relatively fast decaying isotopes.
- An Oscilloscopic Davis Polarograph based on a Tektronix oscilloscope equipped with two CBNM - developed plug-in units. It will be used to measure and display the concentration of electrolytic solutions.
  Preliminary tests have been performed and several improvements have been made.
- A high precision dead-time generator with incorporated life-time gate has been developed. The range can be selected in steps of 1 μs from 1 to 10 μs. The dead-time generator itself has no dead-time and the precision of the life-time gate is about 1 ns. A proto type has been completed and is already in use in the Radionuclides Group.
- An electronic sliding pulser for test and calibration of pulse-height ADCs has been developed. The specifications of this pulser are superior to the pulsers available on the market. Two units are in use by now.
- The development of a μ-processor based accordeon system has been started. In this connection the 8086 Intel processor has been studied in detail.
- A word generator intended to test the digital part of data acquisition systems. It generates 24-bit binary words of constant, sliding or statistical contents and with periodic or random occurrence. The unit has been completed and tested and is now in use.
- A digital delay for testing low resolution time coders has been developed. The delay is adjustable in steps of 128 μs up to 8064 μs, with an output time jitter of 1 μs.
- A paper tape reader controller which interfaces a new optical paper tape reader to the control computer of the data transmission system. A proto type has been built but not yet tested.

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** SCK-CEN, Mol, Belgium

9. TECHNICAL DEVELOPMENTS OF VARIOUS DEVICES

A Fission Ionization Chamber Delivering Fast Pulses Containing Timing and Energy Information

C. Budtz-Jørgensen, H.-H. Knitter

In our current programme of measurements of neutron induced fission cross sections of actinide nuclei a fission chamber has been developed intended for medium alpha active (10^6-5.10^7 a/sec) samples. The chamber itself is of the parallel plate type, filled with methane NTP as counter gas. But, contrary to most other designs used for fission cross section measurements, the distance between the electrodes in the present chamber has been chosen longer than the ranges of the fission fragments, see F. 9.1a. Although the charge signal, Fig. 9.1b., induced by the ionization electrons depends on the direction of the fragment track in the chamber, the current pulse, Fig. 9.1c. is proportional to the number of created ion pairs and hence to the kinetic energy of the fission fragments. The risetime of the current pulse is in principle only dependent on the time ≈ 2 ns it takes to stop the fission fragment in the gas. This means that a differentiation of the current pulse results in the extremely sharp pulse as shown in Fig. 9.1d., which will have an area proportional to the energy. To our knowledge it is the first time that a fast pulse is formed from an ionization chamber capable of fast timing in the n.s. range and carrying the energy information with it. Moreover, the short width and the energy proportionality of the pulses ensure that the chamber can operate with samples of high α-activity without having problems to discriminate the fission signals from the alpha pulse pile up.

