NEUTRON AND GAMMA-RAY SPECTROSCOPY AND ACTIVATION ANALYSIS

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Massachusetts Institute of Technology
77 Massachusetts Avenue
Cambridge, Massachusetts

FINAL REPORT
MITNE-70

Contract No. AF19(604)-7492
Project 5620
Task 562002

Period Covered
January 1, 1961 through January 1, 1966

February, 1966

Air Force Cambridge Research Laboratories
Office of Aerospace Research
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ABSTRACT

This report reviews the principal results of research under Air Force contract number AF19(604)-7492 carried out between January 1, 1961 to January 1, 1966. Some of the work was a continuation of research begun under Air Force contract AF19(604)-3461.

The preparation of lithium drifted solid state γ-ray detectors having active volumes between 0.5 cm$^3$ and 17 cm$^3$ is described. The energy resolution of these detectors is 0.5% FWHM for Cs$^{137}$ γ rays and 0.12% for the 7 MeV capture γ rays in iron. The experimental facility at the MIT reactor for using these detectors to study capture γ rays is described. The Ge(Li) detector is used in conjunction with two large NaI crystals. This system of detectors is operated as a triple coincidence pair spectrometer to study γ rays with energy above 2 MeV and in anticoincidence for Compton reduction when studying the spectrum below 2 MeV. Preliminary results for the spectrum of Fe and Mn are presented.

A six meter bent crystal spectrometer and a pair scintillation spectrometer have been used to study the capture γ ray spectrum of Sc$^{45}$, Ir$^{191}$, Ir$^{193}$, Rh$^{103}$, Dy$^{161}$, Dy$^{164}$, and Ho$^{165}$. The results of these measurements have been used to help construct proposed energy level schemes in Sc$^{46}$, Dy$^{165}$, and Ho$^{166}$. Some additions to present level schemes of Rh$^{104}$ and Dy$^{162}$ are suggested.

A technique for the nondestructive analysis of spent reactor fuel using Ge(Li) γ ray spectrometers is described. The fuel elements are scanned and the concentrations of three fission products, Cs$^{137}$, Cs$^{134}$, and Zr$^{95}$ are determined from the intensity of emitted γ rays. The ratios of the intensities of these fission products are used to determine burnup, flux, and irradiation time of fuel. A comparison of the results of this method with independently determined results showed agreement within $\pm$ 10% in all cases for the MIT reactor fuel studied.

An improved method for determining fast neutron spectra is described. As an example of the application of this method, measurements
of the fast flux of the MIT reactor medical therapy beam are reported. The method uses threshold foil data and computes the spectrum using a weighted orthonormal and a weighted orthonormal polynomial method. A code is also developed for calculating fast neutron dose in hydrogenous material.

A technique using prompt activation analysis for the analysis of boron and lithium is described. Samples are internally mounted inside a gas flow proportional counter. The counter is then placed in a thermal neutron flux of $10^8$ neutrons/cm$^2$ sec and $\alpha$ rays from the $(n,\alpha)$ reaction are detected. The method is capable of determining boron and lithium in the levels of 1 part per million.

Fast neutron activation analysis using the $(n,2n)$ reaction has been investigated. Preliminary work indicates that about $10^{-6}$ to $10^{-7}$ gms of N, F, K, Zn, Ga, Br, Mo, and Ag can be detected using 2 NaI crystals in coincidence to detect annihilation radiation from the induced positron activities.
I. INTRODUCTION

The objectives of this contract were to carry out basic research in the areas of neutron spectroscopy and dosimetry, gamma-ray spectroscopy and dosimetry, and activation analysis techniques. There can be no doubt of the need for better ways of measuring gamma rays and neutron radiation fields. Many areas of investigation find a lack of accurate knowledge of these radiation fields a limitation in their work. This is particularly true of studies of radiation damage to solids, where one of the difficult problems is knowing just how much of what kind of radiation has been received. Improvements in these areas will also be of benefit to such fields as shielding and radiation safety, space exploration, and a host of other areas where either natural or artificial radiation is a problem.

The area of activation analysis has long been recognized as a very sensitive technique for assaying for certain elements. In this contract our efforts have been directed toward the development of new activation analysis techniques. These included prompt activation analysis by the \((n,\alpha)\) reaction for boron and lithium, prompt analysis by \((n,\gamma)\) reaction, and analysis using fast neutrons to produce \((n,2n)\), \((n,p)\), etc. reactions.

The work was all done at the reactor of the M. I. T. Nuclear Engineering Department. This is well suited for such studies because of the large variety of irradiation facilities available. There is also available several bent quartz crystal spectrometers, a fast neutron chopper, a 4096 channel analyzer, and a wide variety of other instruments. Associated with the reactor is a Cockcroft-Walton neutron generator capable of producing 14-MeV neutrons, either pulsed or steady state. In addition to the equipment there is a staff with a wide range of experience in this general field.
The work carried out under this contract is a continuation of work begun under a previous Air Force contract AF19(604)-3461. Many of the equipment items used in this work were provided under that contract.

Most of the research has been carried out by students of the M. I. T. Nuclear Engineering Department as MS or Ph. D. theses under the direction of the principal investigators. Many of these theses have been issued as Air Force reports. A complete list of them appears as Appendix 1.

Chapter II deals with work in γ-ray spectroscopy. During the early part of the contract this work was done mostly with the bent crystal spectrometers and the triple coincidence scintillation spectrometer which were used in capture γ-ray studies. In the last two years, however, all the work in γ-ray spectroscopy has been in the development and application of lithium drifted Ge(Li) solid state detectors. Chapter II includes a description of the fabrication techniques used and the application of these detectors to the study of capture γ rays and spent reactor fuel.

In Chapter III the work on problems of neutron spectroscopy and dosimetry are described. This work resulted in an improved method for measuring fast neutron spectra through the use of foils. Chapter IV summarizes the work done in activation analysis.
II. GAMMA-RAY SPECTROSCOPY

A. Lithium Drifted Ge(Li) Detectors (J. A. Sovka, V. Orphan, B. Hites)

1. Description of Ge(Li) Detectors

In recent years, semiconductors have been frequently used instead of detecting systems such as scintillation crystals and gas counters. The primary reason for this trend is the fact that semiconductors have an improved efficiency for converting the particle energy into an electrical signal. The absorption of a given amount of energy results in a charge about 10 times larger in such a detector than in a gas counter. In scintillators, inefficiencies in converting light to an electrical pulse result in a signal only 1/100 of that from a semiconductor. Since the energy resolution of a system is dependent upon the ratio of signal-to-noise, the semiconductor detector can yield a resolution about a factor of 10 better than NaI and about a factor of 3 better than gas counters. To date, only silicon and germanium have been applied with reasonable success to nuclear radiation spectroscopy. Some of their relevant properties are given in Table 1.

The first attempt to use semiconductors as radiation detectors was reported by McKay in 1949 (1) who tried to detect ionizing particles with point contact rectifiers and p-n junctions. However, because of impure crystals, his results were not encouraging. Now, as a result of improvements in semiconductor material technology, many different types of semiconductor detectors are available (2, 3, 4, 5).

The principles of operation of each is essentially similar to a parallel plate ionization chamber and is exemplified by a silicon p-n junction detector as shown in Fig. 1. Between the n and p type silicon
### TABLE 1

**PROPERTIES OF SILICON AND GERMANIUM (a, b, c, d)**

<table>
<thead>
<tr>
<th>Property</th>
<th>Silicon</th>
<th>Germanium</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atomic Number</td>
<td>14</td>
<td>32</td>
</tr>
<tr>
<td>Atomic Weight</td>
<td>28</td>
<td>72.6</td>
</tr>
<tr>
<td>Density (gm/cm(^3))</td>
<td>2.33</td>
<td>4.32</td>
</tr>
<tr>
<td>Dielectric Constant</td>
<td>12</td>
<td>16</td>
</tr>
<tr>
<td>Energy Gap (eV)</td>
<td>1.09</td>
<td>0.79</td>
</tr>
<tr>
<td>Energy/Electron-Hole Pair (eV)</td>
<td>3.6</td>
<td>2.8</td>
</tr>
<tr>
<td>Electron Mobility at 25(^0)C (cm(^2)/v/sec)</td>
<td>1,350</td>
<td>3,900</td>
</tr>
<tr>
<td>Hole Mobility at 25(^0)C (cm(^2)/v/sec)</td>
<td>480</td>
<td>1,900</td>
</tr>
</tbody>
</table>

**References**


there exists a highly compensated region, called the "depletion region," of width $w$, having a resistivity corresponding to intrinsic silicon. Ionizing particles striking the depletion region create electrons and holes which drift under the action of the applied field. As the carriers move they induce charge on the n and p regions proportional to the potential difference they traverse, thus giving rise to an external signal. The latter is then usually amplified and subsequently processed to determine the amount of energy deposited in the depleted region by the particle. Particles striking the detector away from the depletion zone create carriers which tend to recombine before diffusing to the junction and therefore give rise to no external charge signal.

The thickness of the depletion layer in p-n junctions can be increased by applying a reverse bias and is given approximately by (6)

$$w = \frac{\sqrt{\rho V}}{3}$$

where $w$ is the width in microns, $\rho$ is the resistivity of the lightly doped region in ohm-cms, and $V$ is the applied reverse bias voltage. The depletion region thickness determines the maximum particle energy that will be absorbed. For a typical silicon detector at 400 V bias, $w$ is about 700 microns, which is sufficient to stop a 10-MeV proton.

Increasing the thickness of the depletion region also reduces the detector capacitance, resulting in improved signal-to-noise ratios. The other main factor influencing the signal-to-noise ratio is the detector reverse leakage current which is determined mainly by the resistivity of the intrinsic region, which in turn is inversely dependent upon the operating temperature. Much larger depletion layer thicknesses are required
Fig. 1  SCHEMATIC DIAGRAM OF A P-N JUNCTION DETECTOR.
for complete absorption of photons; consequently, p-n junction counters are seldom used for gamma-ray spectroscopy. The production of relatively large intrinsic depletion volumes was first accomplished by Pell in 1960 with the lithium-ion drift technique (7) outlined in the next section.

An intrinsic region can be achieved by the ion drift technique by which donor and/or acceptor ions are drifted in the field of a reverse-biased n-p junction. The drift temperature must be sufficient to make either the donor or acceptor ions mobile but low enough to retain the n-p junction. Donor-acceptor ion pairing results in almost complete compensation of each other.

In present method, Li$^+$ ions, which are donors, are drifted in p-type silicon or germanium, uniformly doped with acceptor atoms such as boron, gallium or zinc, to a level of $N_A$ acceptors per cc. Lithium is then diffused into the crystal to give a surface concentration of $N_o$ donors per cc where $N_o \gg N_A$. The donor concentration, $N_D$ as a function of distance into the crystal as shown in Fig. 2, is equal to the acceptor concentration at position $x = c$, thus creating an n-p junction.

Applying a reverse bias to this n-p junction, thereby creating an electrostatic field near $c$, causes the positively charged Li$^+$ ions to move from the Li-rich side of the junction to the Li-deficient side. Thus, the donor concentration $N_D$ decreases for $x < c$ and increases for $x > c$, approaching the acceptor concentration $N_A$, thereby producing an intrinsic region of width $w$ as illustrated in Fig. 3. The resulting structure is known as a PIN diode. An extensive theoretical treatment of the ion-drift method, along with experimental verification of the above model is given by Pell (7).
The techniques of the lithium-ion drift method were successfully applied to the preparation of thick silicon detectors at a number of different laboratories (8, 9, 10, 11, 12). Improvements in techniques resulted in decreased drift times and depletion layer thicknesses in silicon of up to 1 cm (13).

Most applications of silicon PIN detectors were for the detection of charged particles although some measurements were made of gamma-ray spectra (14, 15). However, these detectors were of limited usefulness at energies above a few hundred keV because the ratio of photopeak area to the area under the Compton distribution was so small. The use of germanium, which has a Z of 32, compared with 14 for silicon, offers a great improvement since its photoelectric cross-section is about 40 times that of silicon. A comparison between the two for the photo interactions of photoelectric absorption, Compton scattering and pair production is shown in Fig. 4.

Successful germanium gamma-ray detectors prepared by the lithium-ion drift process, now known as Ge(Li) detectors, were first reported by Freck and Wakefield in 1962 (16), followed shortly by Webb and Williams in 1963 (17). The construction of Ge(Li) of sufficiently large volumes (up to 5 cm$^2$ x 8 mm thick) for use as practical gamma-ray spectrometers was first carried out by Tavendale in 1963 (18, 19, 20). These devices obtained photopeak efficiencies of about 0.1% to 1% at 1 MeV while yielding energy resolutions about 10 times better than is possible with the best NaI scintillation spectrometer. Soon after Tavendale's results were reported, a number of other laboratories, including this one, have prepared successful Ge(Li) gamma-ray detectors (11, 12).
Fig. 2  IMPURITY DISTRIBUTION IN CRYSTAL AFTER LITHIUM DIFFUSION
Fig. 3 IMPURITY DISTRIBUTION IN CRYSTAL AFTER DRIFT
Fig. 4 PHOTON ABSORPTION COEFFICIENTS FOR PHOTOELECTRIC EFFECT (τ), COMPTON SCATTERING (σ) AND PAIR PRODUCTION (κ) FOR SILICON AND GERMANIUM.
2. Fabrication Techniques

(a) Planar Detectors

A process for making small Ge(Li) detectors (active volumes less than about 1 cc) was developed in our laboratory. Although our process for making Ge(Li) detectors is generally the same as those initiated at other laboratories, it differs in some of the detailed techniques; consequently a summary of the process is given below. For more details one should refer to Ref. (22) which contains a detailed recipe for making small Ge(Li) detectors.

The germanium used for the fabrication of detectors was supplied by Sylvania Electric Products in the form of p-type, gallium doped, zone-levelled crystals with resistivities between 8 and 44 ohm-cms. Minority carrier lifetimes were greater than 100 µsec and dislocation densities were less than 2000/cm². The crystals were cut with a diamond saw to give thicknesses of 5 to 15 mm and cross-sectional areas between 1 and 8 cm². Surfaces were lapped and etched, and a lithium-in-oil suspension was applied to one face. The lithium was then diffused into the crystal in an argon atmosphere at 500-450°C for 10 minutes. Nickel contacts were applied by the electroless plating method (23, 24), the crystal etched and the resulting n⁺ -p diode was tested for resistance characteristics. Satisfactory diodes were then drifted in the apparatus shown schematically in Fig. 5 at approximately 50 to 55°C with DC reverse bias voltages from 200 volts at initial stages down to 30 volts at final stages of drift. The joule heating generated by the diode during the drift was dissipated by boiling of a fluorocarbon liquid (FX78 supplied by the Minnesota Mining and Manufacturing Company) and the heat of the fluorocarbon removed by
Fig. 5 SKETCH OF APPARATUS FOR LITHIUM-DRIFT PROCESS FOR GERMANIUM DETECTORS
cooling water in the condenser coils. Depletion depths of 1 1/2 to 3 mm were obtained after 1 1/2 to 4 days of drift. Several detectors 4 cm² in area and 1 cm depletion thickness have been prepared with a drift time of approximately one month. The depth of diffusion and drift were checked with a number of different staining techniques. The p-i-n diode was then etched and tested in vacuo at liquid nitrogen temperatures in the cryostat shown schematically in Fig. 6. Reverse bias currents for satisfactory detectors were between $10^{-8}$ and $10^{-10}$ amp at 100 to 300 volts. Currents higher than $10^{-8}$ amp led to excessive noise during operation and thus poor energy resolution. In these cases, the diode was re-etched until the current-voltage characteristics were satisfactory.

(b) Coaxial Detectors

Preliminary capture gamma experiments, using small Ge(Li) detectors, revealed a definite need for developing larger volume detectors. In addition to the obvious advantage of larger Ge(Li) detectors, i.e., greater counting efficiency, there are several less obvious advantages. First, the intrinsic photopeak efficiency will be greater for a large detector than for a small detector. Physically, this means that the larger detector is able to absorb the total energy of a greater fraction of those gammas which have undergone Compton collisions in the detector. As a result, the ratio of the number of counts in the full-energy peak to the number in the Compton distribution increases with detector active volume. Secondly, the intrinsic pair peak efficiency increases with the detector active volume. This increase occurs for two reasons: first, in the large detector, less electrons and positrons from high energy gammas can escape before losing all of their energy; second, less of the bremsstrahlung produced by the high-energy electrons in slowing down is lost from a larger detector. Since in
Fig. 6 SCHEMATIC DIAGRAM OF VACUUM CHAMBER AND ELECTRONICS FOR USE WITH Li-DRIFTED GERMANIUM γ-RAY DETECTORS
our work we rely on the pair production interaction at the higher energies, it was deemed important to develop large detectors that would have greater pair peak efficiencies.

Using present techniques the depth of drift is limited to about 8 mm. The largest germanium ingots presently available have cross sectional areas of about 8.5 cm\(^2\) so use of the planar method of preparation limits one to volumes of about 8 cm\(^3\). One means of getting around this limitation is the fabrication of detectors in a coaxial configuration, in which lithium is diffused, then drifted, from five of the six sides of a crystal. The procedure used for making coaxial detectors is in general the same as that for small detectors except for the application of the lithium to the crystal. The lithium cannot be easily applied to all five sides from an oil suspension as is described in step 2 of the above recipe.

We adopted a convenient technique, first used at McMaster University (25), for applying lithium to five sides of a crystal. This technique, as is shown in Fig. 7, involves dipping the germanium crystal into a 450\(^\circ\)C molten salt bath, consisting of 50\% LiCl and 50\% KCl and contained in a graphite crucible. Lithium is electroplated on to the crystal faces and it diffuses into the germanium. The only purpose of the KCl is to reduce the melting point of the LiCl salt to the optimum temperature range for the diffusion of Li into Ge. The actual diffusion takes about 30 min at currents of 0.5 to 2 amp; however, because of the need for slow heating and cooling of the fragile Ge, the entire process takes several hours. Diffusion depths, revealed by etching with 2/1 solution of HNO\(_3\)/HF, range from 1 to 2 mm.

Table 2 lists successful detectors produced to date in our laboratory. One can see from this table that the trend is definitely towards
Fig. 7 APPLICATION OF Li TO COAXIAL DETECTORS
### TABLE 1

**SUMMARY OF DETECTORS FABRICATED**

<table>
<thead>
<tr>
<th>Detector Designation</th>
<th>Date Started</th>
<th>Type</th>
<th>d = depletion depth for planar type</th>
<th>Active Volume (cc)</th>
<th>Use</th>
</tr>
</thead>
<tbody>
<tr>
<td>9 - 19.1</td>
<td>9-19-64</td>
<td>planar</td>
<td>d = 3.5 mm</td>
<td>0.6 cc</td>
<td>Nondestructive Fuel Burnup Studies</td>
</tr>
<tr>
<td>29</td>
<td>2-19-65</td>
<td>planar</td>
<td>d = 5 mm</td>
<td>2 cc</td>
<td>Given to Radiochemistry Group</td>
</tr>
<tr>
<td>30</td>
<td>2-23-65</td>
<td>planar</td>
<td>d = 5 mm</td>
<td>2 cc</td>
<td>Capture Gamma Work</td>
</tr>
<tr>
<td>31</td>
<td>3-11-65</td>
<td>planar</td>
<td>d = 8 mm</td>
<td>1.2 cc</td>
<td>Given to Radiochemistry Group</td>
</tr>
<tr>
<td>32</td>
<td>3-31-65</td>
<td>planar</td>
<td>d = 8 mm</td>
<td>3 cc</td>
<td>Capture Gamma Work</td>
</tr>
<tr>
<td>7 - 12.1</td>
<td>7-12-65</td>
<td>planar</td>
<td>d = 3.5 mm</td>
<td>3 cc</td>
<td>Given to Rockefeller Generator Group</td>
</tr>
<tr>
<td>34</td>
<td>7-28-65</td>
<td>coaxial</td>
<td></td>
<td>3 cc</td>
<td>Given to Ray Cooper for use in Activation Analysis Studies</td>
</tr>
<tr>
<td>35</td>
<td>9-9-65</td>
<td>planar</td>
<td>d = 4 mm</td>
<td>3.2 cc</td>
<td>Capture Gamma Work</td>
</tr>
<tr>
<td>37</td>
<td>10-15-65</td>
<td>coaxial</td>
<td></td>
<td>5 cc</td>
<td>Capture Gamma Work</td>
</tr>
<tr>
<td>43</td>
<td>11-23-65</td>
<td>coaxial</td>
<td></td>
<td>12 cc</td>
<td>Capture Gamma Work</td>
</tr>
<tr>
<td>44</td>
<td>12-3-65</td>
<td>coaxial</td>
<td></td>
<td>17 cc</td>
<td>Capture Gamma Work</td>
</tr>
</tbody>
</table>
larger detectors. At the present time, we have two 40 cc (approximately) crystals in the drift stage of preparation.

