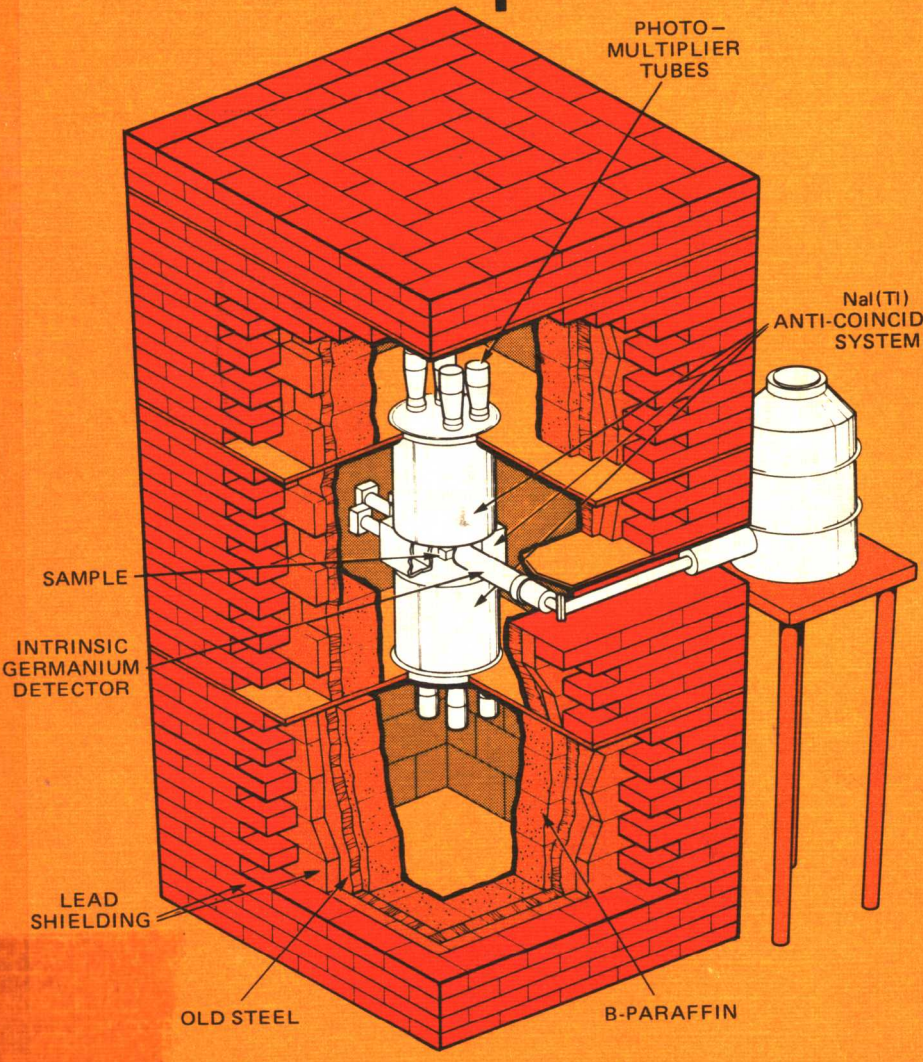
# Methods of Low-Level Counting and Spectrometry

PROCEEDINGS OF A SYMPOSIUM BERLIN (WEST) 6–10 APRIL 1981




INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1981

PROCEEDINGS SERIES

# METHODS OF LOW-LEVEL COUNTING AND SPECTROMETRY

PROCEEDINGS OF AN INTERNATIONAL SYMPOSIUM ON METHODS OF LOW-LEVEL COUNTING AND SPECTROMETRY
ORGANIZED BY THE
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# FOREWORD

Low-level measurements of environmental radioactivity are of increasing potential use for environmental protection and for studying processes in nature. Better instruments and more sensitive methods developed for identifying and determining radionuclides in trace concentration help in understanding physical, chemical and biological processes in the geo-, hydro- and biospheres. Improved dating methods are of importance not only for archaeology, but also in the disposal of radioactive waste.