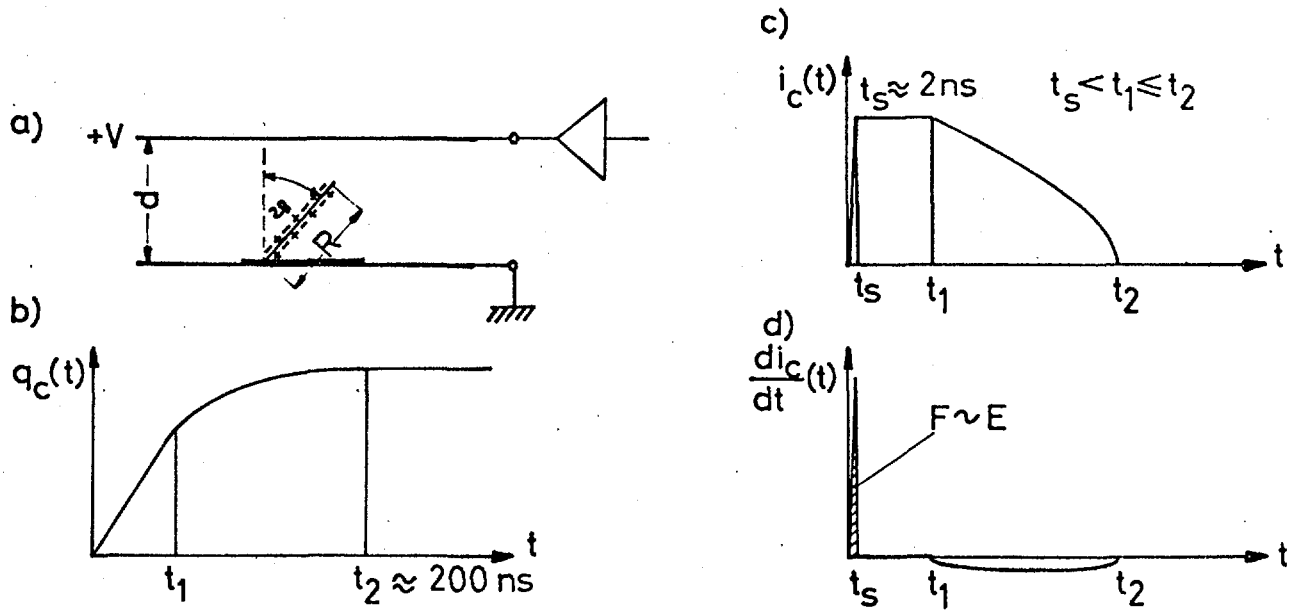

Figure 9.1

With todays commercially available electronics it was possible to obtain a doubly differentiated charge pulse width of 30 n.s., which is sufficiently short to operate the chamber with an alpha-background of 5.10^7 α/s. This is at least a factor 10 more than for the conventional chamber, where the distance between the electrodes is only a fraction of the fission fragment range. The fission fragment energy spectrum was obtained integrating the fast pulse with a fast linear gate opened with 40 n.s. wide gate pulses from a constant fraction discriminator. The chamber is presently operating satisfactorily loaded with 1,5 mg of ^{240}Pu.

Parallel Plate Avalanche Counter Development

A. Wartena, R. Pijpstra

The avalanche counter has proved to be a good neutron flux detector both for fission fragment (^{235}U) and for α-detection (^{10}B). It is now being tested as a general purpose neutron flux detector. It has been reported elsewhere that avalanche counters with low pressure (< 10 Torr) propane and low gas gain have a reduced response for α-particles with respect to fission fragments, a phenomenon which could be interesting for cross section measurements on highly α-active material. Tests were performed with propane (CH-35), household propane, household butane and acetone at pressures between 10 Torr and 4 Torr. The gas gain was varied so that the fission fragments from ^{235}U gave output pulses between 5.10^7 electrons and 3.10^3 electrons. α-pulses corresponding to 500 electrons could be seen. In this range the ratio between the amplitudes from fission fragments and α-pulses was constant so that the reduced response to α-particles could not be confirmed.

Development of Organic Backing Foils

J. Pauwels, J. Van Craen, J. Van Gestel, J. Van Audenhove

Polyvinylchloride and - acetate (VYNS), polyvinyl formal (FORMVAR) and carbon foils were generally used at CBNM as thin backing (10 to 100 $\mu g/cm^2$) for the preparation of actinide deposits. As these foils have relatively poor mechanical-, thermal-, radiation- and/or chemical properties, development work has started for the preparation of higher quality ones. It has been shown that polyimide foils can be prepared at thicknesses interesting for nuclear target preparation via amide-acid spreading and in-situ polycondensation on glass plates vacuum coated with NaCl. First results show that these foils have excellent chemical-, heat-, radiation-, and mechanical resistance, compared with other foils of comparable thickness. Temperature tests indicate that further optimisation of heat treatment must be able to lead to significant increases of their mechanical strength. Further work in order to achieve this is going on. See also list of papers presented at conferences by PAUWELS J. et al. at München (1978).

Development of Spraypainting Technique for the Preparation of Deposits for Nuclear Measurements

J. Pauwels, J. Tjoonk, A. Verbruggen *

A painting method for the preparation of $^{235}U_3O_8$ deposits on metallic backings using an aerograph is developed. The advantages of this method compared to classical brush-painting are that very simple masks can be used to obtain geometrically well defined deposits, practically

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without border effects, and that rather thick deposits can be obtained in one single spraying operation. The adherence of the layer is comparable to that obtained in brush painting.
Targets prepared in a single spraying operation have acceptable homogeneties.
Homogeneity tests were carried out by relative α-counting as illustrated in Fig. 9.2.
The results of these tests are listed in Table 9.1.
See also list of papers presented at conferences by PAUWELS J. et al. at München (1978).