We have five liquid nitrogen dewars for operating and storing the detectors at 77°K. (In order to prevent deterioration of a detector's performance, it must be kept constantly at temperatures below about 150°K.) One of these dewars was specifically designed to operate as part of our pair spectrometer to measure capture gamma rays. It is described in the following subsection of this report. The remaining four dewars are of a type which has been described in detail elsewhere (26). Because of the limited number of dewars, we have made available our excess Ge(Li) detectors to several groups around MIT who have dewars.

3. Performance of Ge(Li) Detectors as Gamma-Ray Spectrometers

Figure 8 shows the gamma spectrum from Cs$^{137}$ as measured by Ge(Li) detector number 31 (8 mm depletion depth x 1.5 cm$^2$ area). The resolution is 3.3 keV (fwhm) and the peak to Compton ratio is better than 4 to 1. This resolution is the best obtained to date with any of our detectors. For the larger detectors, resolutions of 5-7 keV on the 662 keV Cs$^{137}$ gamma are typical.

Figure 9 shows the 2.614 gamma ray of ThC$^{11}$, measured with detector number 9-19.1 (0.6 cc active volume). Figure 10 shows the same spectrum measured with a larger detector--detector number 31 (1.20 cc). Furthermore, Fig. 11 shows the same spectrum measured with a still larger detector--detector number 44 (19 cc coaxial). A comparison of these three spectra reveals several interesting differences. The peak to Compton ratio increases with the active volume of the detector, illustrating one of the previously mentioned advantages of larger detectors. Also, the
Fig. 8  $^{137}$Cs GAMMA SPECTRUM, Ge(Li) DET. NO. 31, 220 VOLTS BIAS, 8 mm DEPLETION DEPTH, 1.5 cm$^2$, 1/13/66
Fig. 9 Th C\textsuperscript{\textprime}\textsuperscript{\textprime} \gamma \textit{RAY SPECTRUM} Li-Drifted Germanium Detector
No. 9-191 3.5mm Depletion Depth, 1.6 cm\textsuperscript{2} 200 Volts Bias,
77°K 20th October 1964
Fig. 10  ThC'' GAMMA SPECTRUM, Ge(Li) DET. NO. 31, 220 VOLTS BIAS, 8mm DEPLETION DEPTH, 1.5 cm², 1/13/66
Fig. II  ThC" GAMMA SPECTRUM, Ge(Li) DET. NO. 44 (17 cc ACTIVE VOLUME), 250 VOLTS BIAS, 1/6/66
Fig. 12  INTRINSIC PHOTOPEAK EFFICIENCY VS. ENERGY
FOR 3 DIFFERENT SIZE Ge(Li) DETECTORS
relative size of the single escape peak increases with detector size. This follows from the fact that a large detector is able to absorb a greater portion of the 511 KeV annihilation gammas.

Figure 12 shows intrinsic photopeak efficiencies as a function of energy for three different size detectors. The efficiencies were measured by using calibrated radioactive point sources of Co$^{57}$, Cs$^{137}$, Mn$^{54}$, Co$^{60}$, and Na$^{22}$. The increase of intrinsic photopeak efficiency with detector size is quite noticeable, especially at higher energies.

B. Neutron Capture Gamma-Ray Studies


A six-meter bent crystal spectrometer was designed and constructed by J. M. Neill and I. U. Rahman (27). This instrument was placed at the 9CH2 port of the MIT Reactor, where a flux of $5 \times 10^{11}$ n/cm$^2$-sec was available, and sources of scandium, iridium, and rhodium were irradiated and surveyed there. The capture gamma rays were detected as lines on an Ilford G5 emulsion. Figure 13 shows a photograph of the overall arrangement of the system.

The emulsion plates required processing at low temperatures since the emulsions were 600 microns thick. The slow diffusion of the developing fluid through the emulsion meant that if development at normal temperature was employed, the surface would be overdeveloped and the subsurface would be underdeveloped. The plates were sent to Livermore, California, for processing, but this resulted in a long delay between exposure and inspection. A refrigerated developing facility was therefore constructed and is located in the dark room of the MIT Reactor Building. The plates were subsequently processed there using the method suggested by Cohan (28).
FIG 13. PHOTOGRAPH OF THE BENT CRYSTAL SPECTROMETER
A systematic way of reading the plates on the comparator was specified in order that a consistent analysis could be performed. Since there were so many lines on each plate, it was decided to program the IBM 7090 digital computer at MIT to calculate the energies and the standard deviation of each unknown line.

The basic procedure in the analysis was to express the spacing of the calibration lines on the plate in a linear form. A least squares fit of the calibration line wavelengths was made to the linear form of the functional dependence of the spacing. The coefficients so obtained were used to determine the wavelength and energy of the unknown lines. Provision was made for three cases:

(a) When the plate did not straddle the beta point, (bent crystal focus), and had two or more calibration lines.
(b) When the plate did not straddle the beta point and had only one calibration line.
(c) When the plate straddled the beta point and had one or more calibration lines.

No quantitative measurements of intensity were made since it was not possible to use a photo-densitometer. The qualitative estimates made by eye were most useful; the key to the description of these estimates is given in Table 3.

It was found that the sensitivity of the bent-crystal spectrometer was flux-limited. Consequently, the scandium and the rhodium were reirradiated at a high flux location of $10^{13}$ n/cm$^2$-sec. A considerable improvement was achieved. The results are given in Tables 3 to 5. The iridium was not irradiated at the higher flux because its high activation level did not permit
TABLE 3

GAMMA RAYS FROM THERMAL-NEUTRON CAPTURE BY SCANDIUM

<table>
<thead>
<tr>
<th>Energy, keV</th>
<th>Intensity</th>
</tr>
</thead>
<tbody>
<tr>
<td>52.014 ± 0.008</td>
<td>M</td>
</tr>
<tr>
<td>61.788 ± 0.005</td>
<td>W</td>
</tr>
<tr>
<td>79.514 ± 0.008</td>
<td>W</td>
</tr>
<tr>
<td>88.938 ± 0.011</td>
<td>VW</td>
</tr>
<tr>
<td>89.821 ± 0.011</td>
<td>W</td>
</tr>
<tr>
<td>142.451 ± 0.029</td>
<td>VVS</td>
</tr>
<tr>
<td>146.974 ± 0.32</td>
<td>VVS</td>
</tr>
<tr>
<td>181.918 ± 0.067</td>
<td>W</td>
</tr>
<tr>
<td>216.39 ± 0.09</td>
<td>VS</td>
</tr>
<tr>
<td>227.71 ± 0.10</td>
<td>VVS</td>
</tr>
<tr>
<td>228.65 ± 0.10</td>
<td>VS</td>
</tr>
<tr>
<td>280.66 ± 0.15</td>
<td>VW</td>
</tr>
<tr>
<td>295.39 ± 0.14</td>
<td>VVS</td>
</tr>
<tr>
<td>383.74 ± 0.25</td>
<td>VVW?</td>
</tr>
<tr>
<td>485.94 ± 0.28</td>
<td>W</td>
</tr>
<tr>
<td>539.31 ± 0.29</td>
<td>W</td>
</tr>
<tr>
<td>546.55 ± 0.32</td>
<td>VW</td>
</tr>
<tr>
<td>554.34 ± 0.30</td>
<td>M</td>
</tr>
<tr>
<td>584.60 ± 0.33</td>
<td>M</td>
</tr>
<tr>
<td>627.12 ± 0.37</td>
<td>M</td>
</tr>
<tr>
<td>775.17 ± 0.74</td>
<td>VVW</td>
</tr>
</tbody>
</table>

Nomenclature for intensity used in Tables 3 - 5 is as follows:

S = Strong
M = Medium
W = Weak
V = Very
? = Questionable
TABLE 4

GAMMA RAYS FROM THERMAL-NEUTRON CAPTURE BY IRIDIUM

<table>
<thead>
<tr>
<th>Energy, keV</th>
<th>Intensity</th>
<th>Energy, keV</th>
<th>Intensity</th>
</tr>
</thead>
<tbody>
<tr>
<td>961.16 ± 2.55</td>
<td>VVW?</td>
<td>144.84 ± 0.09</td>
<td>VW</td>
</tr>
<tr>
<td>477.56 ± 1.41 *</td>
<td>VVW?</td>
<td>136.08 ± 0.09</td>
<td>W</td>
</tr>
<tr>
<td>418.30 ± 0.51</td>
<td>VVW?</td>
<td>112.13 ± 0.08</td>
<td>VVW</td>
</tr>
<tr>
<td>351.76 ± 0.14</td>
<td>M</td>
<td>107.95 ± 0.08</td>
<td>VW</td>
</tr>
<tr>
<td>334.03 ± 0.35</td>
<td>VVW?</td>
<td>94.318 ± 0.054</td>
<td>VVW</td>
</tr>
<tr>
<td>278.34 ± 0.35</td>
<td>VVW?</td>
<td>90.648 ± 0.072</td>
<td>VVW?</td>
</tr>
<tr>
<td>226.28 ± 0.18</td>
<td>VVW</td>
<td>90.376 ± 0.072</td>
<td>VVW?</td>
</tr>
<tr>
<td>218.77 ± 0.17</td>
<td>VVW?</td>
<td>88.724 ± 0.026</td>
<td>W</td>
</tr>
<tr>
<td>216.93 ± 0.15</td>
<td>VVW</td>
<td>86.826 ± 0.045</td>
<td>VVW?</td>
</tr>
<tr>
<td>214.91 ± 0.30</td>
<td>VVW?</td>
<td>84.257 ± 0.016</td>
<td>M</td>
</tr>
<tr>
<td>211.63 ± 0.17</td>
<td>VVW?</td>
<td>77.941 ± 0.008</td>
<td>W</td>
</tr>
<tr>
<td>178.90 ± 0.12</td>
<td>VVW</td>
<td>76.047 ± 0.021</td>
<td>VW</td>
</tr>
<tr>
<td>169.11 ± 0.12</td>
<td>VVW?</td>
<td>58.844 ± 0.012**</td>
<td>VVW</td>
</tr>
<tr>
<td>151.49 ± 0.10</td>
<td>VW</td>
<td>48.058 ± 0.018</td>
<td>VW</td>
</tr>
</tbody>
</table>

* Possible doublet.

** Isomeric decay transition in Ir$^{192}$. 
### Table 5

**Gamma Rays from Thermal-Neutron Capture by Rhodium**

<table>
<thead>
<tr>
<th>Energy, keV</th>
<th>Intensity</th>
<th>Energy, keV</th>
<th>Intensity</th>
</tr>
</thead>
<tbody>
<tr>
<td>645.42 ± 0.42</td>
<td>VVW</td>
<td>219.94 ± 0.12</td>
<td>VVW</td>
</tr>
<tr>
<td>556.12 ± 0.31*</td>
<td>W</td>
<td>217.90 ± 0.04</td>
<td>M</td>
</tr>
<tr>
<td>538.47 ± 0.28</td>
<td>VW</td>
<td>215.54 ± 0.04</td>
<td>M</td>
</tr>
<tr>
<td>447.30 ± 0.19</td>
<td>VVW</td>
<td>213.08 ± 0.04</td>
<td>VW</td>
</tr>
<tr>
<td>440.59 ± 0.17</td>
<td>W</td>
<td>202.96 ± 0.04</td>
<td>VW</td>
</tr>
<tr>
<td>427.70 ± 0.21</td>
<td>VW</td>
<td>200.92 ± 0.05</td>
<td>VVW</td>
</tr>
<tr>
<td>420.89 ± 0.15</td>
<td>W</td>
<td>196.17 ± 0.06</td>
<td>VVW</td>
</tr>
<tr>
<td>385.25 ± 0.13</td>
<td>VVW</td>
<td>185.97 ± 0.04</td>
<td>VW</td>
</tr>
<tr>
<td>374.98 ± 0.13</td>
<td>W</td>
<td>180.85 ± 0.03</td>
<td>S</td>
</tr>
<tr>
<td>371.10 ± 0.14</td>
<td>VVW</td>
<td>178.84 ± 0.03</td>
<td>W</td>
</tr>
<tr>
<td>356.87 ± 0.13</td>
<td>VVW</td>
<td>177.77 ± 0.04</td>
<td>VW</td>
</tr>
<tr>
<td>353.21 ± 0.12</td>
<td>VVW</td>
<td>169.42 ± 0.03</td>
<td>VW</td>
</tr>
<tr>
<td>333.55 ± 0.09</td>
<td>M</td>
<td>168.36 ± 0.06</td>
<td>VVW</td>
</tr>
<tr>
<td>323.88 ± 0.09</td>
<td>W</td>
<td>161.39 ± 0.06</td>
<td>VW</td>
</tr>
<tr>
<td>317.18 ± 0.08</td>
<td>VW</td>
<td>157.04 ± 0.03</td>
<td>VW</td>
</tr>
<tr>
<td>305.99 ± 0.08</td>
<td>VW</td>
<td>135.22 ± 0.03</td>
<td>W</td>
</tr>
<tr>
<td>303.69 ± 0.08</td>
<td>VVW</td>
<td>134.602 ± 0.022</td>
<td>S</td>
</tr>
<tr>
<td>290.25 ± 0.10</td>
<td>VVW</td>
<td>127.317 ± 0.020</td>
<td>S</td>
</tr>
<tr>
<td>288.63 ± 0.10</td>
<td>VVW</td>
<td>118.212 ± 0.018</td>
<td>VVW</td>
</tr>
<tr>
<td>286.18 ± 0.08</td>
<td>VVW</td>
<td>100.804 ± 0.017</td>
<td>M</td>
</tr>
<tr>
<td>273.54 ± 0.07</td>
<td>W</td>
<td>97.104 ± 0.016</td>
<td>VS</td>
</tr>
<tr>
<td>269.22 ± 0.06</td>
<td>W</td>
<td>51.421 ± 0.008</td>
<td>W</td>
</tr>
<tr>
<td>266.71 ± 0.06</td>
<td>M</td>
<td>45.292 ± 0.010</td>
<td>VVW</td>
</tr>
<tr>
<td>261.53 ± 0.06</td>
<td>VW</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*From a level in Pd$^{104}$. 
FIG 14. SAMPLE EMULSION PLATES FROM IRRADIATION OF SCANDIUM
2. Scintillation Pair Spectrometer

The principle of the scintillation pair spectrometer is based upon the measurement by a sodium iodide crystal of the kinetic energy only of the electron pair formed by a pair interaction of a primary gamma ray in that crystal. The pair interaction was selected by means of coincidence circuitry so arranged that two side crystals captured simultaneously the entire energy of the two back-to-back photons from annihilation of the position of the pair.

The geometric efficiency of the MIT scintillation pair spectrometer was increased by reducing the distance from the source to the detector. The resolution of the instrument was improved by using the below listed methods which have been reported from Brookhaven National Laboratories:

(a) Careful selection by the Harshaw Chemical Company of the central sodium iodide crystal, mounted as an integral unit without a light pipe to provide minimum intrinsic resolution.

(b) Optimization of the crystal performance by variation of the cathode-dynode potential on the photomultiplier, by variation of the focusing electrode potential, and by variation of the overall high voltage.

(c) Careful attention to shielding to reduce the external background and to reduce the background arising from scattered neutrons.

(d) Careful collimation and limitation of the maximum counting rate in the central crystal.

A resolution of 3.4\% (FWHM) at 7.6 MeV was obtained on the MIT instrument, as compared to 2.5\% achieved on similar apparatus at Brookhaven. The latter also had the advantages of a lower external back-
ground, a more flexible coincidence circuit and the availability of a 64 x 64 channel two-parameter analyzer. Figure 15 shows the arrangement of the BNL scintillation pair spectrometer, and Fig. 16 shows the electronic circuitry of the equipment. It was felt that the effort spent in developing the MIT instrument could not be justified if use of the BNL apparatus could be obtained.

Dr. Neill received a guest junior research associate appointment at Brookhaven National Laboratory, and made high-energy low-energy coincidence studies there on Dy$^{164}$ and Ho$^{165}$. Low energy coincidence studies were also made with the apparatus, though with less success. No high-energy low-energy coincidence studies were made on the capture gamma rays of rhodium. The high-energy (n, γ) spectrum of rhodium could not be well-resolved by the scintillation pair spectrometer, and the coincident count rate was found to be very low. Measurements were also made of the single pair spectrum of Dy$^{161}$, Dy$^{164}$ and Ho$^{165}$, which are shown in Figs. 17 to 19. The results of the low energy coincidence studies are given in Tables 6 to 10, and Tables 11 and 12 detail the high-energy coincidence measurements on Dy$^{164}$ and Ho$^{165}$. Figure 20 illustrates the high-energy coincidence measurements and shows the pair spectrum of capture gamma rays from Dy$^{164}$ with and without the requirement of a coincidence in the fourth crystal.

The output of the 64 x 64 channel analyzer was prodigious and a program was written for the IBM 7094 digital computer at Brookhaven National Laboratory to perform reduction of the data and to plot it. Also written for the computer was a program to calculate the line spectrum of gamma rays from measurement of the complex pair spectrum. The method used in the calculation was described by Mollenauer (29) as follows. The
complex spectrum $\overline{C}$ is related to the line spectrum $\overline{N}$ of incident gamma rays and the response function of the system $\overline{R}$ by

$$\overline{C} = \overline{R} \times \overline{N}$$

The normal matrix inversion procedure,

$$\overline{N} = \overline{R}^{-1} \times \overline{C}$$

does not work because there are invariably large fluctuations between positive and negative values in the vicinity of the peaks. The procedure is to denote successive approximations to the incident line spectrum by $\overline{N}_i$ and to the calculated complex spectrum by $\overline{C}_i$. Set $\overline{N}_1$ equal to the observed spectrum by $\overline{C}_0$ and apply in succession.

$$\overline{C}_i = \overline{R} \times \overline{N}_i$$

$$P = \frac{(C_0)_j}{(C_i)_j}$$

where $(C_i)_j$ is the $j$th element in $\overline{C}_i$, and

$$(N_{i-1})_j = (N_i)_j \times P.$$  

Convergence of $\overline{C}_i$ to $\overline{C}_o$ should be obtained.

