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ABSTRACT

The sensitivity of large NaI(Tℓ) detectors to background radiation inhibits their use in measuring partial neutron capture cross sections in the keV energy region. Major sources of neutron induced background are the iodine in the crystal and iron in structural materials. In a survey of possible neutron shielding materials, a 70 percent by weight mixture of boron carbide in paraffin most readily reduced keV neutron fluxes. The use of a multi-layered shield assembly, incorporating an inner sleeve of boron carbide and lead shot in a litharge/glycerine cement, gave a factor of 10 improvement in the counts-to-background ratio at the 37 keV resonance in ^{48}Ti.

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BACKGROUND; BORON CARBIDES; CAPTURE; CROSS SECTIONS; GAMMA SPECTRA; GLYCERIN; KEV RANGE 10–100; LAYERS; LEAD; LITHARGE; NEUTRON FLUX; NEUTRONS; PARAFFIN; RADIATION DETECTORS; SCINTILATION COUNTERS; SENSITIVITY; SHIELDING; SODIUM IODIDES; THALLIUM; THICKNESS; TIME OF FLIGHT METHOD

CONTENTS

1. INTRODUCTION

The 8 in. x 6 in. NaI(Tℓ) detector in the 3 MeV Van de Graaff laboratory is used to measure relative partial neutron capture cross sections for high energy γ- ray transitions. The technique makes use of the kinematics of the ^{7}Li(p,n) reaction in which neutrons in the energy range 5-100 keV are produced in a collimated cone. Neutron energy is measured by time-of-flight and γ-ray pulse height spectra are recorded so that the time spectra for individual γ-ray transitions can be measured. Similar detectors have been used previously (Bird, Gibbons and Good 1962, Bergqvist and Starfelt 1961), however sensitivity to background radiation has been a factor which has inhibited their use at flight paths in excess of 50 cm. Longer flight paths (up to 2 m) were needed to obtain the neutron energy resolution required to separate individual resonances.

2. SOURCES OF BACKGROUND

Associated with the (p,n) reaction in the lithium target there is also the (p,γ) reaction which produces γ-rays up to an energy of 17 MeV. These γ-rays occur at zero time-of-flight for the neutrons and serve as the time reference. Apart from the (p,γ) events in the target and the (n,γ) events in the capture sample, any event recorded by the detector between successive beam pulses forms part of the background. The background events can be divided into beam dependent and beam independent events and are characterized by the form of the radiation incident on the detector.

2.1 Neutrons

The maximum current in the neutron energy spectrum from the ^{7}Li(p,n) reaction occurs near 30 keV so that the importance of individual elements as sources of neutron induced background will largely depend on their neutron capture cross sections at this energy. 30 keV Maxwellian-averaged capture cross sections, $\sigma_{n,\gamma}$ (< 30 keV >), for several elements are shown in Table 1. The high capture cross section for iodine makes the NaI crystal an important source of neutron induced background. Furthermore, summing of individual pulse heights following neutron capture in the iodine of the crystal, results in a pulse height spectrum corresponding to that of 6.82 MeV γ-ray. This falls within the range of greatest interest in the capture experiments (5-10 MeV). Although its 30 keV capture cross section is relatively small, the sodium in the crystal can be a source of long-term background. The neutron capture product, ^{24}Na has a 15 hour half life and decays with the release of a 2.76 MeV γ-ray.

The main sources of neutron background are:

(i) direct beam neutrons

(ii) neutrons scattered by the sample

(iii) partially moderated, multiply scattered neutrons originating from the neutron beam

(iv) radioactive neutron sources within the experimental area (e.g. Pu-Be and Am-Be sources)

(v) cosmic neutrons

2.2 Gamma Rays

Background γ-rays originate either from neutron capture or from beam independent ambient events. In most experiments a greater part of the neutron current bypasses the capture sample. These neutrons are scattered in the various structural materials, mainly the concrete walls and floor, and are either captured or decay. Figure 1(a) shows the γ-ray spectrum following neutron capture in materials in the experimental area. γ-ray energies with the most probable assignments are given in Table 2. The ground state-first excited state doublet at 7.65 MeV from neutron capture in ^{56}Fe is the most prominent feature of the high energy end of the spectrum. Although its $<$ 30 keV $>$ capture cross section is relatively small (18 mb), iron occurs in many structural materials (e.g. reinforced concrete).