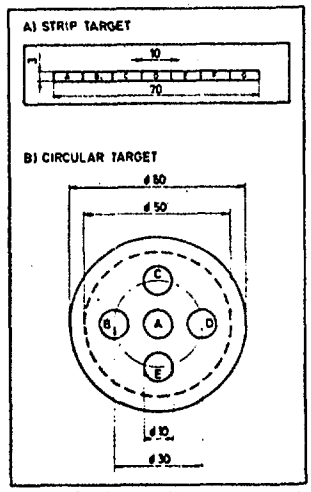

Figure 9.2 Set-up of homogeneity tests by relative α-counting.

Table 9.1 : Results of Homogeneity Tests by Relative α-counting

Measured area	STRIP TARGET ≈ 240 $\mu g/cm^2$/ prepared in one spraying operation	STRIP TARGET ≈ 250 $\mu g/cm^2$/ prepared in three successive spraying operations	CIRCULAR TARGET ≈ 200 $\mu g/cm^2$/ prepared in three successive spraying operations
A	(0.68 ± 0.01)cps	(0.65 ± 0.02)cps	(1.43 ± 0.02)cps
B	(0.67 ± 0.01)cps	(0.64 ± 0.01)cps	(1.53 ± 0.02)cps
C	(0.66 ± 0.01)cps	(0.61 ± 0.03)cps	(1.50 ± 0.02)cps
D	(0.61 ± 0.01)cps	(0.59 ± 0.03)cps	(1.48 ± 0.02)cps
E	(0.57 ± 0.01)cps	(0.59 ± 0.02)cps	(1.50 ± 0.02)cps
F	(0.57 ± 0.01)cps	(0.59 ± 0.02)cps	
G	(0.57 ± 0.01)cps	(0.60 ± 0.02)cps	
Dispersion (1s)	± 7.7 %	± 4.2 %	± 2.6 %
Precision individual meas.(1s)	± 2 %	± 3.5 %	± 1.5 %

Special Vacuum Equipment

W.D. Dobma, I.V. Mitchell, H.L. Eschbach

Two short notes on special vacuum equipment, developed at CBNM, were submitted for publication. The first communication describes a simple method to construct full size models of complex vacuum chambers as they are used in scattering experiments. The second paper gives details on a rotatable support that can be used in ultrahigh vacuum for the evaporation of elemental boron. See DOBMA W.D. et al. in list of publications.

Absolute Ion Counting for Mass Spectrometry Applications

E. Sattler, H. Schipke, L. Traas

The development project for absolute ion counting to measure Pu samples of less than 10^{-8} g is in full progress. The project has three objectives :

Firstly, tests are going on to optimise the electronic instrumentation with respect to double pulse resolution and amplitude overload performance. In order to limit counting errors due to ion coincidences during the dead time of the counting system to less than 10^{-3} for count rates up to 10^{6}/sec, the dead time of the counting chain must be known to within 10^{-9} sec. Electronic tests showed a dead time of the counting system of 40 $\pm$ 0.5 nsec for input pulse amplitudes between 1 and 500 mV or higher. Future tests with well known ion current ratios of 1 : 10 (NBS U-100) will try to confirm the validity of this dead time determination for the whole system.

Secondly, results of measurements with blank filaments show that very small impurities of the Re-filament material lead to considerable measurement error when Uranium samples of nanogram amounts are analyzed. E.g. for a Re-filament normally used for microgram samples an error of 10 % for a $^{235}U/^{238}U$ isotope ratio of 17.3 has been observed on nanogram amounts of NBS U930. As a consequence some development has to be carried out to purify the filament material from impurities (natural Uranium ?). There was no time available to investigate this problem further during the reporting period.

Thirdly, the investigation of the so-called "resin bead" technique in order to minimize the sample amount of Plutonium which has to be transported from the point of inspection to the verification laboratory. The person on this work unfortunately left and work is interrupted.

Evaluation of Accuracy and Reliability of Mass-Spectrometers

E. Sattler, H. Schipke, L. Traas

In order to identify error components limiting the precision of isotope ratio measurements by surface ionisation mass spectrometry, a thorough revision of one of the spectrometers has been effected. As a definite result, the electronic instrumentation has been refined with respect to hardware and operation software. It has been possible to trace the internal precision of an isotopic ratio measurement to the inherent second order effect sources of instability e.g. sudden small changes in source voltage or filament temperature with influence on ionization efficiency, small changes in pump speed, etc. For a single isotope ratio measurement with a value between 1 and 10 an internal standard deviation of $< 5 \times 10^{-5}$ can be reached (total data acquisition time approx. 200 seconds, input currents $\sim 10^{-11}$ Amp. and 10^{-12} Amp. respectively).

