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COMMISSION OF THE EUROPEAN COMMUNITIES

Multiannual Programme
of the Joint Research Centre
1980-1983

1981
Annual Status Report

Nuclear measurements

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NUCLEAR MEASUREMENTS
1981

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PROJECTS

- project Measurements of Nuclear Data:
  neutron data
  non neutron nuclear data
- project Nuclear Reference Materials and Techniques:
  nuclear reference materials
  samples and targets for nuclear measurements
  development of reference techniques
  study for the production of enriched actinide isotopes

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1. INTRODUCTION

The JRC programme "Nuclear Measurements" is carried out exclusively by the Geel Establishment (the Central Bureau for Nuclear Measurements, CBNM), and forms by far the largest fraction of the work commitment of that Establishment. It conforms closely with the aim of the Establishment as envisaged in the Treaty establishing the European Atomic Energy Community. In addition it is designed to extend the exploitation of the CBNM's major nuclear measurement facilities, the performances of which have been increased and improved in recent years. These include the two large accelerator installations (the electron accelerator and the Van de Graaff generator) and the mass spectrometers.
The Nuclear Measurements programme is divided into two main projects, Nuclear Data on the one hand and Nuclear Reference Materials and Techniques on the other.
In the former the JRC actions form part of world-wide sets of actions to establish reliable, and in many cases very precise, figures for important nuclear parameters - e.g. neutron interaction cross-sections, radionuclide half lives - needed for the development and exploitation of nuclear energy for peaceful purposes. In this work the CBNM pays particular attention to the specific needs of the Community and to complement similar actions undertaken in the laboratories belonging to the Member States.
Concerning Nuclear Reference Materials and Techniques the actions are to provide materials to which analytical and other measurements carried out in the nuclear industry or by the nuclear community can be referred. One or more of the properties of the materials must therefore be well characterized and eventually certified and in order to carry this out effectively the important techniques used for characterization must be continuously examined and if possible improved.
The basic aim of the Nuclear Measurement programme is therefore to develop

nuclear metrology with special orientation towards satisfying the demands for basic nuclear data and for materials and methods of reference.

2. RESULTS

Measurements of Nuclear Data

This project is divided into two parts, one concerned with data pertinent for neutron induced reactions and the other with so-called non-neutron nuclear data, i.e. mainly data on radionuclide decay.

Neutron Data

The main objective is to measure accurate differential cross section data needed for fast reactor design and safety, long term irradiation effects on fuel and structural materials, shielding, nuclear waste treatment and storage, safeguards and also for fusion reactors and technology. A prime objective for CBNM remains the improvement of the knowledge of standard cross sections required for these research areas. Also the understanding of underlying physics mainly concerning neutron induced reactions necessary for current and future nuclear power technology is an important objective.
In 1981 substantial contributions have been made to the knowledge of the fission and capture cross sections and alpha of ^{235}U in the keV neutron region (Figure 1).
Whereas good agreement was found between the results and the previous figures for the ^{235}U fission cross section (with a disparity on average of 1.2 %), for the absorption cross section, the Geel results showed data on average 14 % lower than these of Gwin et al. (Nucl. Sci. Eng. 59, 79, 1976). A discrepancy of this magnitude and with such a systematic character can only be due to an error of normalisation of one of the two sets of measurements.
The fission cross section was measured in detail for the Pu-isotopes ^{238}Pu, ^{242}Pu and ^{244}Pu including the sub-

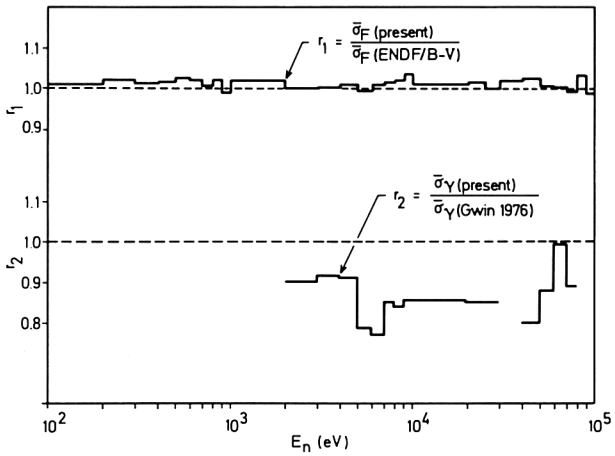


Figure 1. Fission cross section of ^{235}U. Comparison of present results with previously known values.