Ambient γ-rays may be classified according to the source. γ-rays from cosmic ray showers have a dE/dx equivalent of several hundred MeV. Figure 2 shows a response curve for ambient events in large NaI($T\ell$) crystal. The cosmic ray peak occurs at 116 MeV. Other γ-rays in the ambient spectrum originate from natural or induced radioactivity. Figure 1(b) shows an ambient spectrum for the 8 in. x 6 in. crystal. Several strong γ-rays in this spectrum were assigned (Table 2) to radioactive isotopes of antimony created by previous neutron capture events. Antimony has a $<$ 30 keV $>$ capture cross section of 490 mb (Table 1) and occurs as a 5% by weight impurity in the industrial grade lead normally used for γ-ray shielding. It is therefore essential that all γ-ray shielding close to the NaI crystal be made of pure lead.

3. SHIELDING

For most situations where neutron shielding is required, it is common to use 50 percent by weight boric acid in paraffin. However, for neutron capture experiments using a Ge(Li) detector this material was found to be unsatisfactory (Allen 1968). Successful operation of the technique was achieved by the use of a mixture of boron carbide in paraffin (70% by weight B_4C) (Broomhall 1968). However because of the physical size and extreme sensitivity to background radiation of the 8 in. x 6 in. NaI(Tℓ) detector, a special shielding design study was needed, and a survey of the shielding properties of various materials was undertaken.

The neutron transport code 'SLABBO' (Clancy 1969) was used to calculate the transmission through slabs of a standard thickness. The materials investigated contained varying amounts of boron or lithium, carbon, hydrogen and oxygen. The input neutron current and energy distribution used in the calculations were chosen to simulate the experimental conditions. The code 'SLABBO' required the individual macroscopic cross-sections appropriate to the energy groups under consideration; consequently it was used in conjunction with the code 'GYMEA' (Pollard and Robinson 1966) which calculates flux weighted, group average cross sections. Because of the high hydrogen concentrations in some materials considered, it was found that the P_3 approximation to the anisotropic scattering matrix had to be used in order to obtain self consistent results. Table 3 shows the transmitted current in each energy group for the various materials. These calculations were checked experimentally in total transmission for materials 1 and 2 of Table 3 and the agreement between experiment and theory was found to be better than 5 percent. The theoretical value of 0.12 percent transmission over the whole energy range for boron carbide in paraffin (material 2) is a factor of two better than any other shielding materials considered. However the materials 1, 3, 5 and 6 transmit fewer neutrons than material 2 in the keV energy range: because of their higher hydrogen concentrations they have better moderating properties. In the final design the moderating and absorbing properties of materials 1 and 2 were combined to form a multi-layered shield (Figure 3). The outer layer consisted of 8 in. of boric acid in paraffin with a 2 in. inner sleeve of boron carbide in paraffin on all sides except the front face where 4 in. of boron carbide in paraffin was used. The need for further neutron absorbing material inside the 4 in. lead sleeve (γ-ray shield) has been demonstrated by the use of a ^6Li loaded liner in the shielding assemblies of other large crystals (Bird, Gibbons and Good 1962, Bergqvist and Starfelt 1961); the lead cavity forms an almost 4π scattering shell and increases capture probability in the crystal. The composition of the innermost sleeve was litharge (PbO), glycerine ($C_3H_8O_3$), boron carbide and lead shot in the weight ratios 5/1/4/10. Figure 4 shows this sleeve (in two parts) being fitted around the detector. The new material provided neutron absorption and moderation, further γ-ray shielding, and because it 'set' to a hard cement, provided adequate mechanical support for the detector body. The results shown in Figure 5(a) and (b) demonstrate the effectiveness of the new shielding assembly. Figure 5(a) is a time-of-flight spectrum showing the capture yield for transitions to the first, third and fifth excited states of ^{49}Ti using a boric acid in paraffin shield. Figure 5(b) shows the same capture yield using the present shield assembly. The improvement in peak/background ratio for the 37 keV resonance was a factor of 10. A large proportion of the remaining background counts was due to capture in the iron of structural materials in the experimental area. Figure 6 shows the γ-ray spectrum obtained with no sample in the neutron beam. Surrounding the target area on three sides with 4 in. of boric acid in paraffin reduces the iron peak by 30 percent but results in an increase in the intensity of the 2.2 MeV γ-ray from capture in hydrogen (Figure 6).

4. CONCLUSION

Adequate neutron shielding was achieved by the use of a multilayered assembly. For γ-ray transitions above 5 MeV, partial capture yields were obtained using flight paths up to 1.5 m and low cross section samples (Pb and Fe). For lower energy transitions, time-of-flight spectra were subject to high background levels because of the dominance of the 2.2 MeV γ-ray following neutron capture in hydrogen.