The International Symposium on Methods of Low-Level Counting and Spectrometry was held by the International Atomic Energy Agency in Berlin (West) from 6 to 10 April 1981. The Symposium was attended by about 200 participants from 34 countries and two international organizations. The forty papers presented are published in the present volume together with a brief summary of the Symposium. Among the many topics covered at the Symposium were: gamma-ray spectrometry, low-level alpha and beta particle counting, tritium enrichment and measurement, radiocarbon counting and new methods of low-level measurements including accelerator-based techniques.

The papers published in this volume show the considerable progress achieved in this subject since 1967 when the last IAEA Symposium on similar topics was organized in Monaco, and give an up-to-date review of the progress and development in this field.

The Agency wishes to thank the many staff of the Hahn-Meitner-Institut für Kernforschung for their hospitality and co-operation in running the Symposium, which helped so much towards its success.

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# Session 1

# GAMMA SPECTROMETRY, INSTRUMENTATION AND APPLICATIONS

**Chairman:** **H. Moser**
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# A MULTI-CRYSTAL SPECTROMETER FOR GAMMA-RAY COINCIDENCE MEASUREMENTS

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## Abstract

A MULTI-CRYSTAL SPECTROMETER FOR GAMMA-RAY COINCIDENCE MEASUREMENTS.

A multiple γ-ray spectrometer has been constructed using six thallium activated sodium-iodide detectors. Two of the crystals are 170 mm in diameter and 50 mm thick, the other four are 75 mm in diameter and 75 mm thick. They surround a sample volume of about 0.5 ltr. The output of each scintillator is digitized and a microprocessor used to select events of particular characteristics. The apparatus is operated in a shielded enclosure in the Holborn underground laboratory in London. The performance of the spectrometer has been investigated using positron sources of $^{22}$Na and $^{26}$Al. It was then used to measure the cosmogenically produced activities of these isotopes in a 335-g fragment of the Mayo Belwa meteorite. It is shown that the problems due to self-absorption factors and the variation of efficiency with position in the sample volume can be solved by recording both twofold and threefold coincident spectra. It is then practicable to determine the ratio of $^{22}$Na to $^{26}$Al in the meteorite to an accuracy of about 3% and their absolute activities to about 6%. It is suggested that this type of apparatus could be useful for measuring other multiple γ-emitting isotopes in extended specimens.

## Introduction

During the last twenty years there has been an increasing interest in studying meteorites in order to obtain clues both to the early history of the solar system and to the radiation field through which they have passed. Measurement of cosmogenic isotopes, whether stable or not, in meteorites therefore needs to be carried out for as many samples, and as accurately, as possible. Special difficulties arise with meteorites in that it is usually necessary to use non-destructive methods or else to study very small amounts of material which have been removed from them and may not be representative samples. The only isotopes which can thus be studied easily are those such as $^{26}$Al which emit two or more γ-rays simultaneously, as the activities to be measured, $\sim 1\ s^{-1}\ kg^{-1}$, make coincidence methods essential.

It was suggested by Fireman [1] that it would be particularly useful to make precise measurements of the ratio of the positron emitters $^{22}$Na to $^{26}$Al since the very much shorter



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half life of the former, 2.6 a as against $7 \times 10^5$ a , means that its activity should depend on the meteorite's trajectory and the phase of the sun's 11 year cycle in the period before arrival at the earth. Fireman therefore proposed that all recently fallen meteorites should be examined and the ratio plotted against the solar activity. Since the total variation is not expected to be more than about 30%, the requirement is to measure the activities within a few per cent. The original purpose of this investigation was to find out whether this degree of accuracy was feasible using extended specimens. Some initial experiments, using 3-fold coincidences (Barton and Wright, [2]), indicated that the geometry of the detector arrangement could greatly affect the results and that a more detailed investigation would be necessary. The resulting apparatus, though designed primarily for these studies of meteorites, may well have applications in other fields requiring the measurement of very low activities of multiple $\gamma$-emitting isotopes.

## Apparatus

The ideal arrangement for measuring multiple $\gamma$-emitting isotopes would be to surround the sample volume completely with a large number of detectors, each of 100% efficiency. Arrangements of that sort have been developed for high energy physics and are known as "crystal ball" detectors because of their shape; their cost precludes their use for low background counting. The aim of the present investigation was to find out what could be achieved using a relatively simple system. As far as possible it used equipment already available in the laboratory and certainly it does not attain the best possible performance.