threshold region. Neutron capture cross sections for ^{96}Zr and ^{54}Fe have been measured with high neutron energy resolution and, concerning the data requested for the fission products, special attention was given to the study of the resonance parameters of the Pd isotopes as well as to their average capture cross sections. To have a better understanding of neutron spectra emitted from nuclear waste a comprehensive study of the neutron energy spectra from (α,n)-reactions in light elements was performed. In collaboration with KFA Jülich further measurements were performed to solve discrepancies for the cross sections for tritium breeding from ^{7}Li, of importance to fusion technology (Figure 2). Hitherto information on neutron cross sections for ^{7}Li have been based on the ENDF/B-IV evaluation. Later work done elsewhere indicates that some of the accepted values of these cross sections may be too high. Our results so far confirm the general trend of the Los Alamos evaluation showing lower neutron cross sections than previously accepted. An important contribution has been made to the knowledge of the angular distributions of the $^{6}Li(n,t)^{4}He$ reaction, one of the basic neutron standard cross sections.
In conjunction with the measurements on neutron data, good progress has been made towards the fundamental

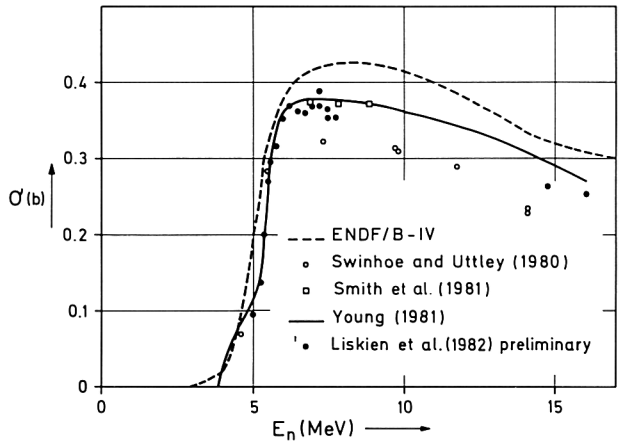


Figure 2. $^{7}Li(n,t)$ results measured by activation together with other results not yet available at the time of the ENDF/B-IV evaluation (dashed line). The other curve (full line) is a recent LA-evaluation.

understanding of the neutron capture process and also to the improvement of knowledge on how the kinetic energy of fission fragments depends on the initial excitation of the compound system.
The 150 MeV linear electron accelerator and the 7 MV Van de Graaff accelerator as well as the IBM 370/138 computer have been used effectively for the execution of this programme.
Good progress has been made towards the acquisition of the necessary hardware to improve pulse compression (Figure 3) on both accelerators and the modifications to the accelerators to obtain more energy in a pulse (or the same energy in a shorter pulse) are expected to be completed in 1982.