5. ACKNOWLEDGEMENTS

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TABLE 1

30 keV MAXWELLIAN AVERAGED CAPTURE CROSS SECTIONS

(MUSGROVE 1970)

Element	$\sigma_{n,\gamma}$ (mb)	Element	$\sigma_{n,\gamma}$ (mb)
C	0.2	Ag	920
Na	2.7	Cd	430
Mg	4.0	In	760
Al	4.6	Sn	95
Ti	20	Sb	490
Fe	18	I	760
Ni	12	W	290
Cu	47	Pb	4.6

TABLE 2

PEAK IDENTIFICATION OF THE SPECTRA IN FIGURE 1

(a) during neutron irradiation				(b) after neutron irradiation			
Line No.	E_γ (MeV)	Emitting isotope	Identification	Line No.	E_γ (MeV)	Emitting isotope	Identification
1	9.32	^{55}Fe	Iron in structures	1	2.75	^{24}Na	Crystal activation
2	8.83	^{55}Fe	Single escape peak of 1	2	2.61	^{208}Th	Thorium source
3	7.65	^{57}Fe	Iron in structures	3	2.07	^{124}Sb	Sb in lead shield
4	7.16	^{57}Fe	Single escape peak of 3	4	1.69	^{124}Sb	Sb in lead shield
5	6.80	^{129}I	NaI crystal	5	1.48	^{40}K	P.M. glass and photo-cathode
6	6.75	^{49}Ti	Target store	6	1.37	^{24}Na	Crystal activation
7	6.44	$^{49}Ti+^{41}Ca$	Target store	7	1.13	^{122}Sb	Sb in lead shield
8	5.98	^{57}Fe	Iron in structures				
9	5.49	^{57}Fe	Single escape peak of 8				
10	4.94	$^{29}Si+^{13}C$	Concrete				
11	4.44	^{11}B	Neutron shield				
12	4.21	^{57}Fe	Iron in structures				
13	3.53	^{29}Si	Concrete				

TABLE 3

NEUTRON TRANSMISSION PROPERTIES OF SHIELDING MATERIALS

Neutron Energy Group	Input Current	Transmitted Current Through 4 in. Thick Material					
		1	2	3	4	5	6
1	5	0.000054	0.000126	0.00036	1.86	0.00044	0.00021
2	10	0.000326	0.000787	0.00020	2.68	0.00244	0.00125
3	5	0.001484	0.003057	0.00163	3.52	0.00919	0.00532
4	0	0.047157	0.019697	0.05330	0.37	0.09972	0.01229
5	0	0.048035	0.000674	0.10100	1.2×10^{-8}	0.01341	0.02711
Total transmission		0.46%	0.12%	0.78%	42.2%	0.63%	0.25%

Materials

1. 50% boric acid in paraffin
2. 70% boron carbide in paraffin
3. 5% boron in polyethylene
4. Sintered boron carbide
5. 50% boron carbide in polyethylene
6. 50% lithium carborate in paraffin

