NEUTRON AND PHOTON DETECTORS FOR URANIUM AND PLUTONIUM APPLICATIONS

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Abstract

Uranium and plutonium storage present different challenges to radiation detectors. The former does not generate significant numbers of neutrons but does produce many gamma- and x-rays below 200 keV. On the other hand, gamma radiation field from plutonium is intense, extends to energies significantly above those from uranium, and plutonium emits spontaneous fission neutrons. Radiation detectors based on the $^{10}B(n,\alpha)$ reaction and using CZT or scintillator are being evaluated for use in storage applications. In addition, a small and relatively inexpensive detector employing a thin polyethylene radiator mounted within an ion chamber was built to measure fast fission neutrons in a high gamma-ray field and at temperatures in excess of 50 ºC. This paper will present the results of measurements of the characteristics of these devices and an evaluation regarding their utility in storage applications.

INTRODUCTION

The large number of items that might be in a storage facility makes it desirable to minimize the amount of radiation detection equipment. This will have the effects of decreasing the initial investment, improving reliability by decreasing component count, and decreasing the operating costs of the facility. To this end, combined neutron/gamma ray have been under development at the Oak Ridge Y-12 Plant[2] and the Oak Ridge National Laboratory. These detectors are based on CZT and $CdWO_4$ scintillator because of their inherent gamma ray spectroscopic characteristics and the high thermal capture cross section of cadmium. The latter implied that is would not be necessary to coat these materials to sensitize them to neutrons.

There are a few reported successes using CdTe solid state detectors[3,4,5] for thermal neutron detection. These devices relied the detection of capture gammas by the diode, in particular the 558 keV de-excitation of the first excited state of ^{114}Cd, which occurs in 74% of captures[6]. Unfortunately, this high-energy gamma ray has a long mean free path and requires a large (approximately 10 mm) cube to achieve reasonable efficiency.

In contrast to CdTe and CZT, however, cadmium tungstate scintillators of large dimension are readily obtainable, and these were studied as well. Crystals a few centimeters on a side have a sufficiently large photoelectric cross section that Cd capture gammas would be easily stopped and a neutron signature more easily obtained than from diodes. To their detriment, however, scintillator crystals do not provide the same resolution as the solid-state devices, although the 7 - 10% resolution obtainable was expected to be adequate for uranium enrichment measurements, and gross gamma activity monitoring.

Detectors based on Cd or B are sensitive primarily to slow neutrons. However, since plutonium is a source of fast spontaneous fission neutrons, it is advantageous to incorporate a detector sensitive to these also. Gas-filled detectors can be made simultaneously sensitive to fast recoil protons and insensitive to gamma radiation, and able to operate at high temperatures.

CADMIUM TUNGSTATE

The long mean free path of the 558 keV gamma ray from cadmium coupled with the large sizes of $CdWO_4$ scintillator readily available made this material an obvious subject of investigation. Like CdTe, it simultaneously serves as a Cd-rich target and an efficient photon detector, the mean free path of 558 keV photons in $CdWO_4$ being only 1 cm. The principle, then, behind the detector was to detect in the body of a sufficiently large crystal capture gammas generated in the outermost 2 mm of the crystal while simultaneously detecting the incident photon flux.

Three crystals, $10 \times 10 \times 10$ mm, $10 \times 20 \times 30$ mm, and $30 \times 30 \times 30$ mm, were evaluated. The smallest and largest crystals had two smooth cleaved faces and four unpolished saw-cut faces. The $10 \times 20 \times 30$ mm crystal had two 20×30 mm cleaved faces and one each of the 10×30 mm and 10×30 mm faces polished. Pulse height spectra from each crystal were obtained with ^{137}Cs, ^{133}Ba, ^{60}Co, and depleted uranium. Resolution of the smallest and largest crystals was approximately 10%, but that of the $10 \times 20 \times 30$ mm crystal viewed through a 20×30 mm face was 7% at 661 keV. It was not determined if this difference could be attributed to counting statistics or if the dimensions of the crystal or the number of polished faces were critical factors in the better resolution.