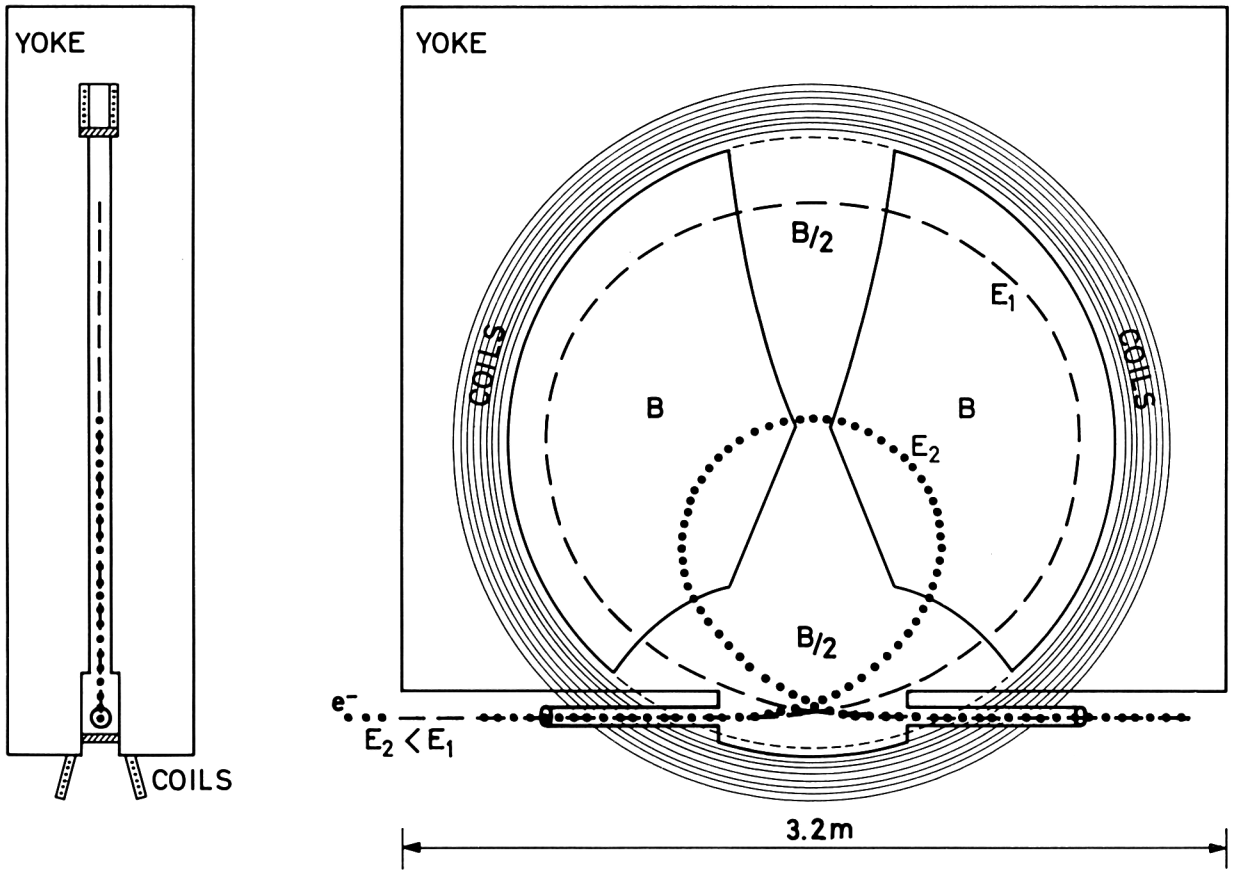
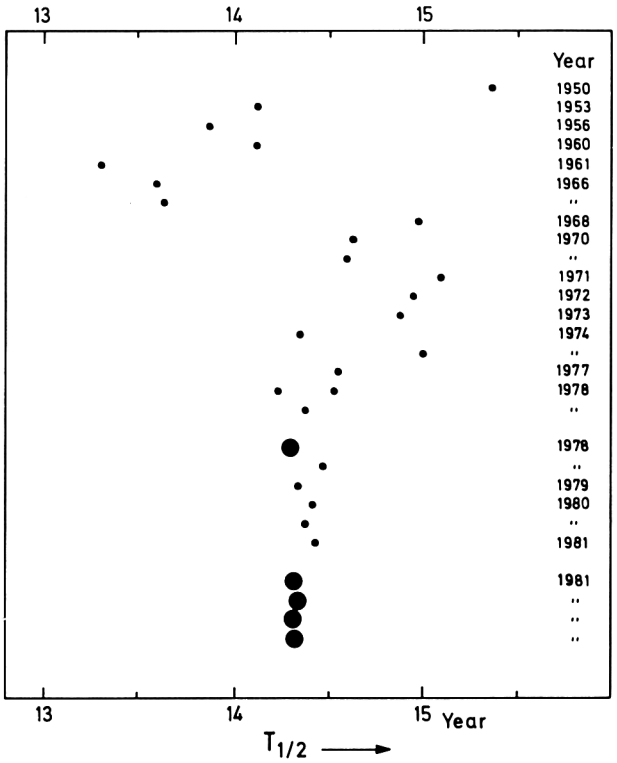


Figure 3. Linear Accelerator: pulse compression magnet.
— — Trajectory of first electrons (E_1) of the pulse.
••••• Trajectory of the last electrons (E_2)