} percent by weight

Neutron Energy Groups

1. 67-53 keV
2. 53-32 keV
3. 32-12 keV
4. 1200-101 eV
5. 101 eV-sub-thermal

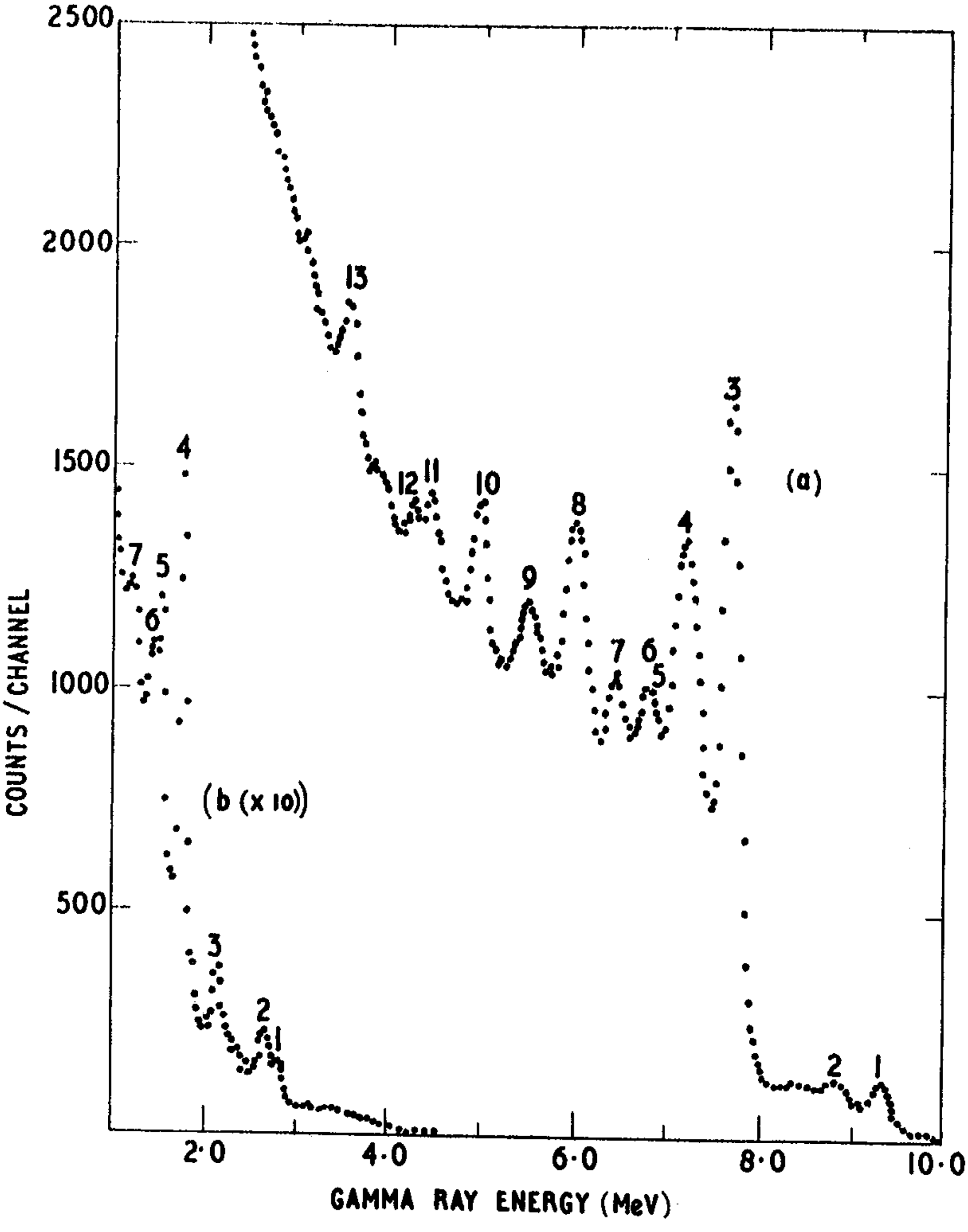

FIGURE 1. GAMMA RAY ENERGY SPECTRA OBTAINED (a) DURING AND (b) AFTER NEUTRON IRRADIATION OF THE 8 in. × 6 in. NaI(Tl) CRYSTAL. PEAKS ARE IDENTIFIED IN TABLE 2.