The reproducibility of the magnetic field setting after a "peak jump" across several mass units (i.e. a sudden switch of the magnetic field of up to 1000 Gauss back and forth) has been tested on several spectrometers. It turned out that spectrometer magnets of older instruments which were not equipped with a peak jump facility are not necessarily suited to meet the requirements in terms of hysteresis characteristics, which are mandatory to ensure a reproducibility of the magnetic field of better than $\pm$ 0.1 Gauss under all measuring conditions.

Instrumentation for Analytical Measurements

Operation and Upgrading of Mass Spectrometers

A. Loopmans, E. Sattler, H. Schipke, L. Traas

In May 1978 the laboratory received on loan a commercial mass spectrometer, in order to assess its capability of measuring U and Pu under normal operational conditions. The agreed period of availability for this purpose is 3 months.

First results have already shown that due to its very modern design (i.e. fully automated process control and data acquisition together with a multi-filament ion source) the instrument would increase considerably the measurement capability for normal routine analyses. The manufacturer installed the spectrometer just before the summer vacation period. The instrument has been under CBNM-MS examination since September and the machine was purchased at the end of the year.

So far, some interesting performances were achieved :

- Measurement of ^{235}U abundance $\approx$ 0.02 % (isotopic ratio approx. 5000) could be carried out with an internal precision of better than 1 %.
- The minimum sample amount needed for a simple isotope ratio determination was found to be 10 nanogram for Faraday Detector and 1 ng for Electron Multiplier integrated current measurement.

S E R V I C E S

10. PROTECTION AND SECURITY

E. de Ras, D. Gelly, A. De Buck, W. Van Suetendael, H. van de Beek, S. Vandevelde

Legislation

Licences for the new Van de Graaff machine and the upgraded Linac machine were granted. A licence procedure for the new Chemical Building has been started.

Environmental Monitoring

Doses revealed by integrating gamma dosimeters, placed during the year at the boundaries of CBNM, did not exceed 120 mrem including background. Activity releases via the three stacks of the CBNM were permanently monitored. No dispersion of significant activities to the environment could be detected.

Because available neutron dose-rate-meters are too insensitive for environmental measurements, a polyethylene sphere of 10" diameter with TLD integrating detectors was calibrated both at PTB-Braunschweig and at CBNM with monoenergetic neutrons to determine the dose equivalent-neutron energy relationship. Results are shown in Fig. 10.1. Begin 1979, 6 of such detectors will be positioned around the accelerators.

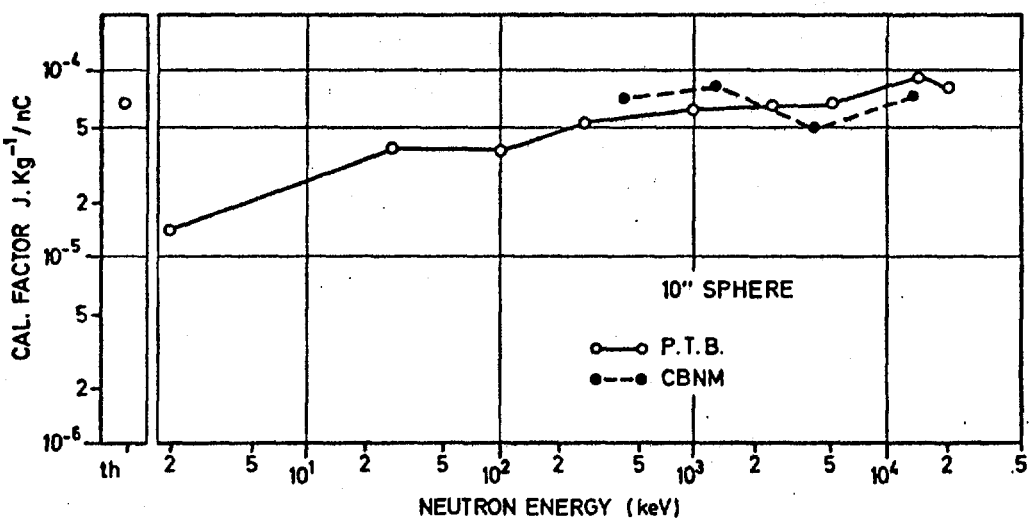

Figure 10.1

Occupational Health Physics and Security

Several minor incidents accompanied with contamination were reported. 35 permissions for work in a controlled area, with special security precautions, were granted. In Table 10.1. information is given on personal and area dosimetry.