Several difficulties were found: the method was slow, the response function of the scintillation pair spectrometer was not known well enough;
and finally, the results were not reproducible for different measurements on the same isotope. However, the work was useful in that an empirical determination of the energy dependence of the instrument response was made, (Figs. 21 and 22). The response was taken as a gaussian peak whose resolution was energy dependent according to the dotted line of Fig. 23. Added to the low-energy side of this peak were an exponential and a triangular tail. The area of the triangular tail was taken to vary linearly with energy, and the area of the exponential tail was taken to vary with the square of the incident-photon energy. Using this response a comparison was made between the measured and calculated pair spectrum of capture gamma rays from iron, (Fig. 24).
TABLE 6

LOW ENERGY GAMMA-RAY COINCIDENCES FROM IRRADIATED Sc\textsuperscript{45}

<table>
<thead>
<tr>
<th>Energies of Coincident Gamma Rays, keV</th>
<th>Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>294 ± 5</td>
<td>Definite</td>
</tr>
<tr>
<td>549 ± 17</td>
<td>Definite</td>
</tr>
<tr>
<td>208 ± 8</td>
<td>Definite</td>
</tr>
<tr>
<td>841 ± 18</td>
<td>Probable</td>
</tr>
<tr>
<td>350 ± 13</td>
<td>Possible</td>
</tr>
<tr>
<td>396 ± 16</td>
<td>Possible</td>
</tr>
<tr>
<td>118 ± 13</td>
<td>Possible</td>
</tr>
<tr>
<td>440 ± 20</td>
<td>Possible</td>
</tr>
</tbody>
</table>

TABLE 7

LOW ENERGY GAMMA-RAY COINCIDENCES FROM IRRADIATED Rh\textsuperscript{103}

<table>
<thead>
<tr>
<th>Energies of Coincident Gamma Rays, keV</th>
<th>Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>634 ± 20</td>
<td>Definite</td>
</tr>
<tr>
<td>92 ± 7</td>
<td>Probable</td>
</tr>
<tr>
<td>177 ± 13</td>
<td>Probable</td>
</tr>
<tr>
<td>174 ± 9</td>
<td>Probable</td>
</tr>
<tr>
<td>441 ± 24</td>
<td>Possible</td>
</tr>
<tr>
<td>174 ± 7</td>
<td>Possible</td>
</tr>
</tbody>
</table>
TABLE 8
LOW ENERGY GAMMA-RAY COINCIDENCES FROM IRRADIATED Dy$^{161}$

<table>
<thead>
<tr>
<th>Energies of Coincident Gamma Rays, keV</th>
<th>Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>185 ± 7</td>
<td>Probable</td>
</tr>
<tr>
<td>76 ± 4</td>
<td>Possible</td>
</tr>
<tr>
<td>307 ± 8</td>
<td>Possible</td>
</tr>
<tr>
<td>200 ± 6</td>
<td>Possible</td>
</tr>
<tr>
<td>385 ± 13</td>
<td>Possible</td>
</tr>
<tr>
<td>283 ± 8</td>
<td>Probable</td>
</tr>
<tr>
<td>819 ± 14</td>
<td>Possible</td>
</tr>
<tr>
<td>336 ± 10</td>
<td>Possible</td>
</tr>
<tr>
<td>604 ± 15</td>
<td>Possible</td>
</tr>
<tr>
<td>438 ± 13</td>
<td>Possible</td>
</tr>
</tbody>
</table>

TABLE 9
LOW ENERGY GAMMA-RAY COINCIDENCES FROM IRRADIATED Dy$^{164}$

<table>
<thead>
<tr>
<th>Energies of Coincident Gamma Rays, keV</th>
<th>Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>181 ± 5</td>
<td>Definite</td>
</tr>
<tr>
<td>74 ± 5</td>
<td>Probable</td>
</tr>
<tr>
<td>182 ± 8</td>
<td>Possible</td>
</tr>
<tr>
<td>380 ± 10</td>
<td>Probable</td>
</tr>
<tr>
<td>101 ± 8</td>
<td>Probable</td>
</tr>
<tr>
<td>1100 ± 30</td>
<td>Possible</td>
</tr>
</tbody>
</table>

TABLE 10
LOW ENERGY GAMMA-RAY COINCIDENCES FROM IRRADIATED Ho$^{165}$

<table>
<thead>
<tr>
<th>Energies of Coincident Gamma Rays, keV</th>
<th>Probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>170 ± 7</td>
<td>Definite</td>
</tr>
<tr>
<td>181 ± 6</td>
<td>Probable</td>
</tr>
<tr>
<td>633 ± 12</td>
<td>Probable</td>
</tr>
<tr>
<td>178 ± 8</td>
<td>Probable</td>
</tr>
<tr>
<td>234 ± 7</td>
<td>Probable</td>
</tr>
<tr>
<td>450 ± 15</td>
<td>Probable</td>
</tr>
<tr>
<td>191 ± 11</td>
<td>Probable</td>
</tr>
<tr>
<td>320 ± 11</td>
<td>Probable</td>
</tr>
</tbody>
</table>
### TABLE 11

**HIGH ENERGY-LOW ENERGY GAMMA-RAY COINCIDENCES**

**FROM IRRADIATED Dy\textsuperscript{164}**

<table>
<thead>
<tr>
<th>High Energy gamma ray</th>
<th>Coincident low energy gamma rays in keV, and remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.58 MeV</td>
<td>Essentially no coincidences. The small 170 and 50 keV peaks are probably from the spread of the 5.15 MeV primary gamma ray.</td>
</tr>
<tr>
<td>5.15 MeV</td>
<td>Broad definite peak whose half width extends from 330 to 450 keV. Definite peak of 170 keV, slightly broad, which seems to shift up slightly as primary gamma-ray energy is reduced. Small 50 keV peak.</td>
</tr>
<tr>
<td>4.62 MeV</td>
<td>Broad definite peak whose half width extends from 434 to 594 keV. Two possible smeared peaks of 131 and 192 keV. Possible peaks at 271 and 358 keV. Probable peak at 615 keV.</td>
</tr>
<tr>
<td>4.10 MeV</td>
<td>Definite broad peak at 185 keV. Broad definite peak whose half width extends from 420 to 560 keV. Definite 1450 keV peak. Broad definite peak comprising perhaps two gamma rays of energies 968 and 1068 keV. Possible peaks at 754 and 1153 keV. Small peak at 1640 keV but this builds up as the energy of the primary gamma ray is reduced.</td>
</tr>
<tr>
<td>3.88 MeV</td>
<td>This primary gamma ray is almost off the Y scale, and coincidences are not well defined. Definite 180 keV peak. Broad definite peak whose center is at about 480 keV. Unresolved coincidences up to a resolved definite peak at 1630 keV.</td>
</tr>
</tbody>
</table>
TABLE 12
HIGH ENERGY-LOW ENERGY GAMMA-RAY COINCIDENCES FROM IRRADIATED Ho$^{165}$

<table>
<thead>
<tr>
<th>High Energy gamma ray</th>
<th>Coincident low energy gamma rays in keV, and remarks.</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.07 MeV</td>
<td>Essentially no coincidences. The small 47 and 230 keV peaks are probably from the spread of the 5.82 MeV primary gamma ray group.</td>
</tr>
<tr>
<td>5.80 MeV</td>
<td>Definite 52 keV peak. Definite 226 keV peak. The peak of 246 keV is probably all from the spread of the 5.85 MeV primary gamma ray. Possible 199 and 86 keV spreading in from a primary gamma ray of lower energy.</td>
</tr>
<tr>
<td>5.54 MeV</td>
<td>Definite 52 keV peak. Unresolvable gamma rays from 80 to 200 keV. Probable double peak comprising gamma rays of energies 242 and 213 keV. Definite broad peak at 438 keV. Broad probable peak at 342 keV.</td>
</tr>
<tr>
<td>5.45 MeV</td>
<td>The coincidences from this gamma ray cannot be resolved easily from the 5.54 MeV primary gamma ray. The coincidence pattern is similar except that the 242 keV gamma ray is slightly more emphasized in the double peak. The 342 keV peak is somewhat more pronounced and a probable 504 keV peak has appeared.</td>
</tr>
</tbody>
</table>
TABLE 12 (Continued)

HIGH ENERGY-LOW ENERGY GAMMA-RAY COINCIDENCES FROM IRRADIATED Ho\textsuperscript{165}

<table>
<thead>
<tr>
<th>High Energy gamma ray</th>
<th>Coincident low energy gamma rays in keV, and remarks</th>
</tr>
</thead>
</table>

3. Proposed Level Schemes of Sc\textsuperscript{46}, Rh\textsuperscript{104}, Dy\textsuperscript{165}, and Ho\textsuperscript{166}.

The data on scandium were compiled from the bent-crystal spectroscopy work at MIT and from the coincidence studies by Fiebiger (30) at Brookhaven. The information existing prior to this work is represented by nuclear energy level diagrams of Groshev (31) (Fig. 25), and Mazari (32) (Fig. 26). The new level scheme for Si\textsuperscript{46} is given in Fig. 27. It has been substantially confirmed by Bostrom (33) and the levels of 142, 582, and 627 keV have been confirmed by recent (d, p) measurements of Rapaport (34). The Q value for the Sc\textsuperscript{45} (n, γ) Sc\textsuperscript{46} reaction was deduced to be 8.766 ± 0.008 MeV.

The measurements of rhodium capture gamma rays by the bent crystal spectrometer were superseded in the course of this work by Buschhorn (35) and later by Gruber (36). However, the coincidence measurements of Table 7 permitted tentative additions to be made to the nuclear energy level scheme of Rh\textsuperscript{104} proposed by Buschhorn. The additions made use of Ritz combinations of Gruber's measurements, and these additions are shown in Fig. 28.
Fig. 15  PHYSICAL ARRANGEMENT OF THE B.N.L SCINTILLATION PAIR SPECTROMETER.  (SCALE DISTORTED)
Fig. 16 ELECTRONIC BLOCK DIAGRAM FOR THE BNL SCINTILLATION PAIR SPECTROMETER
Fig. 17 PAIR SPECTRUM OF IRRADIATED Ho$^{165}$
Fig. 18 PAIR SPECTRUM OF IRRADIATED Dy$^{161}$
Fig. 19 PAIR SPECTRUM OF IRRADIATED Dy$^{164}$
Fig. 20  SINGLES AND COINCIDENCE PAIR SPECTRA
FROM IRRADIATED Dy^{164}
Energy in MeV

Arbitrary Units

Fig. 21 CALCULATED RESPONSE OF THE SCINTILLATION PAIR SPECTROMETER
Fig. 22 BASIC ELEMENTS DESCRIBING THE RESPONSE OF THE SCINTILLATION PAIR SPECTROMETER
Fig. 23 ENERGY DEPENDENCE OF THE RESOLUTION OF THE SCINTILLATION PAIR SPECTROMETER

\[ \left[ \frac{E}{\text{MeV}} \right]^{\frac{1}{2}} \]

\( E \) is gamma-ray energy
Fig. 24 Measured and Calculated Pair Spectra from Irradiated Iron
Fig. 25  NUCLEAR ENERGY LEVEL SCHEME OF 
$^{46}\text{Sc}$ PROPOSED BY GROSHEV G2
Fig. 26 NUCLEAR ENERGY LEVELS OF Sc\textsuperscript{46} MEASURED BY MAZARI M2
Fig. 27 FINAL PROPOSED NUCLEAR ENERGY LEVEL SCHEME OF Sc$^{46}$
Fig. 28  SUGGESTED ADDITIONS TO THE Rh\textsuperscript{104} NUCLEAR ENERGY LEVEL SCHEME OF BUSCHHORN BI
The high energy coincidence measurements of capture gamma rays from Dy$^{164}$ were difficult to interpret since precision measurements by Motz (37) had shown that the primary photons (Fig. 20) were either doublets or triplets. However, there was good evidence to show that the highest energy gamma ray of 5.620 MeV fed the 108 keV isomeric state implying a $Q$ value of 5.728 MeV for the Dy$^{164}$(n, $\gamma$) reaction. Using these coincidence measurements and making Ritz combinations with the data of Hickson (41) from bent crystal spectroscopy, a nuclear energy level scheme has been proposed tentatively for Dy$^{165}$. This scheme is shown in Fig. 29.

A similar approach was taken with the coincidence measurements on capture gamma rays from Ho$^{165}$. From beta decay studies some knowledge of the level scheme of Ho$^{166}$ was shown by Helmer (38) and from neutron capture studies by Estulin (39). The existing information could be summarized by the Ho$^{166}$ level diagram of Fig. 30. Making Ritz combinations with the data of Hickson (41) the level scheme of Fig. 31 has been proposed for Ho$^{166}$, again somewhat tentatively. The levels have been interpreted in terms of rotational excitations extending the original proposals of Gallagher (40). The $Q$ value for the reaction Ho$^{165}$(n, $\gamma$) Ho$^{166}$ was deduced to be $6.127 \pm 0.022$ MeV.
Fig. 29 PROPOSED POSSIBLE NUCLEAR ENERGY LEVEL SCHEME OF Dy\(^{165}\)
Fig. 30  NUCLEAR ENERGY LEVEL SCHEME OF Ho$^{166}$ PROPOSED BY ESTULIN E2
Fig. 31 PROPOSED POSSIBLE NUCLEAR ENERGY LEVEL SCHEME OF Ho$^{166}$
4. Thermal Neutron Capture Gamma Ray Studies Using Ge(Li) Detectors

The object of this work was to develop an improved method for measuring capture γ rays. The Ge(Li) detectors are the first high resolution γ-ray spectrometers that work reasonably well over the entire energy range of 0 to 10 MeV. This makes possible the determination of relative intensities of the lines to a higher degree of accuracy. At the higher energies above about 2 MeV the Ge(Li) detectors offer resolutions as good or better than any other device but they are not as good as bent crystals in the lower energy ranges where the bent crystals can be used.

(a) Experimental Facilities

In order to measure the capture gamma-ray spectra, especially of low cross section elements, it is necessary to observe the sample when it is in a high thermal neutron flux. A facility for doing this was designed and constructed at the M. I. T. R. A 9" x 9" beam was brought out from the thermal column to a heavily shielded sample facility on the reactor face. The Ge(Li) detector was placed inside a shield about 5 ft. from the sample. In this facility the thermal neutron flux at the sample is about $5 \times 10^8$ n/cm$^2$ sec and the cadmium ratio is about 2000.

The Ge(Li) drift detector was used in conjunction with two large NaI crystals to form a triple coincidence pair spectrometer essentially the same as described in Ref. (43) except the central NaI crystal was replaced with the Ge(Li) detector.

In order to keep the Ge(Li) detector between the two NaI crystals a special dewar was designed with a long cold finger. This dewar is shown in Fig. 32. As previously described (43) this detection system is contained with a steel and lead shield as shown in Fig. 33.
The advantage of using the triple coincidence system is shown in Fig. 34 which shows the 2.614 MeV gamma ray of ThC\(^{\prime}\) with and without the triple coincidence operating. Note the background is reduced by more than a factor of 10 while the peak height is reduced by only a factor of four. In addition the photopeak is completely eliminated. This is very helpful in simplifying complicated spectrum.

It is possible to use the same set of detectors in an anticoincidence Compton suppression mode. In this case any event detected by the Ge(Li) detector that is simultaneously accompanied by an event in either of the two NaI crystals is not recorded. Figures 35 and 36 show the effect of the Compton suppression mode of operation on the 1.28 MeV gamma ray of Na\(^{22}\).

(b) Preliminary Results

Since the triple-coincidence pair spectrometer became operational only recently, the bulk of the preliminary results consists of capture gamma spectra taken with different size Ge(Li) detectors--operated in the "free" mode. Figure 37 shows the capture gamma spectrum of iron measured with a small Ge(Li) detector (active volume approximately 3 cc). Figure 38 shows the resolution of the 7.644 MeV doublet of iron as measured with detector number 32 (5 cc active volume). Figure 39 shows a portion of the iron spectrum (4-10 MeV) as measured with a larger detector--number 37, (5 cc active volume). Although the resolution is not as good as that of the smaller detector, the efficiency is greater--revealing weaker lines not seen with the smaller detector.

Figures 40 and 41 show the capture gamma spectrum of manganese measured with a 17 cc Ge(Li) detector (number 44) in the energy range 3.5 to 7.5 MeV. The resolution in keV is indicated on several selected
peaks. In Fig. 41 we note the presence of the 7.644-MeV doublet from iron
capture and the 7.380-MeV capture gamma from $^{208}$Pb. These are back-
ground capture gammas resulting from thermal neutrons striking the col-
limator and detector shield.

The triple coincidence pair spectrometer has been operated with two
different size Ge(Li) detectors, a 3 cc planar detector and a 5 cc coaxial
detector. Figure 42 shows the iron capture spectrum using the pair spec-
trometer with the 3 cc detector. Comparison of Fig. 42 with Fig. 37, which
shows the same spectrum taken in the free mode, reveals that the use of the
pair spectrometer increases the peak to background ratio of the peaks.
Notice that this increase is greater at the lower energies than at the higher
energies. The reason for this is that the Compton background, which is
greater at the lower energies, is essentially removed by the pair spectrom-
eter.

Figure 43 shows the iron capture spectrum measured with the pair
spectrometer using the 5 cc coaxial Ge(Li) detector. Comparison of this
spectrum with that shown in Fig. 40, which is the same spectrum taken in
the free mode, reveals that the peak to background ratio is improved by
1.65 at 7.63 MeV. At 3.23 MeV, a similar comparison reveals an improve-
ment by a factor of 5.65. This behavior illustrates the above statement
concerning better enhancement of peak to background ratios at low than at
high energies. Comparison of the 7.63-MeV doublet in the spectra of Figs.
42 and 43 showed the 5 cc detector used in the pair spectrometer to have an
overall efficiency 13 times that of the 3 cc detector used in the pair spectrom-
eter (at 7.63 MeV).
Fig. 33 ATOMIUM CORPORATION DETECTOR SHIELD

Fig. 32 CROSS SECTION OF DETECTOR DEWAR
Fig. 34  SPECTRUM OF ThC², UPPER CURVE - "FREE", LOWER CURVE - TRIPLE COINCIDENCE, DET. NO. 32, 8 mm x 3.8 cm², 200 VOLTS, 11/3/65 - LENGTH OF RUN = 60 MIN.
Fig. 35  NORMAL SPECTRUM OF 1.26 MEV GAMMA OF Na$^{22}$, Ge(Li) DET. NO. 37 (5 CC - WRAP AROUND), 400 VOLTS BIAS, 12/13/65
Fig. 36  COMPTON - SUPPRESSION SPECTRUM OF 1.28 MEV GAMMA OF Na$^{22}$, Ge(Li) DET. NO. 37. (5CC - WRAP AROUND), 400 VOLTS BIAS - 1/24/66
Fig. 37 "FREE" IRON (n-γ) SPECTRA, Ge(Li) DETECTOR NO. 32, 200 VOLTS BIAS, DEPTH = 8 mm, AREA = 3.8 cm², 9/9/65
Fig. 38 RESOLUTION OF 7644 KeV DOUBLET IN IRON SPECTRUM

Ge(Li) DETECTOR NO. 32
8 mm Depletion Depth, 3.8 cm²
200 volts bias, 77 K
Sept. 13, 1965 190 min RUN
Fig. 39  IRON (n-γ) SPECTRUM (4–10 MeV), Ge(Li) DET. NO. 37 (5 cc WRAP AROUND), 300 VOLTS BIAS, 12/8/65
Fig. 40  MANGANESE (n−γ) SPECTRUM (5.5 TO 7.5 MeV), Ge(Li) DET. NO. 44 (17 cc WRAP AROUND), 250 VOLTS BIAS, 1/13/66
Fig. 41  MANGANESE $(n-\gamma)$ SPECTRUM (3.5 TO 5.5 MeV), Ge(Li) DET. No. 44 (17 cc WRAP AROUND), 250 VOLTS BIAS, 1/13/66
Fig. 42  TRIPLE COINCIDENCE IRON (n - γ) SPECTRUM, Ge(Li) DET. NO. 32, 200 VOLTS BIAS, DEPTH = 8 mm, AREA = 3.8 cm², 11/12/65
Fig. 43  IRON (n-γ) SPECTRUM USING PAIR SPECTROMETER - (1 TO 8 MEV)
Ge(Li) DET. NO. 37 - (5 CC WRAP AROUND) 400 VOLTS BIAS - 1/17/66
C. Nondestructive Analyses of Irradiated MITR Fuel By Gamma-Ray Spectroscopy (J. A. Sovka)

1. Introduction

Quantitative data on the irradiation history of a reactor fuel element is often difficult to obtain. Flux distributions are generally measured with foils or wires prior to initial startup and are usually calculated for full power conditions. Interpretation of these results to ascertain conditions within the fuel itself is difficult and subject to considerable error. The total amount of fuel fissioned is inferred from results of radiochemical and or mass spectroscopic

(c) Development of a Computer Code to Analyze Capture Gamma Data

An improved computer method was developed to analyze neutron capture gamma-ray spectra. This method is a modification of Carnahan's method (52) which treats each peak independently and takes into account the discontinuous nature of the data. The method eliminates the accumulation error present in most other methods.