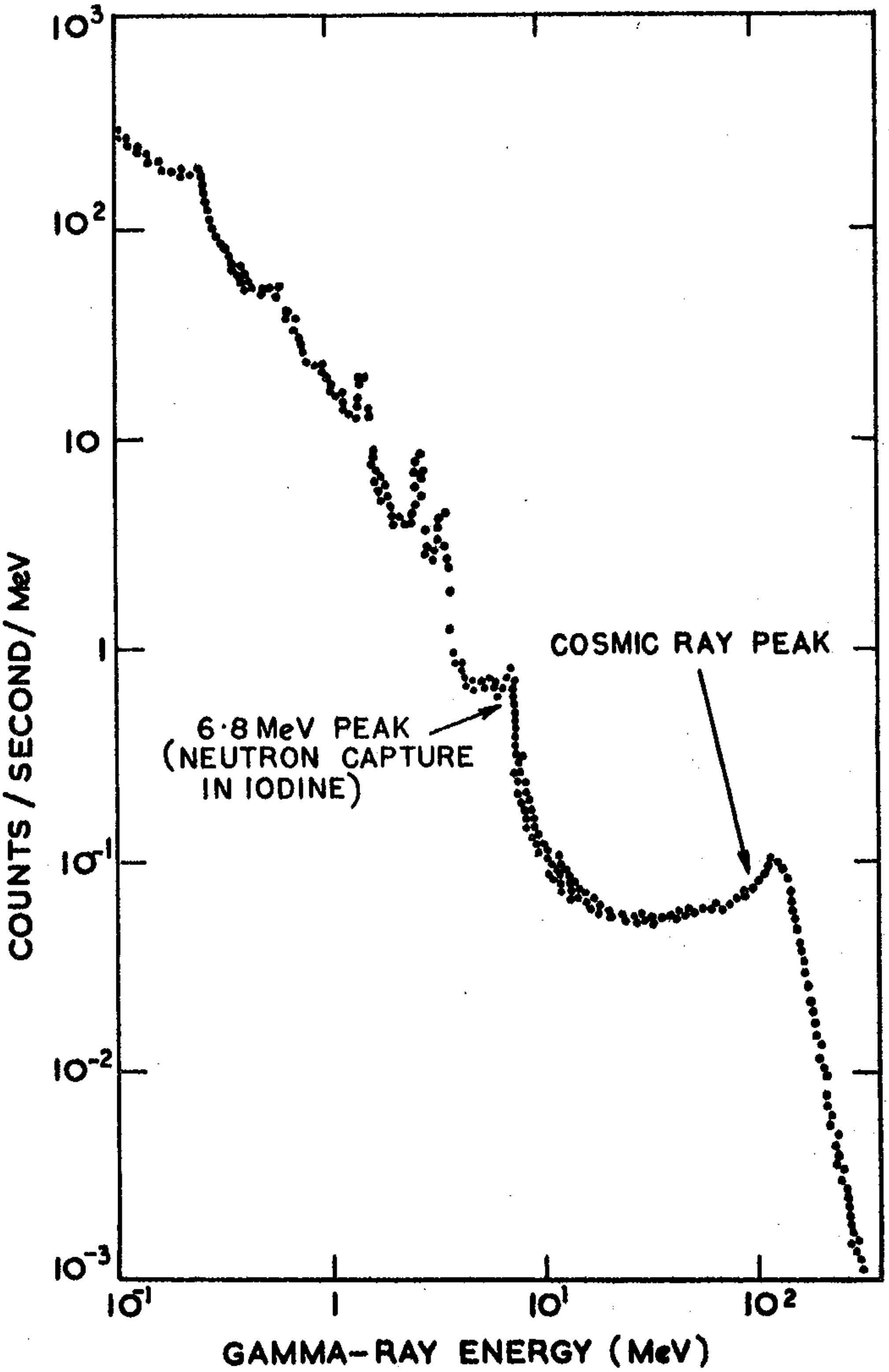

FIGURE 2. AMBIENT SPECTRUM OBTAINED USING THE 10 in. × 12 in. NaI(Tl) CRYSTAL AT O.R.N.L. (BIRD, GIBBONS AND GOOD 1962)