The neutron response of the crystals was extremely disappointing. In marked contrast to the published reports on CdTe, little or no evidence of Cd capture gammas was seen in $CdWO_4$ exposed to thermal neutrons. However, the fact that the reported data has all been obtained from thin (0.25 to 2 mm thick) CdTe diodes, while the present work has been with much larger crystals offers, perhaps, an explanation.

Since all captures occur in a thin surface layer of the crystal, the probability of a capture gamma entering the crystal is approximately 50%. Consequently, subsequent to the capture of each neutron approximately half of all capture gammas enter the crystal essentially simultaneously and result in a light pulse that corresponds to the sum of their energies. However, the mix of gammas entering the crystal is not constant from capture event to capture event, but is itself a random variable. This results in a smooth spectrum with few, if any, distinguishing characteristics. The reason for the success of the CdTe detectors is attributable to their thinness: They do not have sufficient efficiency to detect more than one capture gamma per event and so retain the line spectrum characteristic. Although the experiments were repeated with a $10 \times 10 \times 5$ mm crystal, no significant improvement was observed.

Since the basic difficulties in the use of $CdWO_4$ as both the Cd target and the detector are the size and efficiency of the crystal, and the number and random distribution of photons entering the crystal, the use of a target material emitting only a few photons and placed in close proximity to the scintillator suggests itself. This arrangement would result in a comprehensible spectrum while still enjoying the advantage of compactness that results from the use of a high-Z, dense scintillator.

Boron is known to have a high neutron capture cross section, and the capture reaction results in lithium nuclei in their first excited state. This state de-excites by the emission of a single 478 keV gamma ray, which in the presence of both neutrons and photons, would be superimposed on the photon spectrum. Since boron is a low-Z, low-density material, it does not significantly affect the detection of gamma rays, and the pulse height spectrum from the scintillator indicates both the presence of neutrons and the identity of incident photons.

Figure 1 shows the pulse height spectrum obtained from a $10 \times 10 \times 10$ mm crystal surrounded by boron and placed in a port of heavily moderated Am-Li neutron source. Measurements of the thermal flux with In and Dy activation foils showed it to be approximately 100 /cm^2/sec, in agreement with MCNP calculations. The same MCNP calculations indicated that there was an additional 10 /cm^2/sec neutrons with energy in excess of 10 eV. In addition to neutrons, the detector is exposed to a gamma ray spectrum consisting of 2.23 MeV capture gammas from hydrogen, capture gamma rays from the moderator's cadmium liner, capture gammas from the tungsten-nickel-iron shield around the Am-Li container, and gamma rays from activated tungsten in the shield.

The spectrum in Figure 1 was taken with a 1-cm thick Pb shield in place. The 478 keV line from Li clearly stands out against the background that penetrates the shield. The broad hump to the left of the peak is the backscatter peak.

Figure 2 shows the events caused by fast neutrons. With the Cd cover in place outside the Pb shield, thermal neutrons are excluded from the boron nitride while neutrons over 1 eV easily pass through. Since the boron cross section falls like $1/\sqrt{E}$, and the neutron spectrum's fast component is approximately 10%, there are some captures despite the Cd cover. The broadening of the peak occurs because the cadmium cover introduces a strong 558 keV capture gamma that penetrates 1 cm of Pb and is detected by the crystal.

Figure 3 shows the response of the detector with only the Cd shield in place. The vertical scale is a factor of 2 larger (there are over twice as many counts in the 478 keV peak) than in Figure 2. In addition, the horizontal scale has been expanded to show the separation between the 558 keV peak from Cd and the 478 keV peak from neutron events. The reader should also note that there is now a rapidly rising background at the left in Figure 3.

All shields have been removed for the spectrum in Figure 4. It is seen that the background is unchanged from that in Figure 3 (signifying that the Cd foil does not significantly attenuate the gamma spectrum) while the peak at 478 keV is significantly enhanced. Also the 558 keV peak from neutron captures in Cd has disappeared.