The programs used assume a linear background under each peak and a Gaussian shape for each peak. A study of the experimental data shows these assumptions to be very reasonable for spectra obtained with a Ge(Li) γ-ray spectrometer. The usefulness of this code is demonstrated by analysing the capture γ-ray spectrum of iron. The code analyzed 23 selected peaks in this spectrum in 0.7 min. of IBM 7094 computer time. This is a considerable saving of time over stripping type methods. The code calculates peak energy relative to known standard peaks, peak area, and peak standard deviation, both theoretical and actual. Differences in these two values of the standard deviation allow the identification of unresolved peaks. Reference (44) contains a detailed listing and description of this computer code.
analyses of selected samples of the fuel. These latter methods are lengthy, tedious and costly and, in addition, have often not given agreement amongst themselves.

Attempts have frequently been made to use gamma-ray spectroscopy for nondestructive determinations of the fission product content within irradiated fuel elements. Relative measurements of total gamma activity and also individual fission product activities have been reported. Comparisons of these results with calculated relative power distributions and mass spectroscopic analyses showed that reasonable agreement was obtainable in most cases (within 15%). However, in these experiments, the total absolute fuel burnup can be accurately determined only if the spectrometer and fuel have been properly calibrated. This is usually a difficult task. It will be shown by the results of the present investigations, that even without absolute calibration, considerable additional information can be extracted from the fission product gamma-ray spectra if the gamma-rays can be sufficiently resolved and their intensities measured.

In order to resolve closely-spaced gamma rays, as in the case of irradiated fuel spectra, spectrometers of relatively high energy resolution are required. Lithium-ion drift germanium detectors have this necessary characteristic and, in addition, offer a number of advantages over other types of gamma-ray spectrometers.

The objectives of these investigations have been:

(1) To develop the techniques and apparatus required for the preparation and satisfactory operation of lithium-ion drift germanium gamma-ray spectrometers;

(2) To determine what fission products can be identified from gamma-ray spectra of irradiated MITR fuel elements with the use of such a spectrometer; and
(3) To determine the type of information that could be obtained about fuel element irradiation history from the analysis of the spectrometer data.

The following sections summarize the descriptions of the equipment and procedures used in the experimental investigation, the results obtained, and the methods used in the interpretation of these results.

2. Gamma Scanning of MITR Spent Fuel Elements

Apparatus to permit scanning of irradiated MITR fuel elements in the spent fuel storage tank was built. (Fig. 44) The detector dewar was positioned on a moveable carriage having motion both parallel and normal to the axis of the fuel element. A 1/2 in. I.D. air-filled tube, rigidly attached to the carriage, extended from near the bottom of the dewar, through the water to the surface of the element. This tube permitted a well-collimated beam of gamma rays to reach the detector. Additional collimators with aperture diameters varying between 1/16 and 1/2 in. were used in the tube to reduce counting rates to acceptable levels. Gamma-ray spectra were recorded at various positions for each of 6 fuel elements.

3. Experimental Results

The influence of cooling time upon the gamma-ray spectra from irradiated MITR fuel is shown in Fig. 45. The relative heights of each of the curves was arbitrarily chosen to indicate only how the character of the spectrum changes with cooling time. After cooling periods less than about one month, the most prominent peaks resolved were due to Ba$^{140}$ (12.8 d) and La$^{140}$ (40 hr). Subsequently, the major activity was contributed by Zr$^{95}$ (65 d) and Nb$^{95}$ (35 d), with only Pr$^{144}$ (17.5 m; daughter of 280 day Ce$^{144}$) emitting a high energy
gamma-ray at 2186 keV. An enlarged view of the energy-region from 500 keV to 800 keV for element 2M19 after 9 months cooling time, is shown in Fig. 46. The fission products identified from their gamma-ray peaks were Cs$^{134}$, Rh$^{106}$, Cs$^{137}$, Pr$^{144}$, Zr$^{95}$ and Nb$^{95}$.

A method for calculating the net counts under a gamma-ray peak, such as is shown in Fig. 46 was developed. A computer code, based upon this method, was written to facilitate processing of the data to obtain gamma-ray intensities. The axial distributions of Cs$^{137}$, Cs$^{134}$ and Zr$^{95}$ activities in element 2M19 are shown in Fig. 47, while transverse distributions are given in Fig. 48. The results of a transverse scan in which the edges of the fuel element plates were observed are presented in Fig. 49 and indicate the high spatial resolution possible with this arrangement. The fluctuations in Cs$^{137}$ activity are the result of viewing differing amounts of fuel through the collimator.

The total gamma-ray peak counts from the spectra were then corrected for self-absorption in the fuel and in lead and steel absorbers, for detector efficiency, and for decay since removal from the reactor flux. Corrections were also applied, where necessary, for $\beta$-$\gamma$ branching ratios, internal conversion coefficients and gamma-ray intensities. Ratios were then calculated of the Cs$^{137}$ activity to Cs$^{134}$ and Zr$^{95}$ activities. The axial distribution of $R_1 = A(\text{Cs}^{137}) / A(\text{Cs}^{134})$ is given in Fig. 56, while that for $R_2 = A(\text{Cs}^{137}) / A(\text{Zr}^{95})$ is shown in Fig. 51.

4. Interpretation of Results

(a) Fission Product Activities

The interpretation and analysis of the experimental results required theoretical predictions of fission product activities in MITR spent fuel, utilizing fission yields, half-lives and genetic relations of the nuclides
Fig. 44 SCHEMATIC DIAGRAM OF APPARATUS FOR GAMMA-RAY SCANNING OF MITR SPENT FUEL ELEMENTS
Fig. 45 GAMMA-RAY SPECTRA OF MITR SPENT FUEL AS A FUNCTION OF COOLING TIME

GERMANIUM LITHIUM DRIFT DETECTOR No 9-19.1 3.5 mm DEPLETION DEPTH
170 VOLTS BIAS, 77°K

CURVE | FUEL ELEMENT | COOLING TIME
--- | --- | ---
A | 2M31 | 1 1/2 WEEKS
B | 2M19 | 9 MOS.
C | 2M22 | 1YR 6 MOS.
D | 2M1 | 3YR 6 MOS.
Fig. 46 GAMMA-RAY SPECTRUM OF MITR FUEL ELEMENT 2M19  
AFTER 9 MONTHS COOLING GERMANIUM LITHIUM  
DRIFT DETECTOR No. 9-19.2 3.5 mm DEPLETION DEPTH,  
1.6 cm² 170 VOLTS BIAS, 77°K RUN D4, 5/5/65
Fig. 47 RELATIVE AXIAL DISTRIBUTIONS OF Cs\textsuperscript{137}, Zr\textsuperscript{95} (\(\gamma\)-724) AND Cs\textsuperscript{134} (\(\gamma\)-605) ACTIVITIES FOR MITR FUEL ELEMENT 2M19 AFTER 9 MONTHS COOLING
Fig. 48 RESULTS OF TRANSVERSE SCAN SHOWING Cs\textsuperscript{137}, Zr\textsuperscript{95} (\(\gamma\)-724 keV) AND Cs\textsuperscript{134} (\(\gamma\)-605 keV) ACTIVITIES AS A FUNCTION OF POSITION ACROSS FUEL ELEMENT 2MI9 AFTER 9 MONTHS COOLING TIME.
Fig. 49

RESULTS OF TRANSVERSE SCAN USING 0.125 in. DIA. APERATURE IN Pb COLLIMATOR SHOWING COMPARISON BETWEEN MEASURED RELATIVE Cs$^{137}$ ACTIVITY AS A FUNCTION OF POSITION WITH CALCULATED FRACTION OF FUEL PLATE AREA SEEN BY DETECTOR THROUGH APERATURE
Fig. 50 AXIAL PROFILE OF RATIO OF Cs$^{137}$ TO Cs$^{134}$ ACTIVITIES, AT REMOVAL FROM CORE, FOR FUEL ELEMENT 2M19.  ($R_1$ HAS BEEN CORRECTED FOR ABSORPTION IN FUEL, WATER, DETECTOR EFFICIENCY AND Pb ABSORBERS)
Fig. 51  RATIO OF Cs¹³⁷ ACTIVITY TO Zr⁹⁵ ACTIVITY ALONG FUEL ELEMENT 2M19 - CORRECTED FOR DECAY SINCE SHUTDOWN, EFFICIENCIES AND ABSORPTION
of interest. Table 13 presents a list of the fission products identified in these studies and includes some of their nuclear properties. The nuclides can be divided into three groups, each having a different mode of production. The first group includes those isotopes that are produced directly in the fission process or are the products of precursors having half-lives much shorter than their own. Group 1 type fission products include Zr\(^{95}\), Ru\(^{106}\), Cs\(^{137}\), Ba\(^{140}\) and Ce\(^{144}\). Daughter products such as Rh\(^{106}\), La\(^{140}\) and Pr\(^{144}\) each have half-lives short enough so that in most cases their activities are the same as that of the parent.

The second group includes daughters of the first group that are not in secular equilibrium with their parents and have negligible direct fission yields. Of the fission products considered here, only Nb\(^{95}\) belongs to this group.

The third group contains radioisotopes that have negligible fission yields but are produced by neutron capture reactions on other fission products. The only such radionuclide identified in these experiments was Cs\(^{134}\) formed by the \((n, \gamma)\) reaction on Cs\(^{133}\) which is a stable end-product of the mass 133 fission product chain.

Differential equations describing the net rate of accumulation of each of the above groups of fission products were written. Solutions were obtained for conditions of both constant flux operation and intermittent operation; the latter being required for MITR fuel which undergoes alternating periods of constant flux and shutdown. Numerical values to these solutions were obtained with computer codes written for this purpose.

Theoretical fission product activity ratios as a function of neutron exposure, \(\phi T\), were calculated for various fluxes and for MITR operating
<table>
<thead>
<tr>
<th>FISSION PRODUCT</th>
<th>GAMMA RAY ENERGY, MeV</th>
<th>HALF PERIOD</th>
<th>FISSION YIELD, %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>THERMAL FISSION</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>U²³⁵ (a)</td>
</tr>
<tr>
<td>Zr⁹⁵</td>
<td>0.724, 0.758</td>
<td>65 DAYS</td>
<td>6.27</td>
</tr>
<tr>
<td>Nb⁹⁵</td>
<td>0.766</td>
<td>35 DAYS</td>
<td>0.38</td>
</tr>
<tr>
<td>Ru¹⁰⁶</td>
<td>2.66, 2.40, 2.10, 1.55</td>
<td>1.0 YR.</td>
<td>6.59</td>
</tr>
<tr>
<td>Rh¹⁰⁶</td>
<td>1.05, 0.624, 0.607, 0.513</td>
<td>30 SEC</td>
<td>6.00</td>
</tr>
<tr>
<td>Cs¹³⁴</td>
<td>0.605, 0.796</td>
<td>2.3 YR.</td>
<td>5.62</td>
</tr>
<tr>
<td>Cs¹³³</td>
<td>none</td>
<td>stable</td>
<td>6.44</td>
</tr>
<tr>
<td>Cs¹³⁷</td>
<td>0.662</td>
<td>30 YR.</td>
<td>5.00</td>
</tr>
<tr>
<td>Ba¹⁴⁰</td>
<td>3.00, 2.520, 1.596</td>
<td>13 DAYS</td>
<td>6.44</td>
</tr>
<tr>
<td>La¹⁴⁰</td>
<td>0.92, 0.82</td>
<td>40 HR.</td>
<td>5.00</td>
</tr>
<tr>
<td>Ce¹⁴⁴</td>
<td>0.079, 0.133</td>
<td>285 DAYS</td>
<td>6.44</td>
</tr>
<tr>
<td>Pr¹⁴⁴</td>
<td>0.697, 1.488, 2.186</td>
<td>17 MIN</td>
<td>5.62</td>
</tr>
</tbody>
</table>

REFERENCES: (a) TABLE 10, (b) M9, (c) D2
conditions. These are shown in Fig. 52 for R₁ and in Fig. 53 for R₂.

(b) Determination of Flux, Fuel Burnup and Irradiation Time

The method developed for determining the absolute flux and irradiation time experienced by a fuel element is illustrated with results from Run D4 for element 2M19 shown as dotted lines in Figs. 52 and 53. The intersections of the experimental dotted lines with the theoretical lines for the various fluxes gives two sets of values for neutron flux and exposure. These two sets are now plotted as flux vs. exposure, as shown in Fig. 54. The intersection of the two curves gives a unique solution for the absolute neutron flux, \( \phi \), and the total exposure, \( \phi \tau \). Hence, the irradiation time, \( \tau \), consistent with these values follows directly. With this value of exposure, the \(^{235}\text{U}\) burnup can then be calculated.

The results of the application of this procedure for the data at other positions along the element are summarized in Table 14. The axial distribution of the absolute average neutron flux within 2M19 is shown in Fig. 55. Included for comparison are values calculated from results obtained by Mathews using cobalt wire monitors in MITR fuel. No normalization has been applied to the results and they represent independent absolute flux determinations. Agreement is seen to be within experimental errors.

Similarly, the average irradiation time of the element calculated by the present method agrees within 3.6% of the value determined from operating records. Using the average exposure for the element of 1.08 n/kb, the fraction of \(^{235}\text{U}\) remaining in 2M19 was \( N_{25} / N_{25}^0 = 0.57 \pm 0.06 \). The value predicted for this element by MITBURN, a two-group depletion code, was 0.656. The discrepancy between the two values is about 14%. However, recent investigations indicate that the code underestimates the
TABLE 14

SUMMARY OF RESULTS OF ANALYSIS OF FUEL ELEMENT 2M19 FOR FLUX, EXPOSURE AND IRRADIATION TIME

<table>
<thead>
<tr>
<th>Run</th>
<th>Position Below Fuel Midplane, inches</th>
<th>Neutron Flux $\phi$ $10^{13}$ n/cm$^2$, sec</th>
<th>Neutron Exposure $\omega$ n/kb</th>
<th>Irradiation Time $10^8$ seconds</th>
</tr>
</thead>
<tbody>
<tr>
<td>D4</td>
<td>-1</td>
<td>2.71</td>
<td>1.20</td>
<td>0.443</td>
</tr>
<tr>
<td>D5</td>
<td>0</td>
<td>2.61</td>
<td>1.17</td>
<td>0.446</td>
</tr>
<tr>
<td>D6</td>
<td>+2</td>
<td>2.62</td>
<td>1.21</td>
<td>0.461</td>
</tr>
<tr>
<td>D7</td>
<td>+4</td>
<td>2.61</td>
<td>1.26</td>
<td>0.484</td>
</tr>
<tr>
<td>D8</td>
<td>+6</td>
<td>2.42</td>
<td>1.08</td>
<td>0.448</td>
</tr>
<tr>
<td>D9</td>
<td>+8</td>
<td>2.31</td>
<td>1.19</td>
<td>0.513</td>
</tr>
<tr>
<td>D10</td>
<td>+10</td>
<td>2.23</td>
<td>1.17</td>
<td>0.525</td>
</tr>
<tr>
<td>D11</td>
<td>+11</td>
<td>1.72</td>
<td>0.84</td>
<td>0.488</td>
</tr>
<tr>
<td>D12</td>
<td>-2</td>
<td>2.61</td>
<td>1.20</td>
<td>0.461</td>
</tr>
<tr>
<td>D13</td>
<td>-4</td>
<td>2.50</td>
<td>1.09</td>
<td>0.436</td>
</tr>
<tr>
<td>D14</td>
<td>-6</td>
<td>2.30</td>
<td>1.03</td>
<td>0.448</td>
</tr>
<tr>
<td>D15</td>
<td>-8</td>
<td>2.29</td>
<td>0.984</td>
<td>0.430</td>
</tr>
<tr>
<td>D16</td>
<td>-10</td>
<td>1.93</td>
<td>0.848</td>
<td>0.439</td>
</tr>
<tr>
<td>D17</td>
<td>-11</td>
<td>1.54</td>
<td>0.738</td>
<td>0.479</td>
</tr>
</tbody>
</table>

Average exposure, $\bar{\omega} = 1.08$ n kb.

Mean Irradiation Time, $= (0.464 \pm 0.008) \times 10^8$ seconds

Actual Irradiation Time $= (0.448 \pm 0.013) \times 10^8$ seconds
U\textsuperscript{235} burnup so that the agreement here is also good. Similar, but less precise results, are reported for another fuel element: 2M22.

Results also indicate that with smaller aperture diameters and a more precise scanning mechanism, flux distributions within MITR fuel elements could be obtained by making use of the curvature of the fuel plates.

(c) Determination of Cooling Time

In the cases studied, the cooling time since removal from the core was obtained from records. However, it should be possible to determine this quite accurately from a comparison of a measured gamma-ray spectrum of unknown cooling time with a series of spectra taken after different cooling periods as shown in Fig. 45. It is also shown that it is possible to infer the fuel element's cooling time from the ratios of Zr\textsuperscript{95} to Nb\textsuperscript{95} activities. However, this measurement is not useful for cooling periods greater than three or four months.

(d) Determination of Relative Intensities of Zr\textsuperscript{95} Gamma Rays

Inability to achieve internal consistency in the fuel element results led to an independent determination of the relative intensities of the Zr\textsuperscript{95} gamma rays. The values obtained with (41.7 \pm 2.0)\% and (55.3 \pm 2.0)\% for the 724 keV and 758 keV gamma rays respectively.