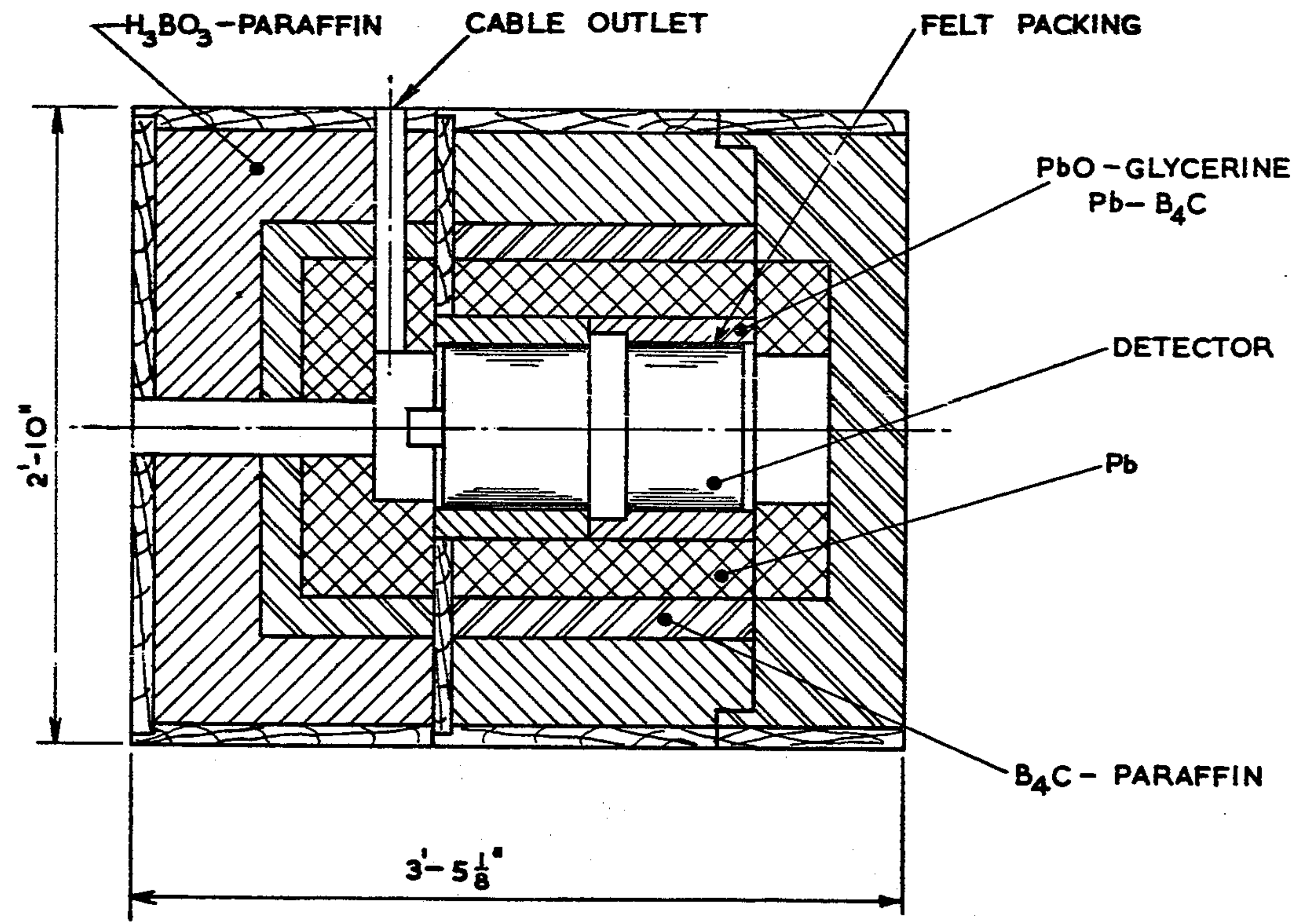

FIGURE 3. LONGITUDINAL SECTION THROUGH THE SHIELDING ASSEMBLY

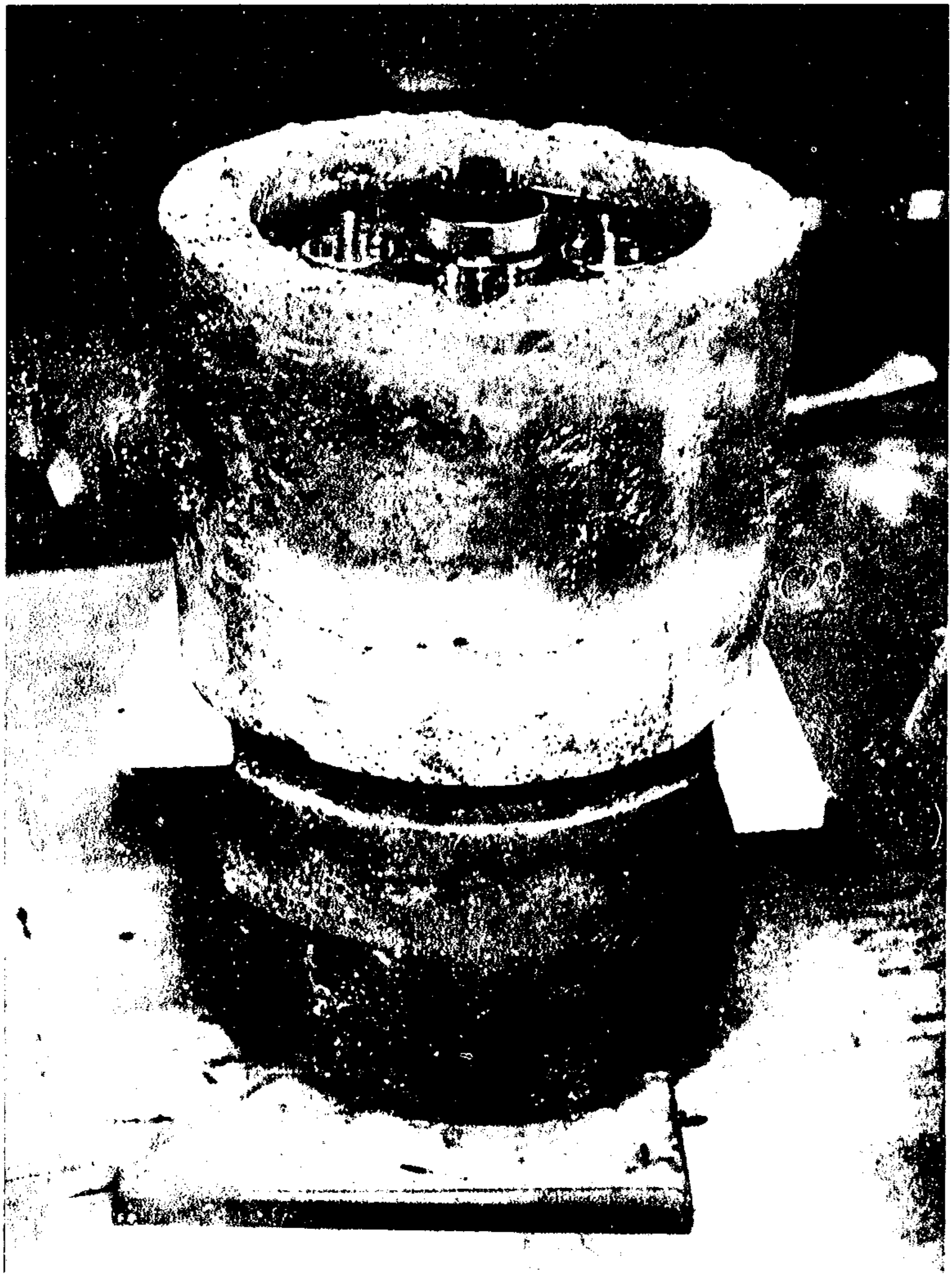

FIGURE 4. BORON CARBIDE/LEAD SHOT/LITHARGE/GLYCERINE INNERMOST SHIELD SLEEVE