The geometry of the detecting system (figure 1) was determined by the obvious advantages of choosing a symmetrical arrangement and the fortuitous availability of the two large thallium activated sodium iodide crystals, each of diameter 170 mm and thickness 50 mm. The resolution and the background of these two crystals were rather poor. The other four detectors, of diameter 75 mm and thickness 75 mm, were of moderately good quality. (The specification of quality of sodium iodide detectors is still unsatisfactory, and the designation "selected for low background counting" does not indicate the actual level of contamination by potassium, uranium and thorium.) The detectors were used in their normal encapsulated form, which meant that there were significant gaps between them and that, even for a point source at the centre, the geometric coverage was less than $4\pi$. The useable sample volume was roughly cubic in shape and of volume $\sim 0.5$ ℓ.

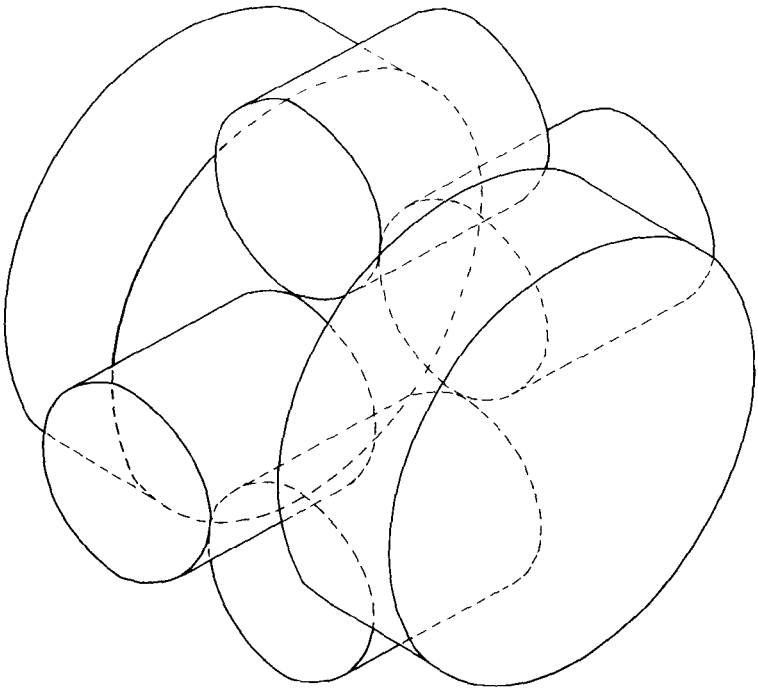

*FIG. 1. Six-crystal gamma-spectrometer.*
*4 crystals: 7.5 cm dia. × 7.5 cm*
*2 crystals: 17 cm dia. × 5.0 cm*
*Thallium-activated sodium iodide.*

The output from each detector was divided and fed both to a majority logic unit and to one input of a standard CAMAC multiple digitiser (Le Croy 2259A) which encoded its value as a 10-bit word. The logic unit could be switched to select all events, 2-fold or 3-fold coincidences; its output was used to gate the multiple analysers. The CAMAC unit "Look-at-me" signal was used to generate an interrupt on a 990/100 microprocessor board, to which the six pulse heights were then transferred. The subsequent classification and accumulation of events was under program control. For studying positron emitters, the pulse height distribution in each crystal were simultaneously recorded, both separately and summed for all the detectors, as follows:

1. All pulses
2. Pulses in coincidence with one 0.51 MeV γ-ray in any other crystal

3. Pulses in coincidence with two 0.51 MeV γ-rays in any other crystals.

Many other modes of operation are possible, such as recording X-Y distributions, but relatively little work has so far been done with them.

The detector assembly was surrounded by shielding, formed by steel sheets of total thickness 15 cm top and bottom and lead walls of thickness 10 cm on all sides. It was operated in the Holborn underground laboratory in London under a thickness of about 25 metres of clay (60 m.w.e.).[1] A large liquid scintillation counter above the shielding was operated in anticoincidence to further reduce the contribution to the background from cosmic rays.

The channel width for the multi-channel analysers was arranged to be 10 keV by appropriate adjustment of the high voltages applied to each photomultiplier. Gating pedestal effects were cancelled for each detector separately in the analysis program. The whole system has been re-calibrated with a $^{26}Al$ source at monthly intervals; any adjustment required has never been more than one channel. The system was linear over the range 0.5 - 2.0 MeV but showed some deviation at higher energies.