5. Conclusions

A new method has been developed for inferring a fuel element's irradiation history from analyses of gamma-ray spectra obtained with a Ge(Li) spectrometer. The results include determinations of absolute neutron flux, total neutron exposure, U\textsuperscript{235} burnup and total irradiation time. Comparisons with independently obtained data showed agreement was within
Fig. 52  RATIO OF $^{137}$Cs TO $^{134}$Cs ACTIVITIES vs. NEUTRON EXPOSURE FOR MITR FUEL AT VARIOUS FLUXES FOR INTERMITTENT OPERATION, 95 HRS ON, 73 HRS OFF
Fig. 53  RATIO OF Cs$^{137}$ TO Zr$^{95}$ ACTIVITY AS A FUNCTION OF NEUTRON EXPOSURE FOR VARIOUS FLUXES. MITR INTERMITTENT. OPERATION 95 HRS ON, 73 HRS OFF.
Fig. 54  CURVES OF NEUTRON FLUX vs NEUTRON EXPOSURE FOR ACTIVITY RATIOS \( R_1 \) AND \( R_2 \) INDICATING METHOD OF SOLUTION FOR FLUX AND EXPOSURE

\[ R_1 = \frac{A(\text{Cs}^{137})}{A(\text{Cs}^{134})} \]

\[ R_2 = \frac{A(\text{Cs}^{137})}{A(\text{Zr}^{95})} \]

SOLUTION AT
\( \phi = 2.71 \times 10^{13} \text{ n/cm}^2\text{-sec} \)
\( \omega = 1.20 \text{ n/kb} \)
Fig. 55 AXIAL DISTRIBUTION OF AVERAGE THERMAL NEUTRON FLUX IN ELEMENT 2M19
the assigned errors. One major advantage of the present method, in addition to being nondestructive, is that the results obtained are representative of the irradiation conditions existing with the fuel itself during normal operation, (i.e., at operating temperatures, pressures, etc.). The operating pattern of the element and the time since its removal from the reactor could also be inferred.

The method has applications to some of the problems of fuel design and fuel reprocessing. It may also have some useful applications to the field of international control of reactor fuel.
III. NEUTRON SPECTROSCOPY AND DOSIMETRY

(R. A. Rydin and G. L. Brownell)

A. **Introduction**

The purpose of this work was to develop and improve the experimental, analytical and calculational methods of obtaining fast neutron spectral information from foil activation measurements. This information was then used to predict fast neutron doses. To illustrate the use of the method the neutron spectrum of the MITR Medical Therapy Facility was measured and predictions were made of the fast neutron dose in tissue-like materials. This section summarizes the principal results of this work, the details of which were issued as scientific report AFCRL-64-404.

B. **Analytical Methods**

The data obtained from a threshold foil activation experiment can be conveniently expressed in the following form,

\[
K_i = \int_0^\infty \sigma_i(E) \phi(E) \, dE
\]

(1)

where \(K_i\) is the experimental result corrected to saturation and corrected for weight, abundance, counter efficiency, etc; \(\sigma_i(E)\) is the absorption cross section as a function of energy, and \(\phi(E)\) is the differential flux.

The primary problem is to extract the differential spectrum, \(\phi(E)\), from a set of experimental quantities.

There are several methods available to treat foil activation data to determine the differential spectrum. All require an accurate knowledge of the neutron absorption cross section curve as a function of energy for the reaction
of interest. Errors in this respect, therefore, affect all methods. Further, the problem is attacked in the following general fashion in all methods: 1) an appropriate form of the spectral shape is assumed, which has unspecified coefficients; 2) the foil activation cross section curves are numerically integrated over an energy interval with respect to the assumed spectral shape, and 3) the experimentally measured activations are used to specify the appropriate coefficients of the assumed shape and hence, specify the measured spectrum. Finally, all methods expect to extract a large amount of differential information from a few integral measurements.

Historically, the Effective Threshold Method is the oldest and perhaps most widely used of the analytical methods. Basically, the expression for the activation of a foil is replaced by an equivalent expression, which assumes that the actual cross section curve can be replaced by a step function, which starts at an effective threshold energy. Thus,

$$K_i = \sigma_{effi} \int_{E_{effi}}^{\infty} \phi(E) \, dE = \sigma_{effi} \bar{\Phi} E_{effi} \quad (2)$$

The experimental activation gives the integral flux above the threshold, and the use of several detectors with different thresholds leads to a histogram plot of the integral flux. To determine the effective threshold values, a fission spectrum is commonly assumed and the effective cross section determined by integration, once we have picked an effective threshold energy or vice versa. An assumption is made that the effective values obtained in this manner are also valid for a nonfission spectrum, which is not always the case.
There are several methods which lead directly to the differential spectrum. First, there is the Polynomial Method in which the spectrum is expanded in a polynomial having as many terms as there are threshold data values. In principle, the coefficients of the polynomial can be determined using the experimental data, but the resulting set of equations is poorly conditioned. This usually results in oscillations in the calculated spectrum and, for this reason, the method is not recommended. Next, there are the Dierckx Method and the Italian Iterative Method in which the spectrum is assumed to be made up of a series of exponentials joined at appropriate energy values. The experimental data are used to determine the coefficient in each of the exponentials and to normalize the curve. These methods are useful only when the measured spectrum has certain specific features; i.e., $\phi(E)$ is a smoothly decreasing function of energy. Finally, there are two variations of what are known as the orthonormal methods, the Weighted Orthonormal Method and the Weighted Orthonormal Polynomial Method. These are suitable, in principle, for any spectral shape.

In the Weighted Orthonormal Method, the spectrum is assumed to be made up of a weighting function times an expansion of known functions of energy, which are required to form an orthonormal set. As many coefficients are used as there are foils and thus the coefficients can be uniquely determined from the experimental data. The advantage is that the coefficients are determined in a best fit sense.

The spectrum is assumed to have the form

$$\phi(E) = W(E) \left[ \sum_{i=1}^{n} B_i \psi_i(E) \right] \left( \frac{\text{neutrons}}{\text{cm}^2 \text{-sec}\cdot\text{MeV}} \right) \quad (3)$$
The functions, \( \psi_i(E) \), are defined to be made up of a linear combination of the cross section curves and are defined to be orthonormal with respect to the weighting function, \( W(E) \). We write

\[
\psi_i(E) = \sum_{j=1}^{j} A_{ij} \sigma_i(E)
\]

The \( A_{ij} \) are determined from the orthonormal conditions

\[
\int_{0}^{E_{\text{max}}} W(E) \psi_i(E) \psi_j(E) \, dE = \delta_{ij} \quad (i, j = 1, n)
\]

Using these coefficients, the experimental activations, and the known cross section curves, the result can be expressed by the following equation

\[
\phi(E) = W(E) \left[ \sum_{k=1}^{n} \left\{ \left( \sum_{i=1}^{n} A_{ki} K_i \right) \left( \sum_{j=1}^{n} A_{kj} \sigma_j(E) \right) \right\} \right]
\]

This calculation has been coded for the IBM 7090 computer and the program is designated FUSE-3. A schematic flow diagram of the program appears in Fig. 56. The Orthonormal Polynomial Method, which is quite similar, has also been coded and is designated FUSE-4.

In the epithermal region, the spectrum is assumed to be asymptotic to \( 1/E \), and thus is given by

\[
\phi(E) = \frac{\Phi_0}{E} \quad \text{(neutrons/cm}^2\cdot \text{sec MeV)}
\]

The value of \( \Phi_0 \) is determined from the conventional flux, \( \Phi_{2200} \), and from cadmium ratio measurements performed with resonance foils. The expression used is
Fig. 56
FUSE-3 WEIGHTED ORTHONORMAL METHOD
SCHEMATIC FLOW DIAGRAM OF MAIN PROGRAM
(WRITTEN IN FORTRAN FOR THE IBM-7090 COMPUTER)

(INPUT) reads cross section data, control data, input activations, and weighting function

(CRSINT) calculates weighted cross section integrals

(ACALC) calculates orthonormal coefficients

(OUTPUT) prints heading, foil order orthonormal coefficients, weighting function

(AVGE) Simpson integration

(RANNF-SETUF) random number generator

(STAT) statistically deviates input activations

(FLUX) calculates differential flux from measured activations

(FLUXAV) linearly averages differential flux values

(INTEG) calculates integral flux and expected activations

(TYPE) prints differential and integral flux and expected activations

(OUTPUT) plots semilogarithmic graph of flux and weighting function

(DESCRIPTIONS)
- (AVGE): Simpson integration
- (CRSINT): Calculates weighted cross section integrals
- (ACALC): Calculates orthonormal coefficients
- (RANNF-SETUF): Random number generator
- (STAT): Statistically deviates input activations
- (FLUX): Calculates differential flux from measured activations
- (FLUXAV): Linearly averages differential flux values
- (INTEG): Calculates integral flux and expected activations
- (TYPE): Prints differential and integral flux and expected activations
- (OUTPUT): Plots semilogarithmic graph of flux and weighting function
\[ \tilde{\phi}_o = \frac{(1.87) \cdot 2200}{(R_{cd} - 1)(1 + \alpha)} \]  

where \( R_{cd} \) is the experimental cadmium ratio result and \( \alpha \) is a known parameter giving the relative ratio of resonance to \( 1/\nu \) captures.

The conventional flux in the beam is measured with dilute gold foils, which are absolute counted by \( \beta - \gamma \) coincidence. The expression used is

\[ \frac{\tilde{\phi}_{2200}}{K} = \frac{R_{cd} - 1}{N \sigma_{2200} g} \]  

where \( K \) is the absolute saturation activity, \( g \) is Westcott's factor, \( \sigma_{2200} \) is the 2200 meter per second cross section, and \( R_{cd} \) is the measured gold foil cadmium ratio. The thermal flux distribution is assumed to be a Maxwellian at 63 \(^\circ\)C and is calculated from

\[ \phi(E) = \frac{4 \pi T_0}{\pi T_0} \int_{0}^{E_{max}} \frac{E}{kT} \exp \left( -\frac{E}{kT} \right) \]  

Finally, dose rates in tissue are obtained using the measured composite neutron spectrum and Snyder's Monte Carlo depth dose curves, one of which is shown in Fig. 57. The dose rates due to protons, gamma rays, and heavy particle recoils are calculated as a function of energy at several different tissue depths. To simplify the dose integrations, the energy variable is converted to lethargy and the depth dose curves are interpolated using La-Grangian interpolation. The resulting dose rates can be expressed as

\[ R_i(x) = \int_{E}^{E_{max}} D_i(E, x) \phi(E) \, dE = \int_{0}^{u} D_i(u, x) \phi(u) \, du \text{ (rads/min)} \]  

where the \( D_i \) represent the interpolated dose curves (taken from the NBS
Handbook 63). This calculation is performed by the computer code DOSE, a schematic diagram of which is shown in Fig. 58.

C. Experimental Methods

In this work, ten different threshold foil reactions were employed. Some of the properties of these reactions are shown in Table 15. All measurements were performed relative to thermally activated standard reactions, leading to products emitting the same or similar gamma ray energies. (See Table 16). Thus, the absolute calibration was referred to the thermal flux, which was measured by absolute \( \beta-\gamma \) coincidence counting of dilute gold foils. In addition, gold monitor foils were used with each of the threshold and thermal standard foils to intercalibrate the results.

Due to the low intensity of the fast neutron flux, the threshold foils used were rather large, while the thermal standard foils were small in comparison. (See Table 17). In order to obtain the same geometric counter efficiency, an \( \sim 4\pi \) scintillation counter apparatus was constructed and used to count all foils. (See Fig. 59). This apparatus was an adaptation of the triple coincidence spectrometer used in J. Neill's work.

Three different techniques were employed to resolve the foil data: 1) For half-lives greater than 1 hour, spectra were measured with a scintillation counter and a 256 channel analyzer. The total count in an appropriate number of channels was used in the comparison between the threshold foil and the thermal standard foil; 2) For half-lives less than 15 min. decay curves were measured, using a 256 channel multiscaler attachment on the analyzer, and the components were fit by FRANTIC, an iterative weighted least squares exponential growth and decay data fitting program written by P. Rogers. A comparison was made between curves which were
Fig. 57 DEPTH DOSE CURVE IN TISSUE, ENERGY ABSORPTION FROM 10 MeV. NEUTRON BEAM
(TAKEN FROM ORNL-1872)
Fig. 58
SCHEMATIC FLOW DIAGRAM FOR DØSE
(WRITTEN IN FORTRAN FOR THE IBM - 7090 COMPUTER)

1. READ IN DOSE CURVES VS ENERGY
2. CONVERT TO LETHARGY, INTERPOLATE EVENLY SPACED SET OF DOSE VALUES
3. READ IN DIFFERENTIAL FLUX VS ENERGY
4. CONVERT TO LETHARGY, INTERPOLATE EVENLY SPACED SET OF FLUX VALUES
5. (AVGE) SIMPSON INTEGRATION
6. PERFORM DOSE RATE INTEGRATIONS AT EVERY ODD LETHARGY POINT
7. CALCULATE DOSE FRACTIONS
8. PRINT DOSE RATE RESULTS

(LINLAG) LAGRANGIAN INTERPOLATION
Fig. 59

\(\sim 4\pi\) SCINTILLATION COUNTER APPARATUS

```
PREAMP

P.M. TUBE

Na I

WELL

Na I

P.M. TUBE

PREAMP

SCINTILLATOR

SCINTILLATOR

LEAD LINED STEEL CASK

BALANCING POTENTIOMETERS

HIGH VOLTAGE SUPPLY

R - C MATCHING NETWORKS

SPECTRUM ANALYSIS
256 CHANNEL ANALYZER

DECAY CURVE ANALYSIS
256 CHANNEL MULTISCALER

N₂ GAS PURGE
```
Fig. 60 BACKGROUND CORRECTED FISSION PRODUCT DECAY CURVES, $^{235}\text{U}(n,f)$; $^{238}\text{U}(n,f)$; $^{232}\text{Th}(n,f)$; $^{237}\text{Np}(n,f)$
RELATIVE COUNTS/CHANNEL (>1.3 MEV) VS TIME
## TABLE 15

**THRESHOLD REACTIONS EMPLOYED IN THIS WORK**

<table>
<thead>
<tr>
<th>REACTION</th>
<th>EFFECTIVE THRESHOLD (Mev)</th>
<th>EFFECTIVE CROSS SECTION (mb)</th>
<th>γ-RADIATION (59)</th>
<th>T₁/₂ (59)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Np²³⁷ (n,f)</td>
<td>0.68 (a)</td>
<td>1450</td>
<td>fission products</td>
<td>—</td>
</tr>
<tr>
<td>U²³⁸ (n,f)</td>
<td>1.48 (a)</td>
<td>580</td>
<td>fission products</td>
<td>—</td>
</tr>
<tr>
<td>Th²³² (n,f)</td>
<td>1.56 (a)</td>
<td>140</td>
<td>fission products</td>
<td>—</td>
</tr>
<tr>
<td>In¹¹⁵⁺ (n,n⁺) In¹¹⁵⁺</td>
<td>1.3 (c)</td>
<td>290</td>
<td>0.335</td>
<td>4.5 hr.</td>
</tr>
<tr>
<td>Ni⁵⁸ (n,p)Co⁵⁸</td>
<td>3.09 (a)</td>
<td>550</td>
<td>0.805</td>
<td>71.3 days</td>
</tr>
<tr>
<td>Al²⁷ (n,p)Mg²⁷</td>
<td>5.46 (a)</td>
<td>80</td>
<td>1.015</td>
<td>9.5 min.</td>
</tr>
<tr>
<td>Si²⁸ (n,p)Al²⁸</td>
<td>6.1 (b)</td>
<td>190</td>
<td>1.78</td>
<td>2.30 min.</td>
</tr>
<tr>
<td>Mg²⁴ (n,p)Na²⁴</td>
<td>6.3 (b)</td>
<td>48</td>
<td>2.754</td>
<td>1.368</td>
</tr>
<tr>
<td>Fe⁵⁶ (n,p)Mn⁵⁶</td>
<td>6.42 (a)</td>
<td>50</td>
<td>0.845</td>
<td>2.58 hr.</td>
</tr>
<tr>
<td>Al²⁷ (n,γ)Na²⁴</td>
<td>7.52 (a)</td>
<td>70</td>
<td>2.754</td>
<td>1.368</td>
</tr>
</tbody>
</table>

a) M. Bresesti et al (813)
b) J. Metoff et al (144)
c) C. Sawyer (529)
# TABLE 16

## THERMALLY ACTIVATED STANDARD REACTIONS

<table>
<thead>
<tr>
<th>THERMAL STANDARD REACTION</th>
<th>( \sigma_{2200}(\text{barns}) )</th>
<th>( \text{ENERGY (MEV)} )</th>
<th>( T_{1/2} )</th>
<th>CORRESPONDING REACTION</th>
<th>( \text{ENERGY (MEV)} )</th>
<th>( T_{1/2} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( ^{235}\text{U}(n,f) )</td>
<td>582. ± 4. ((q = .9665))</td>
<td>fission products</td>
<td>-</td>
<td>( ^{237}\text{Np} (n,f) )</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>( ^{232}\text{Th}(n,f) )</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>( ^{238}\text{Th} (n,f) )</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>( ^{238}\text{U}(n,f) )</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>( ^{23}\text{Na}(n,\gamma) ^{24}\text{Na} )</td>
<td>0.536 ± 0.01</td>
<td>1.368(100)</td>
<td>15.0 hr</td>
<td>( ^{27}\text{Al}(n,\alpha) ^{24}\text{Na} )</td>
<td>1.368(100)</td>
<td>15.0 hr</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.754(100)</td>
<td></td>
<td>( ^{24}\text{Mg}(n,p) ^{24}\text{Na} )</td>
<td>2.754(100)</td>
<td></td>
</tr>
<tr>
<td>( ^{55}\text{Mn}(n,\gamma) ^{56}\text{Mn} )</td>
<td>13.3 ± 0.2</td>
<td>.845 (100)</td>
<td>2.58 hr</td>
<td>( ^{56}\text{Fe}(n,p) ^{56}\text{Mn} )</td>
<td>.845(100)</td>
<td>2.58 hr</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>( ^{58}\text{Ni}(n,p) ^{58}\text{Co} )</td>
<td>.805(100)</td>
<td>71.3 day</td>
</tr>
<tr>
<td>( ^{27}\text{Al}(n,\gamma) ^{28}\text{Al} )</td>
<td>0.21 ± 0.02</td>
<td>1.78(100)</td>
<td>2.3 min</td>
<td>( ^{28}\text{Si}(n,p) ^{28}\text{Al} )</td>
<td>1.78(100)</td>
<td>2.3 min</td>
</tr>
<tr>
<td>( ^{26}\text{Mg}(n,\gamma) ^{27}\text{Mg} )</td>
<td>0.027 ± .005</td>
<td>.843(70)</td>
<td>9.5 min</td>
<td>( ^{27}\text{Al}(n,p) ^{27}\text{Mg} )</td>
<td>.843(70)</td>
<td>9.5 min</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.015(30)</td>
<td></td>
<td></td>
<td>1.015(30)</td>
<td></td>
</tr>
<tr>
<td>( ^{50}\text{Cr}(n,\gamma) ^{51}\text{Cr} )</td>
<td>15.9 ± 1.6</td>
<td>.325(9.8) ( (B18) )</td>
<td>27.8 day</td>
<td>( ^{115}\text{In}(n,\alpha) ^{115}\text{In} )</td>
<td>.335(47.7)</td>
<td>4.5 hr</td>
</tr>
</tbody>
</table>
### TABLE 17