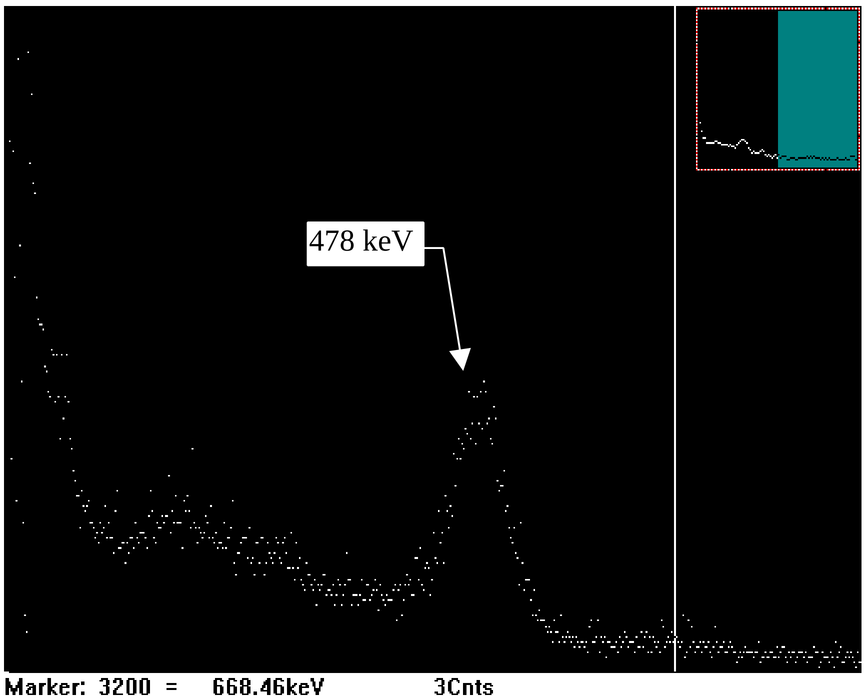

Figure 1. Neutron response. Detector covered with Pb only.

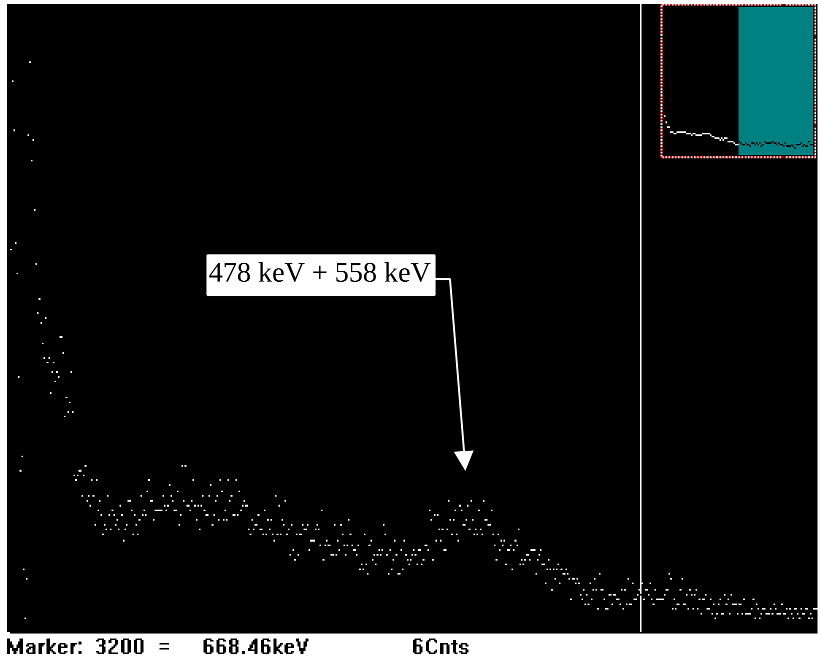

Figure 2. Neutron response. Cd and Pb shields in place.