## Detection efficiency

A series of tests have been carried out to investigate the characteristics of this system. In all cases the results apply to the efficiency for detection in the photo-peak and the background has been subtracted. It is assumed that each disintegration of $^{22}Na$ yields a 1.27 MeV γ-ray and that 0.905 of the disintegrations are accompanied by a pair of 0.51 MeV γ-rays provided the source is surrounded by $\sim$ 0.1 g cm$^{-2}$ of material to stop the positrons. The measured detection efficiencies are given in Table I. The results with the source at the bottom of the sample volume give some idea of how the detection efficiencies vary with position.

An attempt has been made to compare these results with what might be expected theoretically. The geometry is too complicated for these theoretical estimates to be very reliable, but they appear to be compatible with the data using reasonable

[1] m.w.e. = metres water equivalent

TABLE I. ABSOLUTE DETECTION EFFICIENCIES

| | Source at centre | Source at bottom | Ratio |
|---|---|---|---|
| Efficiency for detecting 0.51 MeV γ-ray in peak | 0.269 ± .002 | 0.284 ± .002 | 1.05 ± .01 |
| Efficiency for detecting 1.27 MeV γ-ray in peak | 0.099 ± .001 | 0.111 ± .001 | 1.12 ± .02 |
| Efficiency for detecting two coincident 0.51 MeV γ-rays but not 1.27 MeV γ-ray | 0.123 ± .001 | 0.102 ± .001 | 0.83 ± .01 |
| Efficiency for detecting only one 0.51 MeV γ-ray in coincidence with 1.27 MeV γ-ray | 0.0198 ± .0002 | 0.0252 ± .0002 | 1.27 ± .02 |
| Efficiency for detecting both 0.51 MeV γ-rays in coincidence with 1.27 MeV γ-ray | 0.00781 ± .00007 | 0.00648 ± .00007 | 0.83 ± .01 |

choices for the detector efficiencies and other parameters. The calculations are in any case quite complicated because they must allow for the rejection of events in which the 1.27 MeV γ-ray interacts in the same detector as one of the pair of oppositely directed 0.51 MeV γ-rays. The variation of coincidence efficiency with source position can be understood, at least qualitatively, by similar considerations.

Similar results have been obtained using a small $^{26}Al$ source. Its absolute intensity was not as precisely known as that of the $^{22}Na$ one, so the results were normalised to those of the latter. Further tests were carried out using absorbers around the sources and with inherently thick sources. These experiments confirmed that the coincident rates were reduced by the products of the corresponding γ-ray absorption factors.