**FOIL MATERIALS – APPROXIMATE WEIGHTS AND TIMES**

<table>
<thead>
<tr>
<th>MEASUREMENT</th>
<th>MATERIAL</th>
<th>WEIGHT</th>
<th>IRRADIATION TIME</th>
<th>TIME TO COUNTING</th>
<th>COUNTING TIME</th>
</tr>
</thead>
<tbody>
<tr>
<td>Np(^{237}) Threshold</td>
<td>NpO(_2) in Al can (Cd)</td>
<td>.25 gm</td>
<td>20 min</td>
<td>—</td>
<td>2 hrs</td>
</tr>
<tr>
<td>U(^{238}) Threshold</td>
<td>Depleted (18 ppm) Uranium Metal in Mylar (Cd)</td>
<td>1 gm</td>
<td>20 min</td>
<td>—</td>
<td>2 hrs</td>
</tr>
<tr>
<td>Th(^{232}) Threshold</td>
<td>Thorium Metal in Mylar (Cd)</td>
<td>3 gm</td>
<td>20 min</td>
<td>—</td>
<td>2 hrs</td>
</tr>
<tr>
<td>U(^{235}) Standard</td>
<td>Nat. Uranium Metal in Mylar</td>
<td>2 mg</td>
<td>20 min</td>
<td>—</td>
<td>2 hrs</td>
</tr>
<tr>
<td>Al(^{27}(n,\gamma)) Threshold</td>
<td>Aluminum Metal (Cd)</td>
<td>7 gm</td>
<td>4 hrs</td>
<td>12 + hrs</td>
<td>10–20 min</td>
</tr>
<tr>
<td>Mg(^{24}(n,p)) Threshold</td>
<td>Magnesium Metal (Cd)</td>
<td>6 gm</td>
<td>4 hrs</td>
<td>12 + hrs</td>
<td>10–20 min</td>
</tr>
<tr>
<td>Na(^{23}) Standard</td>
<td>Na(_2)CO(_3) Powder in Mylar</td>
<td>4 mg</td>
<td>4 hrs</td>
<td>12 + hrs</td>
<td>1–10 min</td>
</tr>
<tr>
<td>Fe(^{56}(n,p)) Threshold</td>
<td>Iron Metal (Cd)</td>
<td>10 gm</td>
<td>2 hrs</td>
<td>—</td>
<td>5–10 min</td>
</tr>
<tr>
<td>Mn(^{55}) Standard</td>
<td>MnO(_2) Powder in Mylar</td>
<td>5 mg</td>
<td>5 min</td>
<td>—</td>
<td>1–5 min</td>
</tr>
<tr>
<td>Ni(^{58}(n,p)) Threshold</td>
<td>Nickel Metal (Cd)</td>
<td>8 gm</td>
<td>4 hrs</td>
<td>4 + days</td>
<td>30 min</td>
</tr>
<tr>
<td>Si(^{28}(n,p)) Threshold</td>
<td>Silicon Metal (Cd)</td>
<td>8 gm</td>
<td>10 min</td>
<td>—</td>
<td>30 min</td>
</tr>
<tr>
<td>Al(^{27}) Standard</td>
<td>Aluminum Foil</td>
<td>1 mg</td>
<td>10 min</td>
<td>—</td>
<td>30 min</td>
</tr>
<tr>
<td>Al(^{27}(n,p)) Threshold</td>
<td>Aluminum Metal (Cd)</td>
<td>7 gm</td>
<td>20 min</td>
<td>—</td>
<td>1(\frac{1}{2}) hrs</td>
</tr>
<tr>
<td>Mg(^{26}) Standard</td>
<td>Magnesium Metal</td>
<td>30 mg</td>
<td>20 min</td>
<td>—</td>
<td>1(\frac{1}{2}) hrs</td>
</tr>
<tr>
<td>In(^{115}(n,n)) Threshold</td>
<td>Indium Metal (Cd)</td>
<td>4 gm</td>
<td>4 hrs</td>
<td>12 + hrs</td>
<td>1–5 min</td>
</tr>
<tr>
<td>Cr(^{50}) Standard</td>
<td>Cr(_2)O(_3) Powder in Mylar</td>
<td>70 mg</td>
<td>4 hrs</td>
<td>12 + hrs</td>
<td>1–5 min</td>
</tr>
<tr>
<td>Au(^{197}) Resonance</td>
<td>1% Gold in Aluminum Alloy</td>
<td>5 mg</td>
<td>2–4 hrs</td>
<td>1 + hrs</td>
<td>1–5 min</td>
</tr>
<tr>
<td>Au(^{197}) Resonance</td>
<td>1% Gold in Alum. Alloy (Cd)</td>
<td>30 mg</td>
<td>2–4 hrs</td>
<td>1 + hrs</td>
<td>1–5 min</td>
</tr>
<tr>
<td>Mn(^{55}) Resonance</td>
<td>MnO(_2) powder in Mylar</td>
<td>4 mg</td>
<td>1–5 min</td>
<td>—</td>
<td>1–5 min</td>
</tr>
<tr>
<td>Mn(^{55}) Resonance</td>
<td>MnO(_2) powder in Mylar (Cd)</td>
<td>50 mg</td>
<td>1–5 min</td>
<td>—</td>
<td>1–5 min</td>
</tr>
<tr>
<td>Na(^{23}) Resonance</td>
<td>Na(_2)CO(_3) powder in Mylar</td>
<td>2 mg</td>
<td>2–4 hrs</td>
<td>12 + hrs</td>
<td>1–10 min</td>
</tr>
<tr>
<td>Na(^{23}) Resonance</td>
<td>Na(_2)CO(_3) powder in Mylar (Cd)</td>
<td>40 mg</td>
<td>2–4 hrs</td>
<td>12 + hrs</td>
<td>1–10 min</td>
</tr>
<tr>
<td>Au(^{197}) Monitor</td>
<td>1% Gold in Aluminum Alloy</td>
<td>5 and 30 mg</td>
<td>10 min – 4 hrs</td>
<td>1 + hrs</td>
<td>1–5 min</td>
</tr>
</tbody>
</table>
fit to the threshold foil data and the thermal standard foil data; 3) For
the fissionable reactions, it was found that the gross fission product decay
curves were similar for different fissionable nuclides, (see Fig. 60) and
that these curves could be fit by FRANTIC to the sum of two or three
fictitious components. A comparison was made between curves fit to a
fictitious 52.5 minute component present in the threshold foil data and the
thermal standard foil data. A calibration was also performed to relate
these results to the relative fission rates.

Impurity activity effects were handled by a variety of methods.
These included correction of the results by cadmium ratios, and the use
of cadmium filters, gamma energy discrimination, background subtrac-
tion, and decay time, irradiation time, and curve stripping procedures
to minimize the contribution of these effects to the experimental data.

Three resonance foil materials were used to measure the epi-
thermal spectrum. The properties of these reactions are given in Table
18. The experiments consisted of cadmium ratio measurements performed
with these detectors.

D. Results

The experiments performed in the Medical Therapy Beam con-
sisted of the following: 1) An absolute thermal flux calibration to specify the
thermal neutron spectrum and normalize the other spectral measurements;
2) A set of measurements to determine the spacial uniformity of the Medical
Beam flux; 3) A set of resonance foil measurements to specify the epi-
thermal spectrum; and 4) A set of threshold foil measurements to specify
the fast neutron spectrum. Results of these experiments are presented
here. In addition, results of the fast neutron spectrum calculations are
presented. These consist of calculations made with "exact" data, with data
<table>
<thead>
<tr>
<th>RESONANCE REACTION</th>
<th>RESONANCE (^{(H9)}) ENERGY (ev)</th>
<th>((.48\text{ev})^{(R3)}) (\alpha)</th>
<th>((.35\text{ev})^{(\alpha')})</th>
<th>GAMMA (^{(59)}) ENERGY (Mev)</th>
<th>(T_{1/2}^{(59)})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\text{Au}^{197}\ (n, \gamma) \text{Au}^{198})</td>
<td>4.904</td>
<td>36.2</td>
<td>30.9</td>
<td>.412</td>
<td>2.697d</td>
</tr>
<tr>
<td>(\text{Mn}^{55}\ (n, \gamma) \text{Mn}^{56})</td>
<td>337.</td>
<td>1.41</td>
<td>1.205</td>
<td>.840</td>
<td>2.58 hr.</td>
</tr>
<tr>
<td>(\text{Na}^{23}\ (n, \gamma) \text{Na}^{24})</td>
<td>2895.</td>
<td>0.031</td>
<td>0.0265</td>
<td>2.754</td>
<td>1.368</td>
</tr>
</tbody>
</table>
from the ISPRA-1 Reactor, and with the MITR data. Finally, the results of dosimetry calculations in tissue, performed using Snyder's Monte Carlo depth dose curves and the composite MITR spectrum, are presented.

The conventional flux in the central portion of the Medical Therapy beam, at a position 2 1/2 inches beneath the beam port, was measured with four dilute gold foils. These were irradiated and subsequently absolute counted in a $\beta$-$\gamma$ coincidence counting apparatus. The conventional flux in the beam, at a power level of 2MWT, was calculated to have the value

$$\Phi_{2200} = 2.84 \times 10^9 \frac{\text{neutrons}}{\text{cm}^2 \cdot \text{sec}}$$  \hspace{1cm} (12)

The spatial uniformity of the beam in the plane of the foil holder, located 2 1/2 inches beneath the beam port, was measured using seventeen 1/2-inch diameter dilute gold foils for the thermal flux uniformity and nine 1-inch diameter aluminum threshold foils for the fast flux uniformity. The results are plotted as isoflux maps in Figs. 61 and 62. Note that the thermal flux is uniform to within 10% in the 3-inch diameter central circle, and also that the fast flux is more spatially uniform than the thermal flux. There is also a thermal flux nonuniformity associated with the vertical direction, which amounts to about a 5% change in flux for a quarter inch change in height.

The results of the epithermal flux measurements are given in Table 19. When these values are plotted on log-log paper, a line can be fit through the data which has a slope of -.895. (See Fig. 63). This indicates that the epithermal spectrum obeys the law,

$$\phi(E) \propto \frac{1}{E^{0.895}}$$  \hspace{1cm} (13)

The results of the MITR threshold foil measurements for 10 different
Fig. 61 SPACIAL UNIFORMITY OF THE THERMAL FLUX IN THE CENTRAL PORTION OF THE MEDICAL BEAM NORMALIZED ISOFLUX MAP
Fig. 62 SPACIAL UNIFORMITY OF THE FAST FLUX IN THE CENTRAL PORTION OF THE MEDICAL BEAM
NORMALIZED ISOFLUX MAP
Fig. 63 RESONANCE FOIL DATA, DIFFERENTIAL FLUX VS ENERGY
TABLE 19
RESONANCE FOIL EPITHERMAL FLUX DATA

<table>
<thead>
<tr>
<th>Foil</th>
<th>$R_{CD}$</th>
<th>$E_{RES}^{(ev)}$</th>
<th>$\Phi_0 \left( \frac{\text{neutrons}}{\text{cm}^2 \cdot \text{sec}} \right)$</th>
<th>$\Phi(E_{RES}) \left( \frac{\text{neutrons}}{\text{cm}^2 \cdot \text{sec} \cdot \text{MeV}} \right)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gold</td>
<td>22.9 ± 0.7</td>
<td>4.904</td>
<td>$7.60 \times 10^6$</td>
<td>$1.55 \times 10^{12}$</td>
</tr>
<tr>
<td>Manganese</td>
<td>222. ± 22</td>
<td>337.</td>
<td>$1.09 \times 10^7$</td>
<td>$3.24 \times 10^{10}$</td>
</tr>
<tr>
<td>Sodium</td>
<td>335. ± 14</td>
<td>2895.</td>
<td>$1.55 \times 10^7$</td>
<td>$5.36 \times 10^9$</td>
</tr>
</tbody>
</table>
**TABLE 20**

RESULTS OF THE M.I.T.R. THRESHOLD FOIL EXPERIMENTS

<table>
<thead>
<tr>
<th>THRESHOLD FOIL</th>
<th>NUMBER OF MEASUREMENTS</th>
<th>AVERAGE VALUE ( K_i = \int_0^{\infty} \phi_i(E) \sigma(E) dE )*</th>
<th>STANDARD DEVIATION OF THE MEAN</th>
<th>ESTIMATED ACCURACY (RMS)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{Np}^{237} ) (n, f)</td>
<td>5</td>
<td>( 9.69 \times 10^7 )</td>
<td>( \pm 8% )</td>
<td>( \pm 18% )</td>
</tr>
<tr>
<td>( \text{U}^{238} ) (n, f)</td>
<td>5</td>
<td>( 5.33 \times 10^6 )</td>
<td>( \pm 12% )</td>
<td>( \pm 20% )</td>
</tr>
<tr>
<td>( \text{Th}^{232} ) (n, f)</td>
<td>5</td>
<td>( 2.64 \times 10^4 )</td>
<td>( \pm 7% )</td>
<td>( \pm 19% )</td>
</tr>
<tr>
<td>( \text{In}^{115} ) (n, n')</td>
<td>4</td>
<td>( 1.66 \times 10^4 )</td>
<td>( \pm 2% )</td>
<td>( \pm 15% )</td>
</tr>
<tr>
<td>( \text{Ni}^{58} ) (n, p)</td>
<td>3</td>
<td>( 1.11 \times 10^5 )</td>
<td>( \pm 8% )</td>
<td>( \pm 13% )</td>
</tr>
<tr>
<td>( \text{Al}^{27} ) (n, p)</td>
<td>3</td>
<td>( 3.78 \times 10^4 )</td>
<td>( \pm 5% )</td>
<td>( \pm 25% )</td>
</tr>
<tr>
<td>( \text{Si}^{28} ) (n, p)</td>
<td>3</td>
<td>( 1.10 \times 10^5 )</td>
<td>( \pm 2% )</td>
<td>( \pm 50% )</td>
</tr>
<tr>
<td>( \text{Mg}^{24} ) (n, p)</td>
<td>3</td>
<td>( 4.35 \times 10^4 )</td>
<td>( \pm 1% )</td>
<td>( \pm 10% )</td>
</tr>
<tr>
<td>( \text{Fe}^{56} ) (n, p)</td>
<td>3</td>
<td>( 1.97 \times 10^4 )</td>
<td>( \pm 6% )</td>
<td>( \pm 12% )</td>
</tr>
<tr>
<td>( \text{Al}^{27} ) (n, a)</td>
<td>3</td>
<td>( 2.18 \times 10^4 )</td>
<td>( \pm 2% )</td>
<td>( \pm 10% )</td>
</tr>
</tbody>
</table>

* \( \text{sec}^{-1} \times 10^{24} \)
reactions are given in Table 20. These results were analyzed by the Weighted Orthonormal and Weighted Orthonormal Polynomial Methods to obtain the fast neutron spectrum. It was found that the Weighted Orthonormal Method was less sensitive to inconsistencies in the data than the Orthonormal Polynomial Method and is therefore preferred. It must be pointed out that these methods are integral in nature and, as such, are incapable of predicting fine structure from a few integral measurements. Observance of bumps or peaks in the calculated spectra is insufficient reason to expect that such features exist. However, for dosimetry purposes, only the gross features are important, and the methods are believed to be adequate in this case.

The Orthonormal Methods were tested before being applied to the MITR data. First, they were tested on "exact" data, which was made up by assuming a spectral shape and numerically integrating over the cross section curves with respect to this spectrum to obtain input activations. The results of one of these tests are shown in Figs. 64 and 65, which are examples of a $1/E^3$ spectrum calculated using a similar ($1/E$) and an exact ($1/E^3$) weighting function. It is to be noted that the use of a proper weighting function greatly improves the accuracy of the calculation, and also that the calculation does not provide information below approximately 1 MeV. Further, the calculated curve is not smooth, which is a reflection of the fact that the cross sections which make up the orthonormal functions are not generally smooth.

Next, the Orthonormal Methods were tested on data from the ISPRA-1 Reactor. This data had been previously analyzed by the Dierckx Method and the Italian Iterative Method. The results of the calculations with the ISPRA data are shown in Figs. 66 and 67 where the general agreement with the other methods may be noted.
Fig. 64  WEIGHTED ORTHONORMAL METHOD TEST USING EXACT DATA AND SIMILAR WEIGHTING FUNCTION, DIFFERENTIAL FLUX VS ENERGY
Fig. 65 WEIGHTED ORTHONORMAL METHOD TEST USING EXACT DATA AND EXACT WEIGHTING FUNCTION, DIFFERENTIAL FLUX VS ENERGY
Fig. 66 DIFFERENTIAL FLUX VS ENERGY, DATA FROM THE ISPRA-1 REACTOR, COMPARISON OF THE WEIGHTED ORTHONORMAL, ITALIAN ITERATIVE, AND DIERCKX METHODS
Fig. 67 INTEGRAL FLUX VS ENERGY, DATA FROM THE ISPRA-1 REACTOR (REF B-13), COMPARISON OF THE ORTHONORMAL METHOD AND THE EFFECTIVE THRESHOLD METHOD
The results of the Weighted Orthonormal Method calculation using the MITR data are shown in Figs. 68 and 69. It is to be noted that the spectrum is greatly depleted compared to a fission spectrum, which is most likely due to the presence of oxygen (D₂O) and bismuth. Many combinations and permutations of the ten data points were tried (which can be easily performed by the computer code). The result was an indication of the consistancy of the data. On this basis, it was found that the thorium point was perhaps 60% high, while the uranium point was perhaps 35% low. All of the other points were consistant within their accuracy of measurement except for the In¹¹⁵(n, n')In¹¹⁵m reaction. This was a factor of 2-3 low and leads one to suspect that the cross section curve is incorrect.

The composite neutron spectrum is shown in Fig. 70. Two possible joining functions are shown between the fast neutron spectrum and the epithermal neutron spectrum, since there is no experimental information available in this region. Dosimetry calculations were performed with these spectra, and some of the results of these calculations are shown in Figs. 71, 72, and 73. In particular, Fig. 72 compares the results of the gamma ray dosimetry calculation with some experimental results obtained in a polyethylene phantom using a polyacrylamide dosimeter. The agreement is viewed satisfactory considering that the depth dose curves are calculated for a broad beam of neutrons instead of a finite phantom.

E. Conclusions

The primary conclusions to be made from this work is that the Medical Therapy Beam cannot be successfully modified for therapy purposes simply by filtering the beam and using the epithermal neutrons. The beam is too highly thermal to use this procedure. However, if the beam
Fig. 68 MITR DIFFERENTIAL FLUX VS ENERGY, WEIGHTED ORTHONORMAL METHOD RESULTS (FUSE-3)
Fig. 69 MITR INTEGRAL FLUX VS ENERGY, COMPARISON OF THE EFFECTIVE THRESHOLD METHOD AND THE WEIGHTED ORTHONORMAL METHOD (FUSE-3)
Fig. 70 MITR COMPOSITE SPECTRUM, $\phi(u)$ VS LETHARGY
Fig. 71 INTEGRAL PROTON DOSE IN TISSUE FOR THE COMPOSITE MITR SPECTRUM, DOSE RATE VS LETHARGY
Fig. 72 GAMMA RAY DOSE RATE IN TISSUE DUE TO NEUTRONS FROM THE MITR MEDICAL FACILITY BEAM, DOSE RATE VS DEPTH
Fig. 73 INTEGRAL GAMMA RAY DOSE RATE IN TISSUE FOR THE COMPOSITE MITR SPECTRUM, DOSE RATE VS LETHARGY
were less thermal, without greatly affecting the fast neutron flux, then the use of a filtered epithermal beam would be practical.