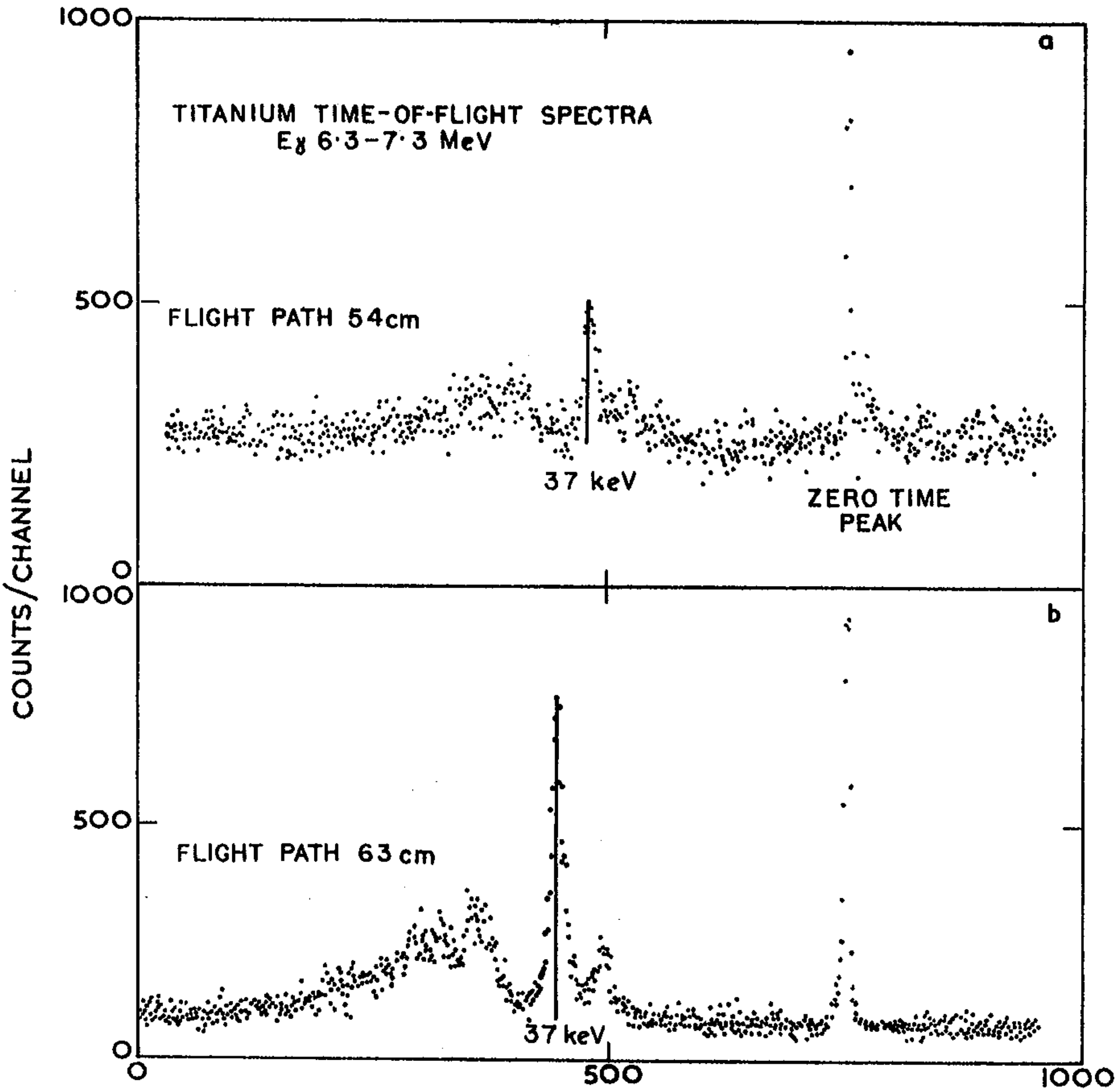

FIGURE 5. TIME-OF-FLIGHT SPECTRA FOR A TITANIUM SAMPLE USING (a) A BORIC ACID IN PARAFFIN/LEAD SHIELD AND (b) THE ASSEMBLY SHOWN IN FIGURE 3