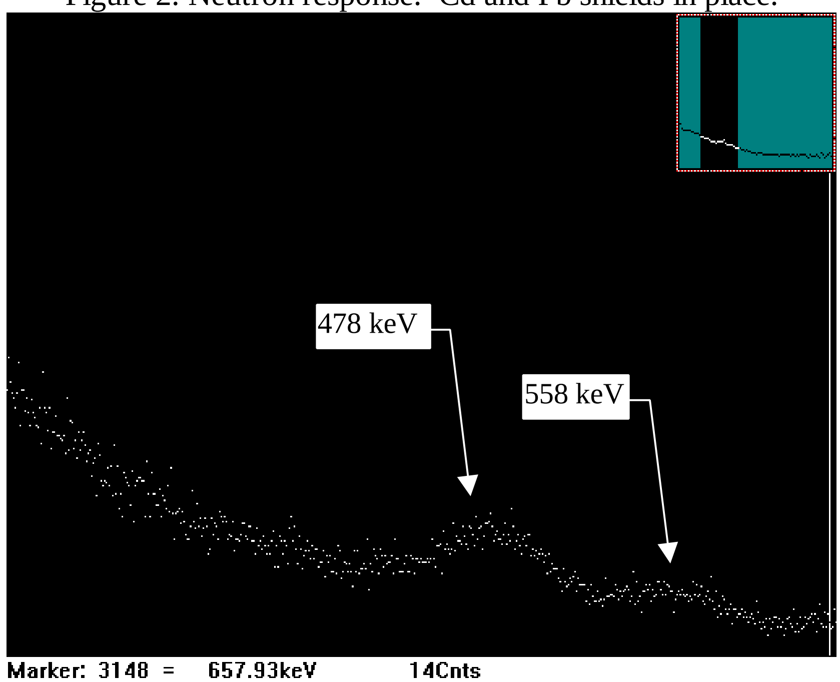

Figure 3. Neutron response. Only Cd shield in place.

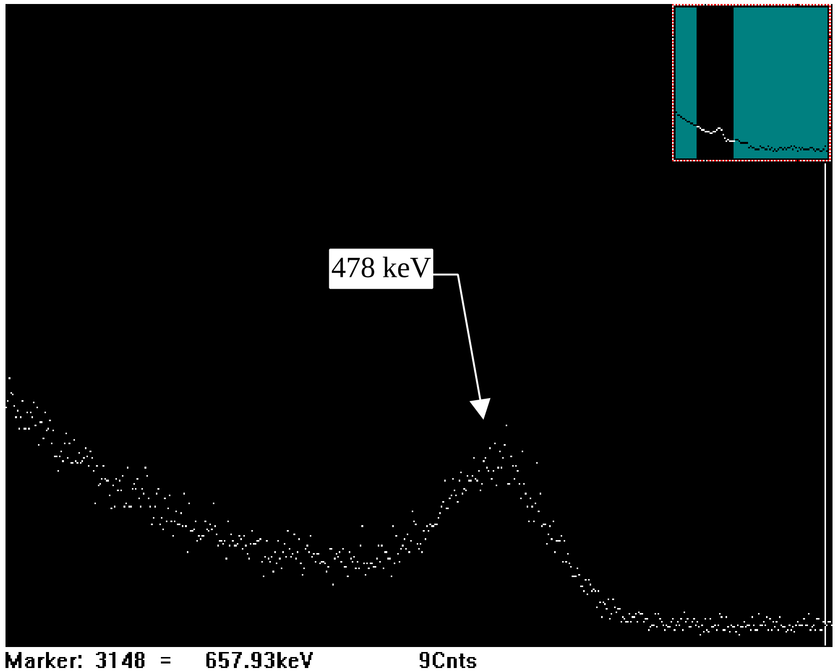

Figure 4. Neutron response. No shields in place.

Monte Carlo analysis of the expected detector response yielded an absolute efficiency of 4 - 5% (photopeak counts/incident neutron) depending on thickness of boron coating.

CADMIUM ZINC TELLURIDE

The success of the boron-coated scintillator led to the evaluation of boron coated CZT diodes and Monte Carlo calculations were used to calculate the optimal thickness of a 8 × 8 mm ^{10}B chip coupled to a 10 × 10 × 5 mm crystal CZT. Results were calculated for a centered 0.025 eV monoenergetic neutron point source emitting isotropically into the ^{10}B chip. Only the 480 keV gamma flux and energy deposition were calculated; other features in the spectrum (e.g., Compton scattering, neutron capture in cadmium, etc) were not calculated.