In order to change the spectrum, material would have to be removed between the core and the beam port. This is a physics design problem, first to attempt to calculate a spectrum which agrees with the measured spectrum, and then to calculate the effects of modifications on both the spectrum and the reactor. If the Medical Therapy Beam is to be successfully modified, this design study should be undertaken.
IV. ACTIVATION ANALYSIS

A. Prompt Activation Analysis for Boron and Lithium (L. Clark, Jr.)

1. Introduction

Investigation in the field of artificially-induced radioactivity established at an early stage the fact that the radiation emitted by each nuclide was characteristic of that nuclide and could be used as a positive means of identification. This has led to a great deal of work in the field known as activation analysis, particularly during the past ten or fifteen years. Most of the work in this area has centered around activation by neutrons. The reasons for this are the lack of a Coulomb barrier against neutron penetration of the atomic nucleus and the practical availability of neutrons in the many nuclear reactors now being operated. Furthermore, neutron activation generally gives a radioactive product, most of which decay with emission of one or more gamma rays. Scintillation spectrometry provides a convenient and accurate method for measuring the energies and the decay rates of these gammas, thereby providing identification of their sources.

The well-investigated method of thermal neutron activation plus gamma spectrometry is not amenable, however, to activation of the light elements. This is indicated by the fact that a listing of 70 elements investigated by the Activation Analysis Group at Oak Ridge National Laboratory, using such methods, during a six-year period ending in 1959, contains no record of work on elements lighter than oxygen (45). Also, Koch (46) gives no references to analyses using the (n, γ) reaction for the light elements up to and including oxygen. The reasons are apparent from a study of the nuclear properties of the light elements. Only a few neutron
capture processes, essential to the formation of radioactive nuclides which decay by $\beta^-$ and gamma emission, are seen to occur. Where such reactions do occur--$^\text{15}_\text{N}(n,\gamma)^\text{16}_\text{N}$ and $^\text{18}_\text{O}(n,\gamma)^\text{19}_\text{O}$--the half-life is either very short or the yield is low.

For the light elements, $(n,p)$ and $(n,\alpha)$ reactions occur with thermal neutrons, but the radioactive end products decay only by $\beta^-$ emission with no gammas.

Other methods of activation analysis have helped to fill the void resulting from the inability of neutron activation plus gamma spectrometry to function in that part of the periodic table occupied by the light elements. Koch (46) lists many charged-particle reactions which may be, and in general have been, used to produce radioactive nuclides, which are then in turn identified and measured by various methods as they decay. Charged particles (protons, deuterons, tritons, and alphas) for these reactions are commonly obtained by means of accelerators, although sometimes they may be obtained indirectly by use of a reactor. Leddicotte, et al, (45) activated oxygen by using the thermal neutron reaction, $^\text{6}_\text{Li}(n,\alpha)^\text{3}_\text{H}$, to produce tritons, which in turn activated the oxygen, $^\text{16}_\text{O}(\text{H}^3,n)^\text{18}_\text{F}$, to produce 112-minute $^\text{18}_\text{F}$. The $\beta^+$ decay of $^\text{18}_\text{F}$ produces 0.511-MeV annihilation gamma rays, which are detected and counted. Other neutron capture reactions useful for the production of charged particles are $^\text{3}_\text{He}(n,p)^\text{3}_\text{H}$ and $^\text{10}_\text{B}(n,\alpha)^\text{7}_\text{Li}$. Alphas may also be obtained from the decay of naturally radioactive materials, such as radium or polonium.

Instead of measuring the radiation which comes from the radioactive product of a nuclear reaction, it is also possible to measure directly one or more of the products of such a reaction. Various means are available for detecting an emitted proton or alpha particle, such as photographic emulsions
or zinc sulfide screens with photomultiplier tubes. The purpose of this paper is to report on the design and operation of a detector to measure trace quantities of two of the light elements, lithium and boron, by direct counting of the prompt particles from the reactions, \( \text{Li}^6(n,\alpha)\text{H}^3 \), and \( \text{B}^{10}(n,\alpha)\text{Li}^7 \). These reactions have been used by Wanke and Monse (47) who employed a zinc sulfide screen with a photomultiplier tube to determine the isotopic abundance of \( \text{Li}^6 \), \( \text{B}^{10} \) and \( \text{U}^{235} \). The second reaction was also used by Fiti, Manteseu, and Costea (48) to measure boron in minerals, employing a somewhat similar detection system. In these investigations, thermal neutron fluxes on the order of \( 10^2 - 10^3 \text{ n/cm}^2\text{-sec} \) were obtained by moderating with paraffin or graphite the fast neutrons from Po-Be or Ra-Be sources. For the purposes of the present work, a detector was designed for operation in the neutron fluxes of the thermal column of the Massachusetts Institute of Technology Research Reactor (MITR) where \( 10^8 - 10^{10} \text{ n/cm}^2\text{-sec} \) are available.

2. Design Considerations

The design of the detector for measuring boron and lithium was based on a consideration (45) of the desirability of operating the detector in the reactor at moderately high neutron fluxes with a capability for frequent changing of samples and (46) of the various types of ionizing radiations which would be encountered.

From a radiation damage point of view, the fluxes are relatively low and did not impose stringent requirements on the materials to be used. However, the desirability of being able to change samples frequently did effect the choice of materials and the design. The use of complicated shielding devices was avoided by making the counter small and construct-
ing it primarily from materials which do not activate appreciably (e.g. polyethylene) or which have short half-lives (e.g. aluminum). (The use of a small detector was dictated basically by other considerations, as explained below.) This permitted removing the detector from the reactor after a brief cooling period, in order to change the sample. Materials like Cu were avoided entirely or kept to a minimum.

The design of the detector was based on a review of the types of radiation which would be encountered. These are summarized in Table 21. Items 1-4, the alpha, Li\(^7\), and H\(^3\) particles from the B\(^{10}(n,\alpha)Li\(^7\) and Li\(^6(n,\alpha)H\(^3\) reactions are the ionizing irradiations to be detected and measured as an indication of the amounts of boron or lithium present. Items 5-11 are interfering radiations which will constitute the background in the detector. Electronic noise is another item which might contribute to the background, but this is not expected to be an important factor since only heavy ionizing particles are to be counted.

The design objective was to maximize the counter response to Items 1-4 while minimizing its response to Items 5-11. It was decided to use a proportional counter for this work, since it preserves the relationship between the particle energy and the detector output pulse height and because its gas multiplication would reduce the amount of electronic amplification required. A gas-flow type was required due to the need for frequent sample changing.

In order to maximize the response to the heavy ionizing particles to be counted, it is desirable that the particle expend all of its energy within the sensitive volume of the detector. Table 22 lists the product particles resulting from the B\(^{10}(n,\alpha)Li\(^7\) and the Li\(^6(n,\alpha)H\(^3\) reactions and,
TABLE 21

SOURCES OF IONIZING RADIATION IN THE DETECTOR

<table>
<thead>
<tr>
<th>Item</th>
<th>Particle</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\alpha$</td>
<td>B to be measured, by reaction $\text{B}^{10}(n,\alpha)\text{Li}^7$</td>
</tr>
<tr>
<td>2</td>
<td>$\text{Li}^7$</td>
<td>B to be measured, by reaction $\text{B}^{10}(n,\alpha)\text{Li}^7$</td>
</tr>
<tr>
<td>3</td>
<td>$\alpha$</td>
<td>Li to be measured, by reaction $\text{Li}^6(n,\alpha)\text{H}^3$</td>
</tr>
<tr>
<td>4</td>
<td>$\text{H}^3$</td>
<td>Li to be measured, by reaction $\text{Li}^6(n,\alpha)\text{H}^3$</td>
</tr>
<tr>
<td>5</td>
<td>Electrons</td>
<td>Primarily Compton recoils from capture, decay, reactor core, and other gamma rays.</td>
</tr>
<tr>
<td>6</td>
<td>Beta rays</td>
<td>Decay of radioactive nuclides (such as $\text{Al}^{28}$) formed in matrix of sample or in structural materials of detector.</td>
</tr>
<tr>
<td>7</td>
<td>$\alpha$, $\text{Li}^7$, $\text{H}^3$</td>
<td>B and Li present as impurities in structural materials of detector.</td>
</tr>
<tr>
<td>8</td>
<td>$\alpha$</td>
<td>Uranium and other naturally radioactive nuclides present as impurities in sample and in structural materials of detector.</td>
</tr>
<tr>
<td>9</td>
<td>Fission fragments</td>
<td>Uranium present as an impurity in the sample and in structural materials.</td>
</tr>
<tr>
<td>10</td>
<td>Recoil nuclei</td>
<td>Collisions between fast neutrons and light nuclei of sample and structural materials.</td>
</tr>
<tr>
<td>11</td>
<td>$\alpha$, $\text{Li}^7$, $\text{H}^3$</td>
<td>Contamination of sample or other interior surfaces with impurities containing boron or lithium.</td>
</tr>
</tbody>
</table>
TABLE 22

KINETIC ENERGY AND RANGES FOR PRIMARY PARTICLES

<table>
<thead>
<tr>
<th>Isotope and Natural Abundance</th>
<th>Reaction and Cross Section</th>
<th>Product Particles</th>
<th>Kinetic Energy (MeV)</th>
<th>Range in Air (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{B}^{10} ) 18.7</td>
<td>( \text{B}^{10} (n,\alpha)\text{Li}^7 ) 4000 b.</td>
<td>( \alpha ) 7 %</td>
<td>1.78</td>
<td>0.89</td>
</tr>
<tr>
<td></td>
<td></td>
<td>( \text{Li}^7 ) 7 %</td>
<td>1.47</td>
<td>0.73</td>
</tr>
<tr>
<td></td>
<td></td>
<td>( \text{Li}^7 ) 93 %</td>
<td>0.84</td>
<td>0.24</td>
</tr>
<tr>
<td>( \text{Li}^6 ) 7.5</td>
<td>( \text{Li}^6(n,\alpha)\text{H}^3 ) 945 b.</td>
<td>( \alpha ) 100 %</td>
<td>2.06</td>
<td>1.03</td>
</tr>
<tr>
<td></td>
<td></td>
<td>( \text{H}^3 ) 100 %</td>
<td>2.73</td>
<td>6.02</td>
</tr>
</tbody>
</table>

TABLE 23

ESTIMATED COUNT RATES

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Particle</th>
<th>Energy (MeV)</th>
<th>Range in Al (cm)</th>
<th>Counting Efficiency</th>
<th>Counting Rate CPM cm² Particle</th>
<th>Counting Rate CPM cm² Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{B}^{10} (n,\alpha)\text{Li}^7 )</td>
<td>( \alpha )</td>
<td>1.47</td>
<td>0.45 x 10⁻³</td>
<td>0.0304</td>
<td>76</td>
<td></td>
</tr>
<tr>
<td></td>
<td>( \text{Li}^7 )</td>
<td>0.84</td>
<td>0.15 x 10⁻³</td>
<td>0.0101</td>
<td>25</td>
<td>101</td>
</tr>
<tr>
<td>( \text{Li}^6(n,\alpha)\text{H}^3 )</td>
<td>( \alpha )</td>
<td>2.06</td>
<td>0.64 x 10⁻³</td>
<td>0.0432</td>
<td>16</td>
<td></td>
</tr>
<tr>
<td></td>
<td>( \text{H}^3 )</td>
<td>2.73</td>
<td>3.7 x 10⁻³</td>
<td>0.250</td>
<td>92</td>
<td>108</td>
</tr>
</tbody>
</table>

Basis: 1 ppm boron or lithium

\( 10^8 \) neutrons /cm²-sec
in particular, the ranges in air of these particles. Fortunately, the α and Li\(^7\) particles have ranges of 1 cm or less, and only the H\(^3\) has a longer range, 6 cm. The actual ranges in the detector will be somewhat different, depending upon the counting gas to be used.

In order to reduce interference from boron and lithium which might exist in the structural materials of the counter, it was decided that the sample material to be assayed should be distributed on the inner surfaces of the detector cavity in a layer thick enough to absorb the unwanted alpha, H\(^3\), and Li\(^7\) particles coming from the structural materials. For most of the experimental work reported on in this paper, aluminum was used as the matrix material with known amounts of boron or lithium mixed in it. Aluminum was selected because it is commonly used for structural purposes in reactors, because its impurity content—particularly those impurities having large neutron absorption cross sections—is of importance to reactor designers; because it is a good electrical conductor, and because standards containing known amounts of boron or of lithium were readily available.

The general type of detector having been determined, it is now possible to estimate the counting rates which trace amounts of boron or lithium in aluminum can be expected to produce. The interfering radiations, which will limit the sensitivity of the detection system, will then be considered, and an estimate made of the counting rates which might result from each.

Table 23 again lists the particles to be counted. It also gives the ranges in aluminum, the longest being \(3.7 \times 10^{-3}\) cm for H\(^3\). The counting efficiency in the next column is based on a layer thickness equal to the H\(^3\) range. The efficiency for counting the α and Li\(^7\) particles is much less due to their comparatively short ranges. For purposes of estimating the counting rates to be expected, calculations were based on boron or lithium con-
centrations of 1 part per million and on a thermal neutron flux of $10^8$ n/cm$^2$-sec. The final columns indicate that count rates in excess of 100 counts per minute/cm$^2$ of surface can be expected. This should prove ample, even if the counter volume is small, provided that the background can be kept low.

The electron and beta rays, Items 5 and 6 of Table 21 will traverse the counting chamber in great quantities, but they have a comparatively low specific ionization. This will vary considerably, depending upon whether the electron has an energy of several MeV or is close to the end of its range when it crosses the chamber. Based on work by E. J. Williams, as reported by Evans (49), on the relationship between range and path length for 19.6 keV electrons in oxygen, it is estimated that an electron will lose 32 keV in its last centimeter of range. This, on the average, will be the maximum energy loss by electrons. The minimum energy loss occurs at about 1.5 MeV and is calculated to be approximately 2 keV/cm. Because of scattering and branching, some electrons will give up more than 32 keV in the chamber, but even these will give small pulses in comparison with the 1.47-MeV alphas from 93% of the $^{10}$B(n,α)$^{7}$Li reactions, and it should be possible to effectively discriminate against them. It is apparent, however, that the pulse heights for wanted and unwanted pulses will be in the best ratio when the sensitive volume of the detector has dimensions approximately equal to the maximum ranges of the alpha and other particles of interest.

The background due to Item 7, the α, Li$^7$, H$^3$ particles entering the chamber as a result of (n,α) reactions by boron and lithium present as impurities in the structural material of the detector, was partially
overcome by lining the chamber with the material to be assayed, as was mentioned earlier. This was accomplished by lining the cavity with a cylindrical sleeve, and will be described in more detail later. The anode and its support, if not relatively free of boron and lithium, could contribute substantially to the background.

Using the rule of thumb that most substances contain an average of about $10^{-12}$ curies of naturally radioactive nuclides per gram of material, it was calculated that the count rate due to alphas from this source would not exceed 0.2 counts/hr-cm$^2$ of chamber surface. Background counting with the finished detector indicated that this was the right order of magnitude and, therefore, not important in this application.

In background from the $\text{U}^{235} (n,f)$ reaction, Item 9 of Table 21 was calculated to be of the order of 1 count/hr-cm$^2$ of chamber surface at a flux of $10^8$ n/cm$^2$-sec. This was the approximate magnitude of the measured count rate due to fission fragments from $\text{U}^{235}$ in the aluminum used for the tests and it could become a problem if the method is extended to boron and lithium concentrations below 1 part per billion.

The recoil nuclei, Item 10 of Table 21, which result from collisions between fast neutrons and light nuclei must also be considered due to the fact that the neutron energy spectrum contains many neutrons which have not been completely thermalized. It is difficult to estimate this contribution to the background with any accuracy due to lack of detailed information about the high energy end of the neutron spectrum in various parts of the MIT reactor. Table 24 shows the maximum and average energies transferred to the nuclei of several elements of interest by a 0.72-MeV neutron, this being the most probable energy of the neutron fission spectrum for
TABLE 24

KINETIC ENERGIES OF RECOIL NUCLEI

<table>
<thead>
<tr>
<th>Recoil Nucleus</th>
<th>Atomic Mass</th>
<th>Max. Energy Transferred (MeV)</th>
<th>Average Energy Transferred (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>1</td>
<td>0.72</td>
<td>0.36</td>
</tr>
<tr>
<td>C</td>
<td>12</td>
<td>0.20</td>
<td>0.10</td>
</tr>
<tr>
<td>O</td>
<td>16</td>
<td>0.16</td>
<td>0.08</td>
</tr>
<tr>
<td>Al</td>
<td>27</td>
<td>0.10</td>
<td>0.05</td>
</tr>
<tr>
<td>A</td>
<td>40</td>
<td>0.068</td>
<td>0.034</td>
</tr>
<tr>
<td>Au</td>
<td>197</td>
<td>0.014</td>
<td>0.007</td>
</tr>
</tbody>
</table>

Basis: Calculations based on a neutron energy energy of $E_0 = 0.72$ MeV (most probable energy for fission spectrum)
When it is recalled that 21% of the fission neutrons are in excess of 3 MeV, and 1% are in excess of 7 MeV, (or more than 10 times the most probable energy) (50), it will be seen that the energies imparted to the struck nuclei, even heavy ones, are not negligible in comparison with the kinetic energies of the $\alpha$, $\text{Li}_7$, and $\text{H}_3$ particles in Table 22. A very rough estimate was made for a counter with an aluminum lining based on measurements of the leakage neutron energy spectrum by Anderson (51) with the MIT Fast Chopper. The calculation showed that the count rate at the inner end of a radial port in the MITR (just outside the heavy water core tank) might be as high as 100 counts/cm$^2$-sec. Although only a rough approximation, the estimate shows the desirability of eliminating the light elements, particularly hydrogen, from sensitive regions of the detector and of operating the counter in a region where the neutron flux has been thermalized as much as possible.

The problem of background due to boron and lithium contamination of the sample surfaces, Item 11 of Table 21, is one of cleanliness in preparing the samples and other interior surfaces of the detector.

In general, the background and the signal are both proportional to the surface area of the chamber. Therefore, there are no advantages to be gained by increasing the size of the sensitive volume of the detector beyond the ranges of the heavy particles in the counting gas. Increasing the size of the detector beyond this will simply increase the electron and beta pulse heights, making it more difficult to discriminate against them.

The counting gas used was 90% argon-10% carbon dioxide. This was selected because of the high atomic weight of argon and the relatively
high weights of the other constituents, the relatively low operating voltage which results from the addition of the CO\textsubscript{2}, the chemical inertness of all the constituents, and the reasonably low neutron activation when used in a small chamber.

The ranges of α, H\textsuperscript{3}, and Li\textsuperscript{7} in 90\% A-10\% CO\textsubscript{2} are 16\% greater than the ranges shown in Table 22 for these particles in air. Except for the tritons, the maximum range is 1.2 cm, and so 1 cm was chosen as the diameter for the counting chamber. As will be seen later, it turns out to be advantageous to size the chamber so that the tritons give up only a fraction of their energies in the sensitive volume.

3. Description of Counter

The counter which resulted from the above design considerations and which was used for most of the experimental work in this investigation is shown in Fig. 74. It is cylindrical in shape, has a diameter of 0.610", because it was necessary that it fit into a 0.620" bent tube in the thermal column of the MITR. This proved just large enough to accommodate a cavity with a diameter of 1 cm. The anode is a 0.002 tungsten wire 1-cm long, so that the sensitive length of the detector is approximately 1 cm.

In order to permit sample changing, the anode is supported at only one end. The aluminum plug at the right hand end of the drawing unscrews so that a sample of the size shown in Fig. 74b may be inserted into the cavity. In this manner the sample material itself forms the inside surface of the counting chamber and almost completely surrounds the anode. It was found that the anode wire was stiff enough for this application, but it had the disadvantage that the wire was not stretched tight and was not supported precisely on the center axis. A spherical covering was provided
Polyolefin Shrink fit tubing (Thermo fit CR) Al tube for counting gas (1/8" O.D.)