Non neutron nuclear data

In this section the objective is to meet the needs for information on the decay properties of radionuclides by experimental determination of the relevant data (including the development of techniques to improve accuracy) and by detailed evaluations of information already available.
During 1981 important progress was made on a measurement of the half life value for ^{241}Pu, for which a long standing discrepancy exist. Since the early 1950s the estimates of the half life of ^{241}Pu have followed an unusual pattern with a scatter of results in the area of 13-14 years and another scatter of results somewhat higher at about 15 years. In order to eliminate some of the problems and sources of errors between laboratories, the changing isotopic composition as a function of time was analysed for a highly enriched ^{241}Pu sample. This yielded a half life value of 14.33 ± 0.02 y (Figure 4). Two samples of plutonium from UKAEA Harwell were also measured. The resulting values for the half life are 14.33 y and 14.35 y with an uncertainty of 0.01 y. Results using different methods at Geel show a consistent figure of between 14.32 and 14.35 with an uncertainty of between 0.01 and 0.03. Despite these seemingly good results, there are still inconsistencies between results from various laboratories and different methods used.
Agreement has been found in collaboration with CEN Grenoble for the half life of $^{93}Nb^{m}$, which in spite of its importance for fast neutron dosimetry was discrepant by a factor of two some years ago.
The gamma-ray emission probabilities in the decay of the daughters of ^{235}U and ^{231}Th and ^{232}U and ^{228}Th, important for accurate mass determinations of uranium samples by α counting and for the determination of natural Th in environmental samples, were determined. The emission probability of the 5.449 MeV α particles in the decay of ^{224}Ra to the 241 keV level in ^{220}Ra was also measured, this result being significant for geochronological studies. Considerable progress was also made in the development of techniques to prepare radioactive sources of low self absorption suitable for radionuclide decay studies. Of special mention is the preparation and characterization of thin $^{93}Nb^{m}$ sources via ion beam sputtering.



Remark: All ingrowth estimates calculated with $T_{1/2}$ ^{241}Am = 432.6 yr.

Figure 4. Half-life of ^{241}Pu
• Measurements of different laboratories.
● Measurements of CBNM.

Nuclear Reference Materials and Techniques

This project is divided into four parts.

Nuclear Reference Materials

There exists a need for certain actinide reference materials within the European Community. This need, and the priorities for the various materials, were established a few years ago and CBNM is now engaged in

trying to satisfy the need. Three types of such material, necessary for the calibration of analytical methods, can be identified: those for destructive chemical analysis, those for destructive isotope anallysis by mass spectrometry and those for non destructive isotope analysis by measuring the emitted γ radiations. During 1981 a milestone was reached by finalizing the procedure for the issue of the first "EC-Certificate" for a nuclear reference material viz for a plutonium metal to be used for calibration of chemical analyses (Figure 5). This signified that in

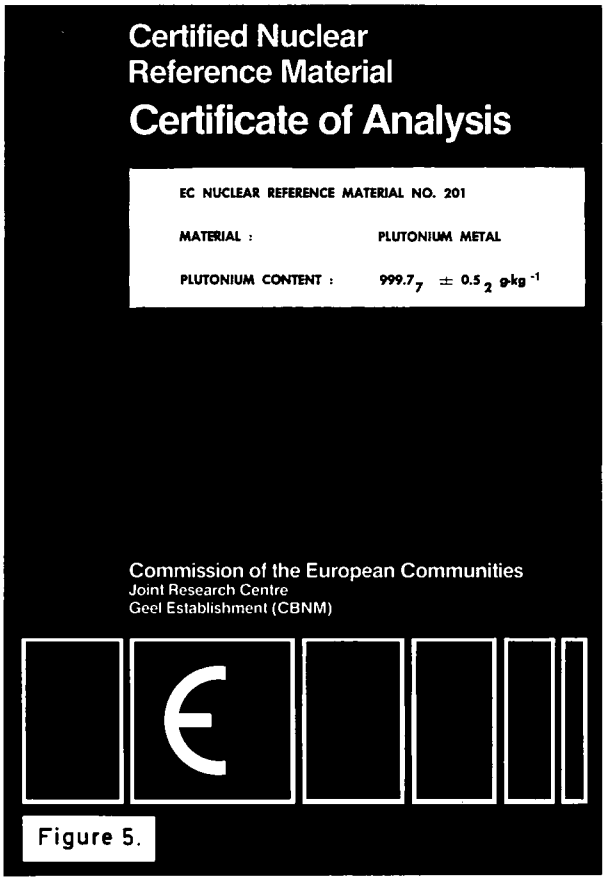


Figure 5.

the case of this material, all the necessary characterizations had been carried out and that there had been sufficient verification for the stock of samples to be accepted as reference material on a Community basis. The corresponding project for uranium metal has also passed through this procedure and for the case UO_2 reference material the necessary experimental and evaluation work required for certification has been finished. In the area of isotopic references, the intercomparison of UF_6 of French, UK and CBNM origin was completed, showing reasonable compatibility. Progress has been made on the preparation of synthetic mixtures of enriched uranium and plutonium isotopes; these mixtures are intended to serve as primary reference materials of high accuracy for the calibration of other isotopic reference materials. The preparation of uranium oxide reference materials for non destructive analysis by measuring the emitted γ-rays made substantial progress in 1981. This project is performed in collaboration with the National Bureau of Standards, USA (Figure 6).