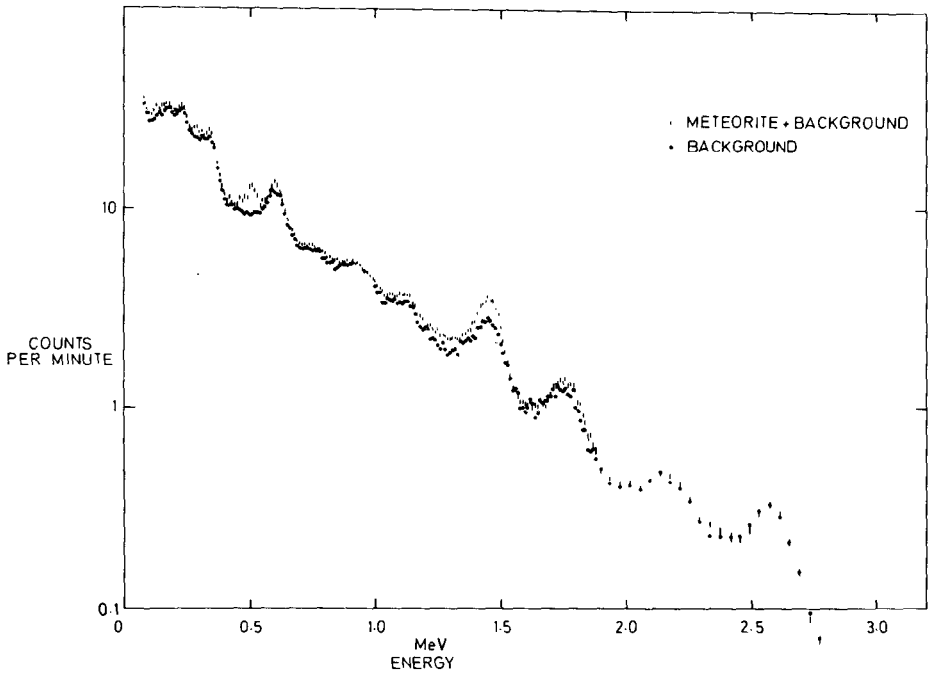


*FIG. 2. Total rates.*

## Results

The meteorite sample available for this investigation was a 335 g irregularly shaped fragment of Mayo Belwa, which fell on 3rd August, 1974. Figures 2, 3 and 4 show the spectra of all pulses and those in coincidence with either one or two 0.51 MeV γ-rays.

The counting rates in the 0.51, 1.27 and 1.81 MeV peaks for the sample and the background are collected in Table II. It is evident that the total rates for the sample are not sufficiently large compared with the background to be reliable. The measurement of the difference is limited both by the statistics and by uncertainties in the stability of the background counting rate. Part of the background is due to contamination of the detectors and surrounding material, which should be constant, but another part, due to radon and its products, is certainly variable. The extent of this variation is uncertain and there is a further correction needed for the absorption of the background by the sample. The effect of

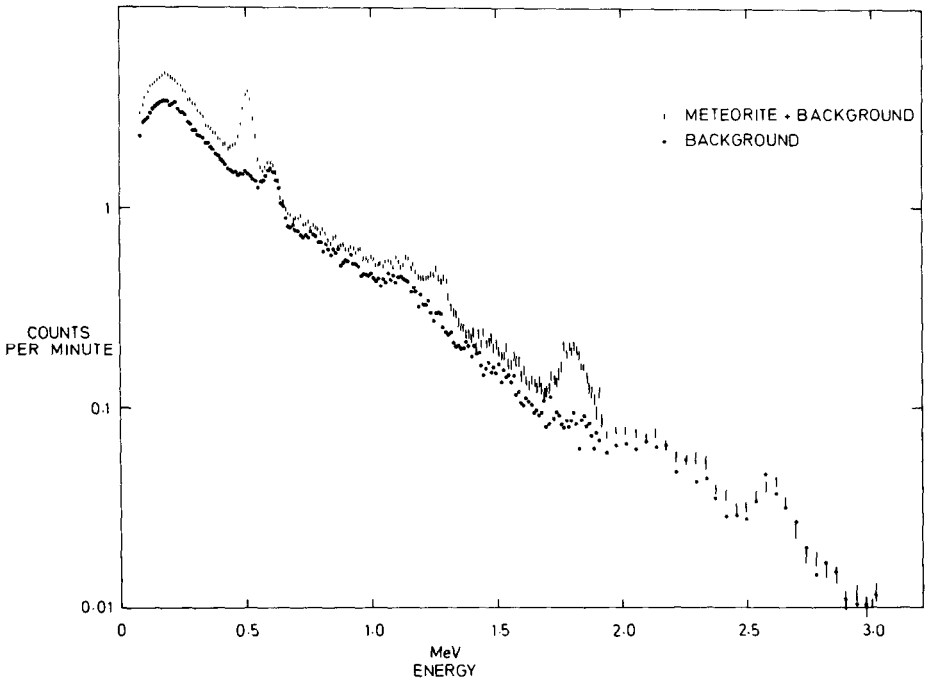


*FIG. 3. Rate in coincidence with one 0.51-MeV γ-ray.*

background variation or absorption on the 2-fold coincidences rates should be quite small and negligible for the 3-fold ones. For the latter case the limitation should be purely the statistical one; even for the 9-day run used to obtain this data the intensity of the $^{26}$Al peak is only determined to ∿ 5%.

The activities of $^{22}$Na and $^{26}$Al in the sample can be deduced from these data and those for the individual test sources. A number of problems must be considered during the analysis:

1. There may be interference from other activities. For most meteorites there are no significant contributions from thorium or uranium and the spectra for Mayo Belwa give no indication of any other isotopes that would contribute to the coincident events.
2. The two positron emitting isotopes contribute to both high energy peaks due to Compton scattering, escape peak and sum peak effects.

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