Table 10.1 : Personal and area dosimetry

Personal Dosimetry			Area Dosimetry		
N° of persons checked	by means of	maximum dose or activity	controlled objects	by means of	N° of active samples
133	γ-dosimeter total body	1090 mrem/year	laboratory surfaces	25000 smeartests	9
88	neutron dosimeter total body	< 40 mrem/month	laboratory air	4000 airfilters	3
16	γ-dosimeter hands	10 4 rem/year	liquid waste disposal	326 water-samples	1
3	nasal swabs	$6,4.10^{-13}$Ci α-activity $1,2.10^{-11}$Ci β-activity			
100	urine	< 0.1 of max.allowable level			

Discussion on the results mentioned in Table 10.1. :

The range of isotopes sought for in the urine test was as follows;
Unat, U-233, U-235, Pu-238, Pu-239, Pu-240, Pu-241, Am-241, Np-237, Th-232, Cf-252, H-3.
No detectable activity was revealed except in two cases where the measured activity (Pu-239) was less than 10 % of the max. allowable level.

Smear-tests: 7 smears taken proved contamination on H-3 between 1.10^{-3} and 8.10^{-3} $\mu Ci.cm^{-2}$. 1 smear showed activity of 10^{-2} $\mu Ci.cm^{-2}$ ^{103}Pd and 1 smear a β-activity of 6.10^{-3} $\mu Ci.cm^{-2}$. All these contaminations are about 10 times of what can be accepted as maximum permissible surface contamination for active areas and the decontamination of those areas was executed.

Air filters: during short periods of time the measured air activity concentration was in the three cases resp. $3.10^{-12}Ci.m^{-3}$ (α), $1,2\cdot 10^{-14}Ci.m^{3}$ (β) and $1,2\cdot 10^{-11}Ci.m^{-3}$ (α). The first case is 1,5 times and the last case 6 times the maximum permissible air activity concentration for continuous exposure. The β-contamination was far below the maximum permissible level.

Water samples: one sample of waste water was active up to 8.10^{-4} $\mu Ci.ml^{-1}$ due to a tritium contamination. The maximum permissible concentration for the radionuclide tritium in drinking water for the population in the neighbourhood is 3.10^{-3} $\mu Ci.ml^{-1}$.

11. TECHNICAL SERVICES

Buildings and Infrastructure

F. Daems, U. Leurs, M. Van Duffel, R. Verwimp

In the Main building the old 220 V Δ power distribution is being transformed into the European 220/380 V system. An air-conditioning plant has been installed for two Radionuclides labs. The central cooling facility has been completely renewed and adapted to the recent extensions. In connection with the modernisation of the Van de Graaff, the special air-conditioning system of the tower and the magnet hall has been put into operation.
At the Linac two new detector stations at 10 and 30 m have been constructed. Transport facilities have been improved by an hydraulic elevator.
In the Mass spectrometry building the new 10 kV/800 kVA transformer has been installed. In connection with the renewal of the old air-conditioning and ventilation plant, a series of technical studies have been undertaken. Orders have been placed for the extension of the building as well as for the refrigerators, cooling towers, filter boxes, etc.
To group the Technical services and in replacement of the old prefab building, an extension of the workshop/store building has been studied and ordered.
Orders have been placed for the construction of a Chemistry building as well as for the related air-conditioning and emergency power plant.
Construction and technical equipment of the Conference hall has been largely completed.

Mechanical Constructions

K. Geerts, U. Leurs, W. Schreiber, B. Uylenbroek, J. Van Saene

The most important realisations of Design Office are : a homogeneity meter for evaporated layers, a goniometer facility, a vacuum chamber for cold welding, a lift for a rotating Linac target, a break-mechanism for the rabbit project, collimators, etc. Graphical and photographical work for publications was an important task for the design office.
The Workshop made the following constructions : the rotating Linac target with heat exchanger, the terminal of the rabbit system, adaptations of flight paths, vacuum facilities, collimators and experimental set-ups; a series of wobble targets with accessories for the Van de Graaff accelerator, a dust sampler, electro spraying facilities, a goniometer, several special containers, about 14 000 cubes of hard metals.
The glassblower made several pieces of apparatus for the Van de Graaff, Mass Spectrometry and Sample Preparation laboratories.