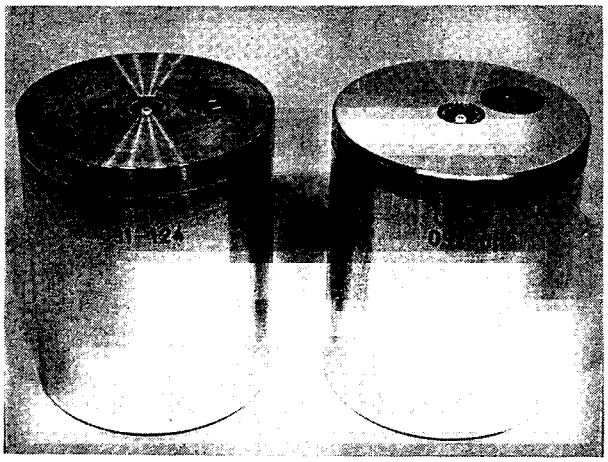


Figure 6. Uranium oxide canned samples for NDA measurements.

Samples and Targets for Nuclear Measurements

More than 200 research laboratories and university departments have made requests to CBNM to provide very specialized samples and targets and in 1981 nearly 650 items were delivered. Such samples and targets were used for various nuclear measurements such as targets used in accelerating machines or in-pile dosimeters. A variety of uranium, plutonium and thorium compounds were provided in isotopically pure forms on different mounts or in different special containers as required.

Other materials included, for example, a sample of NaCl in an iron can. In this particular case the salt was made from the unusual isotope of sodium ^{22}Na rather than ^{23}Na and was requested by the University of Diepenbeek for measurements of positron annihilation in irradiated samples.

Reference Techniques

The object of this project point is to develop and improve analytical methods and characterization techniques used for the production of reference materials and special samples and targets. During 1981 a precise gravimetric potentiometric titration method for uranium determinations was improved.
Photon activation analysis, particle induced X-ray emission and Rutherford backscattering methods were further developed and applied to some interesting fields, e.g. surface analyses. Some attention was focussed on the further instrumental development in mass spectrometry, especially in view of increasing the accuracy in measuring low abundant isotopes. Another development concerned the quality testing of stretched polypropylene as potential substrate material for nuclear targets.

Study of the Production of Enriched Actinide Isotopes

The CBNM has been charged to investigate the future European Community needs for actinide isotopes and the ways they may be obtained. The list of isotopes, has now been evaluated according to materials required in such high purity that electro-magnetically separation techniques are necessary.
Among the high priority materials ^{244}Pu is of particular interest for measurement purposes as it is not present naturally or produced in significant quantity during normal nuclear fuel consumption. By introducing a known amount of this isotope into material containing other plutonium isotopes, the other material can be measured against this ^{244}Pu reference in a mass spectrometer.
In 1981 the project to supply small scale production of this isotope entered the phase of ordering a design study for a prototype electromagnetic actinide-isotope separator.
Twenty ^{242}Pu samples were irradiated in the BR2 reactor at SCK/CEN (Mol) and at the High Flux Reactor at JRC Petten. A preliminary comparison of yields indicates a systematic overestimation of about 30 % in the amount of ^{244}Pu produced, such a material would be used as a feed stock for an electro-magnetic separator.

3. CONCLUSIONS

Substantial contributions to the knowledge of neutron cross sections have continued to be made in the course of 1981. These relate both to data for nuclear reactors particularly fast breeders, and also to the fusion programme.
Work has continued on the half life of ^{241}Pu for which an intriguing long standing discrepancy exists, though the matter has yet to be fully resolved. Work on improving the two main accelerators of the establishment has proceeded with the aim of shortening the pulse length and therefore increasing the accuracy of measurements.
The first nuclear reference material bearing an "EC Certificate" of its chemical quality represents an important milestone in European collaboration in this field.
Provision of samples and targets continues to be an important service to customers with 647 items delivered during the year.
A design study has been ordered for a prototype electro-magnetic actinide isotope separator with the production of ^{244}Pu in mind.
The mutual collaboration with external organizations has continued. Such collaborations are essential to ensure the full exploitation of the Linac accelerator in the area of nuclear data measurements and also in the characterization and verification work for nuclear reference materials.

The Nuclear Measurements Programme is proceeding at about the expected pace although some delays have arisen due to some hot laboratory facilities having to be taken out of use while infrastructure improvements are in progress.

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