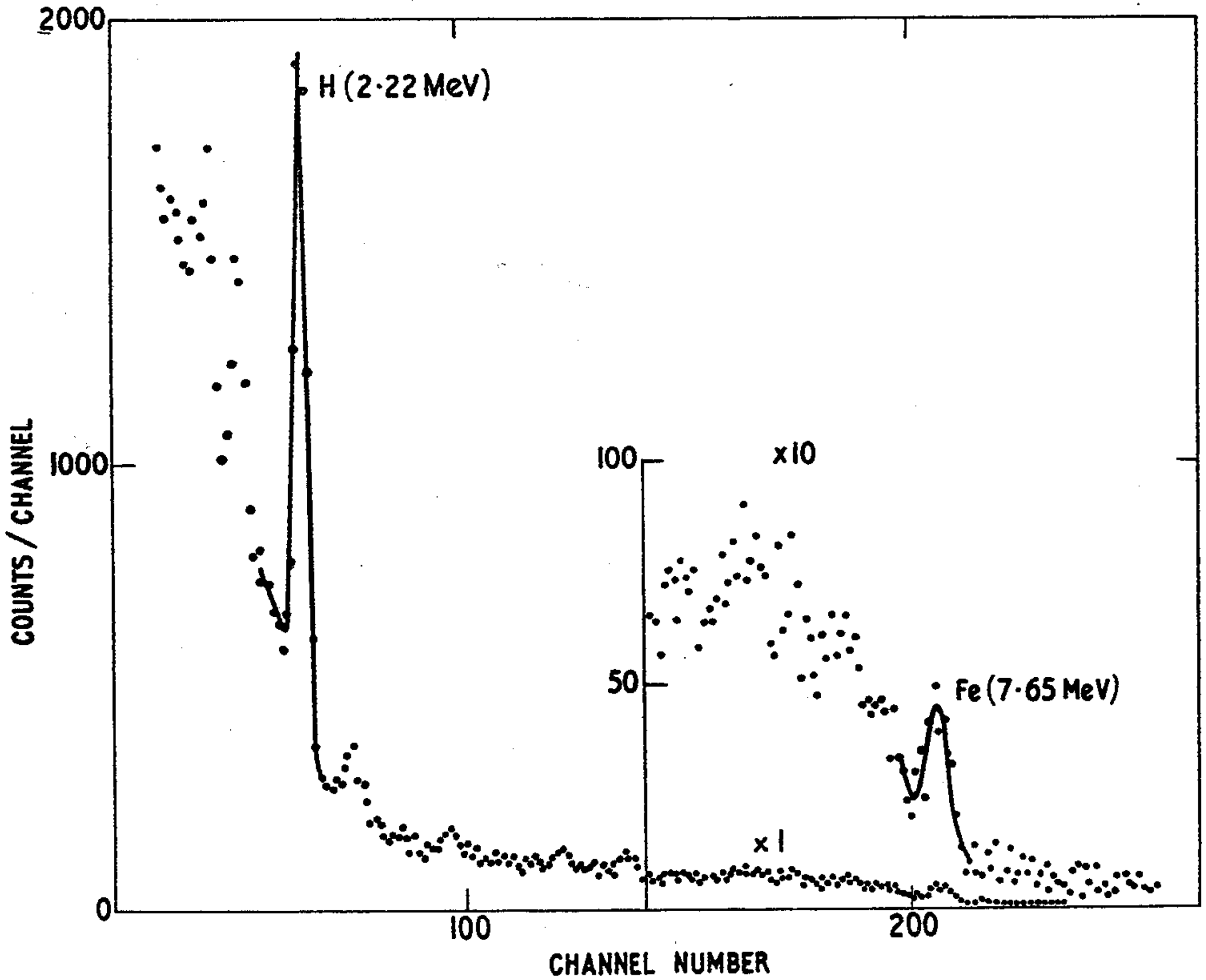

FIGURE 6. GAMMA RAY PULSE HEIGHT SPECTRUM OBTAINED WITH NO SAMPLE IN THE NEUTRON BEAM