ANNEX 1 : LIST OF PUBLICATIONS, REPORTS AND CONFERENCE PAPERS

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BAMBYNEK W., MITCHELL I.V., WEIGMANN H., Central Bureau for Nuclear Measurements, Geel, Europhysics News 9 nr. 5, May (1978) 1-4.

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ter MEER P., Camac Real Time Clock CBNM/DE/19/78.

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ANNEX 2 : CINDA ENTRIES LIST

ELEMENT S	A	QUANTITY	TYPE	ENERGY MIN	ENERGY MAX	DOCUMENTATION REF	VOL	PAGE	DATE	LAB	COMMENTS
B	010	N,ALPHA	EXPT-PROG	10+5	22+6	INDC(EUR)12	21	579		GEL	VIESTI+B10(N,AG) REL TO T(P,N)ANG DI
FE	054	TOTAL	EXPT-PROG	20+4	25+6	INDC(EUR)12	20	579		GEL	BRUSEGAN+LINAC TOF MEASM
FE	054	N,GAMMA	EXPT-PROG	50+2	60+5	INDC(EUR)12	20	579		GEL	BRUSEGAN+LINAC TOF MEASM
FE	054	N,ALPHA	EXPT-PROG	60+6	17+7	INDC(EUR)12	5	579		GEL	PAULSEN+REL TO 56FE AND 54 FE(N,P)
FE	054	N,PROTON	EXPT-PROG	60+6	10+7	INDC(EUR)12	5	579		GEL	PAULSEN+REL TO 56FE(N,P)
FE	056	N,GAMMA	EXPT-PROG	50+2	60+5	INDC(EUR)12	20	579		GEL	BRUSEGAN+LINAC TOF MEASM
ZR	091	RESON PARAMS	EXPT-PROG	10+2	20+4	INDC(EUR)12	13	579		GEL	BRUSEGAN+ANALYS IN COLLAB CNEN BOL
NB	093	RESON PARAMS	EXPT-PROG	10+2	70+3	INDC(EUR)12	14	579		GEL	WINTER+WN,WG AND WT
RH	103	TOT INELASTC	EXPT-PROG	20+5	41+6	INDC(EUR)12	6	579		GEL	PAULSEN+REL RH103M ACTIV MEASM
PD	105	RESON PARAMS	EXPT-PROG	10+1	21+3	INDC(EUR)12	15	579		GEL	STAVELOZ+WN,WG AND WT
PD	106	RESON PARAMS	EXPT-PROG	45+1	30+4	INDC(EUR)12	15	579		GEL	STAVELOZ+WT
PD	108	RESON PARAMS	EXPT-PROG	30+1	50+3	INDC(EUR)12	15	579		GEL	STAVELOZ+WN,WG AND WT
PD	110	RESON PARAMS	EXPT-PROG	45+1	30+4	INDC(EUR)12	15	579		GEL	STAVELOZ+WT
IN	115	TOT INELASTC	EXPT-PROG	30+5	41+6	INDC(EUR)12	6	579		GEL	LISKIEN+IN115M ACTIV MEASM
TH	230	N,FISSION	EXPT-PROG	60+5	50+6	INDC(EUR)12	6	579		GEL	BLONS+TOF MEASM REL TO NP237(N,F)
TH	232	RESON PARAMS	EXPT-PROG	20+1	22+3	INDC(EUR)12	16	579		GEL	CORVI+P-WAVE ASSIGNM
PA	231	N,FISSION	EXPT-PROG	50+4	20+6	INDC(EUR)12	17	579		GEL	BUDTZ-JORGENSEN+WORK STARTED
U	235	N,FISSION	EXPT-PROG	25-2	30+4	INDC(EUR)12	23	579		GEL	WAGEMANS+FRAG DET
NP	237	RESON PARAMS	EXPT-PROG	NDG		INDC(EUR)12	17	579		GEL	MEWISSEN+WN,WG AND WT
PU	239	N,FISSION	EXPT-PROG	50+0	30+4	INDC(EUR)12	18	579		GEL	BARTHELEMY+FRAGM AND FISS NEUTR DET
PU	239	NUBAR (NU)	EXPT-PROG	12+1	46+2	INDC(EUR)12	17	579		GEL	WEIGMANN.NU IN DIFF RESON
PU	240	N,FISSION	EXPT-PROG	14+5	98+6	INDC(EUR)12	9	579		GEL	BUDTZ-JORGENSEN+REL TO U235(N,F)
PU	240	FRAG SPECTRA	EXPT-PROG	SPON		INDC(EUR)12	18	579		GEL	WAGEMANS+KIN ENERGY AND MASS DISTR
AM	241	N,FISSION	EXPT-PROG	10+2	53+6	INDC(EUR)12	10	579		GEL	KNITTER+REL TO U235(N,F)
AM	241	RESON PARAMS	EXPT-PROG	12+0	15+1	INDC(EUR)12	11	579		GEL	KNITTER+FISS WIDTHS