Type 1100 Al shell Al₂O₃ hard surface (Duralectra) Gas passages

Removable plug for changing samples

ALJAK Coax No. 21-607 (0.325" O.D.)

Polyethylene O-ring Teflon .002" Tungsten tip, aquadag coating

Fig. 74a DRAWING OF GAS-FLOW PROPORTIONAL COUNTER

Fig. 74b STANDARD SAMPLE

0.497" diam. 0.031" hole for gas flow

SCALE 1:2

Fig. 74c CADMIUM-WRAPPED SAMPLE
to cover the sharp end of the anode wire which otherwise would be a region of very high gas multiplication. It was found that sulfur had the property of adhering to the wire and of assuming a spherical shape when melted. It was covered with aquadag to make it conducting.

The shell of the counter was machined from Type 1100 aluminum. In order to avoid galling of the outer surface, it was covered with a hard coating of aluminum oxide, 0.002" thick, by an electrolytic process known as "Duralectra."

A combination of aluminum and polyethylene plug screws into the left hand end of the shell. A 7-ft. length of Aljak No. 21-607 coaxial cable is fixed in position in the aluminum portion of this plug. This cable has an aluminum jacket which, while providing some flexibility, reduces the activation which would otherwise result in standard coax when it is exposed to neutrons. The center conductor is copper, but it did not build up to troublesome activities during the test runs. The center conductor passes through the polyethylene portion of the plug to an aluminum rod which serves to join it electrically to the anode and also to support the latter in position. This support extends about 1 cm into the cavity, so that heavy ionizing particles from boron or lithium impurities at the end of the chamber will not reach the sensitive portion. A teflon wafer separates the polyethylene from the chamber, so that recoil protons cannot enter.

The counting gas is brought to the chamber through one of a pair of 1 8"O.D. aluminum tubes. The gas passes through a drill hole in the polyethylene to a circular groove in the face of the plug. 0.031" diameter holes are provided in the teflon wafer and are drilled at an angle so that any knock-on protons passing through them will be directed toward the
anode support. Electrons from any ionization which occurs inside the chamber should be collected without multiplication on the anode support. The gas then passes through the chamber, out through a 0.031" hole at the end, through passages in the end plug, and back around the outside of the chamber by means of several grooves cut on the inside surface of the counter shell. It leaves the counter through a circumferential groove in the polyethylene, two drill holes, and the other piece of aluminum tubing. Buna N O-rings provide gas seals at both ends of the counter shell. The gas was under a pressure of a few inches of water. Epoxy resin was used to improve the gas tightness of the joint where the coax and the aluminum tubing enter the counter. Expanded polyolefin tubing (Thermofit CR) was heat shrunk over the coax and aluminum tubing, which provided a smooth, non-galling surface and which made it easier to slide the counter through the bent tube which passes through the reactor shielding.

Most of the samples used in the test runs were made from aluminum spectrochemical standards, purchased from the Aluminum Company of America. One set of standards contained known amounts of boron, ranging from 5 to 290 parts per million. A second set contained lithium ranging from 5 to 280 parts per million. The Company would not specify the lithium content of the boron standards or the boron content of the lithium standards. Also, it would provide no information regarding the probable error in the analysis. Therefore, in order to cover both of these situations, it was arbitrarily assumed that there was a standard error of 5 parts per million in all of the analyses furnished by the Company.

Two cylinders of the type shown in Fig. 74b were machined from each of the standards. Since the surfaces undoubtedly became contaminated during the machining operations, the finished pieces were etched for about
5 minutes at 70-75°C in a 3.7% hydrochloric acid solution. They were rinsed with distilled water, soaked in acetone, given a final rinse in acetone, drained and dried.

In addition to the aluminum standards described above, cylinders were also made from an aluminum blank obtained from ALCOA and from a 99.9999% aluminum ingot, the boron and lithium contents in both cases being unknown.

In hopes of finding a material which would have a lower boron or lithium content than the aluminum, graphite from two sources was used for making additional samples. Two pieces were machined from a bar of reactor grade graphite. In addition, two bars of graphite manufactured by National Carbon Company for use as spectroscopic electrodes were used to make additional samples.

In order to determine, if possible the extent of the background caused by recoil nuclei, the thermal flux entering the cavity was reduced as much as possible by wrapping several of the standards in 0.020" thick cadmium. A cylinder of this type is illustrated in Fig. 74c.

The characteristics of the detector were determined with an alpha source which had been made by depositing about $10^{-4}$ grams of $^{235}$U in the form of $\text{U}_3\text{O}_8$ on an aluminum foil, which was then fitted to the standard samples described earlier. Figure 75 shows the count rate-operating voltage curve for the detector. The counter was operated at 1400 volts, where the curve has an alpha plateau of 15% per 100 volts.

Figure 76 shows the energy spectrum which was obtained with the above $^{235}$U alpha source. The peak which occurs at Channel No. 32 helps to provide an energy calibration for the differential analyzer. It was calculated that an alpha particle which travels straight across the
Fig. 75 COUNT RATE - OPERATING VOLTAGE CHARACTERISTIC CURVE FOR GAS-FLOW PROPORTIONAL COUNTER
Fig. 76 ENERGY SPECTRUM FOR U-235 ALPHA SOURCE
chamber should give a 0.94-MeV pulse. This fact and the end points in the energy spectra for alphas and tritons from boron and lithium were averaged to obtain a calibration of 0.026 MeV per channel.

4. Operation of Counter

The counter was operated in the neutron flux at the very end of the MIT reactor thermal column. A horizontal cross section of the reactor is shown in Fig. 77. The original thermal column has been extended by adding a heavy water exponential facility on that side of the reactor. Neutrons from the core diffuse through 52" of graphite to an empty space at the end of the thermal column which has the shape of a parallelepiped and which has dimensions of several feet on each face. This empty space, or hohlraum, is lined on each side with about one foot of graphite. Its purpose is to cause neutrons to diffuse upwards into the heavy water exponential tank which is shown in the figure. This tank is not on a level with the reactor core, as is indicated by the figure, but rather it is several feet above it.

The bent tube which was mentioned earlier is located in a shielding plug in the port labeled 12CH1. It is 70-1 2" long and extends through the shielding, through the graphite lining of the hohlraum, which is 16" thick at this point, and then into the hohlraum cavity. The counter was positioned so that it was in the graphite region, where the thermal flux is approximately $10^8$ n/cm$^2$-sec, for the major part of the work.

For most of the experimental runs, it was found extremely helpful to know the energy spectrum of the ionization within the counter. After amplification in a Baird Atomic Model No. 255 Preamplifier, the signal was analyzed in a Technical Measurements Corporation 256 Channel Analyzer.
Fig. 77 HORIZONTAL CROSS SECTIONS THROUGH THE REACTOR AND EXPONENTIAL FACILITY.
This consisted of a Digital Computer Unit, Model - CN-110-A256, with a Pulse Height Logic Unit, Model 210, and a Data Output Unit, Model 220. Information was printed by a Hewlett-Packard Digital Recorder, Model 561B.

5. Results

Figure 78 shows the total counting rate obtained for the various spectrochemical aluminum standards which contain varying amounts of boron and lithium in the range from 5 to 290 parts per million. Although the natural abundance and the \( (n,\alpha) \) cross-section for \( \text{Li}^6 \) are 7.5% and 945 barns respectively, as compared to 18.7% and 4000 barns for \( \text{B}^{10} \), the count rate for a given amount of lithium is higher than that for boron. This is due to the greater range, and consequently greater counting efficiency, of the tritons. With regard to the errors of measurement indicated at the upper ends of curves, it was noted that there often existed a much larger difference in the count rate between two apparently identical samples than could be accounted for by the probable error in the counting. This may have been due to differences in the amount of etching between the two samples, or possibly due to segregation of the boron or lithium in the aluminum. This is an area which could well bear further investigation. The overall count rate for boron is \( 130 (\pm 3) \text{ CPM PPM} \) and, for lithium, \( 169 (\pm 4) \text{ CPM PPM} \). This is the same order of magnitude as the estimates of Table 23. These estimates were based on \( 1 \text{ cm}^2 \) of surface area, as compared to about \( 3 \text{ cm}^2 \) effective area in the detector, and also on the assumption that every particle which entered the chamber would produce a pulse large enough to be counted.

The energy spectra for the various samples is of considerable interest. Figure 79 shows the spectra for the boron containing standards.
Fig. 78 PLOT OF MEASURED COUNT RATES FOR ALUMINUM STANDARDS CONTAINING KNOWN CONCENTRATIONS OF BORON OR LITHIUM (MAY 14 AND JUNE 6, 1963)

FLUX: $1.14 \times 10^8$ N/CM$^2$-SEC
Fig. 79  ENERGY SPECTRA FOR ALUMINUM STANDARDS CONTAINING INDICATED AMOUNTS OF BORON (MAY 14 AND JUNE 6, 1963)
Fig. 80 ENERGY SPECTRA FOR ALUMINUM STANDARDS CONTAINING INDICATED AMOUNTS OF LITHIUM (May 14 and June 6, 1963)
Fig. 81 ENERGY SPECTRA FOR ALUMINUM STANDARDS CONTAINING BORON OR LITHIUM AS INDICATED

- 290 PPM BORON
- SLOPE: - 1.37/MEV

- 280 PPM LITHIUM
- SLOPE: - 4.46/MEV
The curves are almost a straight line up to about 1 MeV, and the slopes for the different concentrations are almost identical, except for the sample with five parts per million, where the slope appears to be a little greater.

Figure 80 shows similar energy spectra for the lithium standards. It is immediately obvious that the shape of the curves is quite different from those of the boron. In particular, the slope of the straight portions of the curves between about 0.3 MeV and 0.8 MeV are much steeper, and there is a fairly sharp bend in the curves at about 1 MeV. About 1.5 MeV the curves turn downward again and, like the boron, drop quickly to zero. The steep slope for the low energy half of the curves is a result of the fact that the tritons, although possessing a maximum kinetic energy of 2.06 MeV, will give up a maximum of only 0.8 MeV, approximately, in traversing the chamber.

Again, the shapes of the curves are very similar with the exception of the 5 PPM sample. On the basis of the slopes, it appears that the 5 PPM lithium standard also contains a detectable amount of boron, while the 5 PPM boron sample also contains a detectable amount of lithium.

In order to bring out the differences in the shape of the curves for boron and compared to the shape of those for lithium, Fig. 81 repeats the energy spectra of the 290 PPM boron sample and the 280 PPM lithium sample. The former has a logarithmic slope of \(-1.37/\text{MeV}\) while the straight portion of the lithium curve has a slope of \(-4.46/\text{MeV}\). The difference in the slopes provides an opportunity for determining the ratio of the amounts of boron and lithium where both elements are present as impurities in a material. A table has been constructed for the detector used in this work which gives the slopes of the curves to
be expected for mixtures of these two elements and the total count rates per part per million which such mixtures would give. By reading the slope of the 5 PPM of Fig. 76, and comparing it with slopes calculated for various mixtures of boron and lithium, we estimate that the sample has a ratio of boron to lithium of 3.8 to 1. Based on the count rate, it appears that the concentrations are about 3.2 PPM B and 0.9 PPM Li. Likewise, it is estimated that the 5 PPM Li standard contains about 5.6 PPM Li and 2.1 PPM B.

In order to obtain some indication of the sensitivity of this detection method, measurements were made on samples machined from two different sources of aluminum and on samples made from two different sources of graphite. Figure 82 shows the energy spectra for the aluminum samples. The aluminum for the upper curves was obtained from ALCOA, but no information concerning the boron and lithium concentration could be obtained. The slope of this curve is slightly greater than that for the boron standards, and it is estimated that the aluminum contains 3 PPM boron and 0.4 PPM lithium. The lower curve is for the 99.9999% aluminum ingot, and the measurements here indicate that it contains approximately 0.4 PPM boron and 0.6 PPM lithium.

Figure 83 shows the results for the graphite samples. The upper curve is for reactor grade graphite, and the lower one is for the spectroscopic electrode material. The low energy end of these curves reveal a shape which was not evident in the curves for the aluminum samples. The count rates are nearly double what would be expected from the $\text{B}^{10}(n,\alpha)\text{Li}^7$ and the $\text{Li}^6(n,\alpha)\text{H}^3$ reactions. It is possible that the increased count rate in this region is due to the recoil of carbon nuclei after collisions with fast neutrons, although an
Fig. 82 APPROXIMATE BORON AND LITHIUM CONCENTRATIONS AS DERIVED FROM ENERGY SPECTRA OF TWO ALUMINUM UNKNOWNS.
Fig. 83 APPROXIMATE BORON AND LITHIUM CONCENTRATIONS AS DERIVED FROM ENERGY SPECTRA OF TWO GRAPHITE UNKNOWNS
Fig. 84  LOG-LOG PLOT OF STANDARD AND UNKNOWN SAMPLES (MAY 14 AND JUNE 6, 1963)
experimental run described below appears to be inconsistent with the explanation.

The slopes of the straight portions of the graphite curves indicate that the reactor grade graphite contained approximately 1 PPM boron and 0.3 PPM lithium while the electrode material contained about 0.4 PPM boron and 0.2 PPM lithium.

Figure 84 is a log-log plot of the total count rate versus parts per million of boron or lithium, and it collects the results of all the above measurements. The boron and aluminum standard samples are plotted along the solid portions of the lines. The unknown aluminum and graphite samples are plotted at the upper end of the dotted portions of the lines. The points for the unknown graphite samples lie slightly above the lines because of the extra counts which were noted at the low energy end of the spectrum curves.

In order to determine the magnitude of the background count rate due to secondary electrons, betas, recoil nuclei, and other ionizing particles the number of which would be independent of the thermal neutron flux level, samples of the purest aluminum and graphite measured were wrapped in cadmium, as described earlier, and then run under conditions which were otherwise similar to those pertaining to the standard samples. A similar test on samples containing 290 PPM boron and 280 PPM lithium had shown that the cadmium wrapping would reduce the count rate to approximately 1% of the level without the cadmium. For the relatively pure aluminum and graphite, however, the count rates dropped to only about 5% of the count rate without the cadmium. These count rates are indicated by the arrows on the lower portion of Fig. 84.

Wrapping the sample in cadmium should eliminate only those
counts which are due to the boron and lithium in the matrix material itself, in the anode or other structural material inside or close to the sensitive volume, or in impurities which were introduced during preparation of the samples. (The counts due to fission fragments would also be eliminated by this, but these were only 0.02 CPM or considerably less than the count rates under consideration here). It is impossible to determine how much, if any, of the counting rate for the standard and unknown samples was due to impurities in the structural materials of the counter or due to impurities introduced during sample preparation. The differences detectable between samples would indicate that the former was responsible for very little background. It is difficult to see how the machining of graphite with clean tools could introduce boron or lithium as impurities into the cavities of these samples. Therefore, it appears that the reduction in count rate to as low as 6 CPM, due to wrapping in cadmium, must be due to the shielding of boron and lithium in the sample matrix material from thermal neutrons. If this is the case, and if appropriate standards of cleanliness can be maintained, it should be possible to make analytical determinations of boron and lithium at concentrations as low as 0.05 PPM.

The results of the test run made with a graphite sample wrapped in cadmium appear to be in conflict with the explanation given above for the increase in count rate on graphite at the low energy end of the spectrum curve. For pulses around 0.25 MeV, the count rate for the cadmium-wrapped graphite was only 0.5 CPM channel, whereas the apparent increase in count rate for the unwrapped graphite samples was on the order of several counts per minute.
6. Summary

Successful measurements have been made of boron and lithium present in trace amounts in aluminum for concentrations down to 5 PPM and it is believed that the method is reasonably accurate for concentrations a factor of 10 below this, as a result of measurements made on samples of aluminum and graphite containing low, but unknown, amounts of boron and lithium. Measurements made on samples of aluminum and graphite wrapped in cadmium, to simulate the effect of materials with still lower boron and lithium contents, indicate that the method should be practical at least down to levels of 0.05 PPM boron or lithium. Even at a flux level of $10^8$ n/cm$^2$-sec, the count rates are reasonably high compared to background, and it is possible that they could be increased by utilizing a higher flux level before the pileup of beta pulses places a limit on the sensitivity of the system. Considerable work still remains to be done in determining the effect, not only of pulse pileup, but of count rates due to recoil nuclei and the effect of sample surface preparation. Since only aluminum and graphite were used in this study, there is a need for further work to investigate the possibility of extending this method to other materials.

B. Fast Neutron Activation Analysis (M. Ahsan)

This study was done to determine the sensitivity of activation analysis using the (n, 2n) reaction and the equipment available at M. I. T. The neutron source used was a Texas Nuclear accelerator which produced 14-MeV neutrons by the (d, t) reaction at a maximum source strength of $10^{11}$ n/sec. The detection system consisted of 2 6" diameter by 3" thick NaI crystals as described for the foil counting in Chapter II of this report. The discriminator windows were set to accept 0.51 MeV annihilation radia-
tion from the positron activity resulting from the 

\((n, 2n)\) reaction. The two crystals were also operated in coincidence which greatly reduced the background caused by nonpositron activities produced by the fast neutrons.

Using this accelerator and detection system it was determined that the following elements, N, F, K, Zn, Ga, Br, Mo, Ag, and Pe could all be detected at levels between \(10^{-6}\) to \(10^{-7}\) gms, assuming interfering reactions were not important. The most common interfering reactions are proton reactions, i.e., \((p, n)\), \((p,\alpha)\), \((p, \gamma)\), etc. The energetic protons are produced by elastic neutron scattering when hydrogen is present in the sample. Because water is often present in the sample this can be an important problem. In most cases no other neutron reaction causes interference.

The element studied most carefully was nitrogen since it is not suitable for thermal neutron activation. In this case the 10 sec positron activity of \(^{13}\text{N}\) was measured. The sample used was ammonium carbonate. It was found that a sample containing \(100 \times 10^{-3}\) gm of \((\text{NH}_4)\text{CO}_3\) was irradiated to saturation giving a coincidence counting rate of about 300 cpm.
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**ABSTRACT**

The preparation of lithium drifted solid state Mass. detectors having active volumes between 0.5 cm$^3$ and 17 cm$^3$ is described. Results of measurements of neutron capture γ-ray spectra with these detectors is reported. Capture γ-ray spectra of Sc-45, Ir-193, Rh-103, Dy-161, Dy-164, and Ho-165 in the energy range below 1 MeV as measured with a six meter bent quartz crystal spectrometer have been measured and this data in combination with coincidence measurements of the high energy part of the spectrum using a scintillation triple coincidence pair spectrometer have been used to construct proposed energy level schemes for Sc-46, Dy-165, and Ho-166. A technique for the nondestructive analysis of spent reactor fuel using Ge(Li) detectors is described. Determination of burnup, irradiation time and flux done on the MIT reactor fuel are good to +10%. A method for determining fast neutron spectra from foil activation data is developed. It has been used to determine the neutron spectra and dose in the MITR medical therapy beam. A prompt activation analysis technique for lithium and boron sensitive to 1 part per million is reported.
1. Activation analysis
2. Capture γ rays
3. γ-ray spectroscopy
4. Neutron dosimetry
5. Neutron spectroscopy
6. Ge(Li) detectors
7. Prompt activation analysis
8. Energy levels
9. Spent fuel analysis
10. Burnup determination
11. Bent crystal
12. Pair spectrometer

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