ANNEX 3 : LIST OF RESIDENT AND VISITING SCIENTIFIC AND TECHNICAL STAFF

JRC-Geel Staff

Research :

Division Head : Rose B.

Group Leaders : Bambynek W.
Böckhoff K.H.
De Bièvre P.J.
Eschbach H.L.
Horstmann H.
Liskien H.
Meyer H.
Van Audenhove J.

Administration and Infrastructure :

Gubernator K.

Leurs U.

Scientific Secretariat :

Debus G.H.

Health Physics :

de Ras E.

Aerts M.	De Keyser A.	Luyten F.	Rietveld P.	Van Gorp A.
Arnotte F.	De Pooter D.	Lycke W.	Rohr G.	Van Hengel L.
Babeliowski T.	De Roost E.	Maes F.	Ruts H.	Van Heuckelom W.
Barthelemy R.	De Spiegeleer A.	Mailly M.	Ryngaert E.	Vaninbroukx R.
Bastian C.	Deckx J.	Massardier F.	Salomé J.M.	Van Overstraeten J
Beckers F.	Denecke B.	Mast H.	Sattler E.	Van Reeth F.
Berthelot Ch.	Dijckmans B.	Mast J.	Schipke H.	Van Rhee L.
Besenthal R.	Dijckmans M.	Melis F.	Schreiber W.	Van Rillaer C.
Blockx E.	Dobma W.D.	Melis G.	Segers F.	Van Roy P.
Borgers R.	Dufrasne A.	Meloni U.	Sempels J.	Van Saene J.
Bortels G.	Ehrenfreund G.	Mencarelli T.	Shelley R.	Vansant J.
Bouwens J.	Falque P.	Mensch H.	Silvester H.	Van Suetendael W.
Bouwmeester E.	Forni R.	Menu F.	Siméone P.	Van Waelderen H.
Broothaerts J.	Freistedt E.	Merla M.	Smets R.	Verdingh V.
Brusegan A.	Gallet M.	Meynants K.	Stal B.	Verdonck J.
Budtz-Jørgensen C.	Geeraerts R.	Michiels A.	Stüber W.	Verheyen F.
Buyl R.	Geerts K.	Mitchell I.V.	ter Meer P.	Verwimp R.
Carraro G.	Gelly D.	Mouchel D.	Teuling C.	Vogt R.
Celen E.	Grosse G.	Müschenborn G.	Tjoonk J.	Waelbers J.
Cervini C.	Hansen H.	Nagel W.	Traas L.	Waller C.
Ceunen L.	Hendrickx F.	Nerb H.	Triffaux J.P.	Wartena J.
Claes K.	Hens E.	Nijs R.	Uylenbroek B.	Wattecamps E.
Cools H.	Herrmann S.	Noyens J.	Van Baelen F.	Weigmann H.
Cools R.	Hofmans K.	Noyens Jos.	Van Baelen G.	Werz R.
Corvi F.	Idzerda B.	Nylanstedt-Larsen A.	Vanbergen C.	Widera R.
Crametz A.	Janssens V.	Oldenhof W.	Van Bijlen J.	Winter J.
Daems F.	Kennis W.	Parduyns G.	Van Bijlen R.	Wolters W.
Daems L.	Klopf P.	Parengh M.	Van Broekhoven A.	Zehner W.
Daems P.	Knitter H.H.	Paulsen A.	van de Beek H.	
Damen	Le Dez G.	Pauwels J.	Vandevelde S.	
Danckers G.	Le Duigou Y.	Peetermans F.	Van de Vonder R.	
De Backer L.	Leidert W.	Pijpstra R.	van der Veen T.	
De Bolle W.	Leonard J.	Prins H.	Van Duffel E.	
De Buck A.	Loopmans A.	Quik F.	Van Endert J.	
de Jonge S.	Louwerix E.	Reher D.	Van Gestel J.	