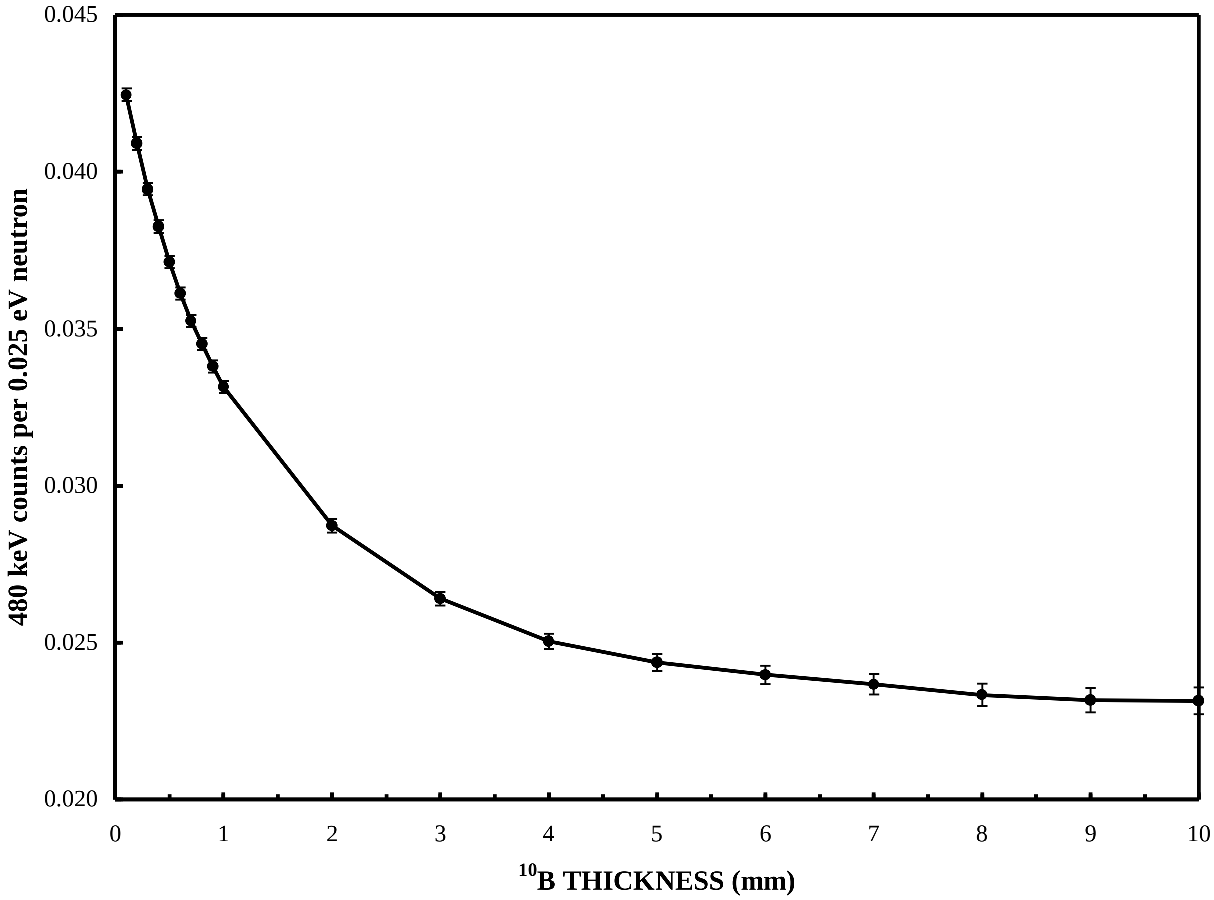

Figure 5. Calculated absolute efficiency of boron-coated CZT diode.

Figure 5 shows the results of calculation of the absolute efficiency (photopeak counts/incident neutron) as a function of boron layer thickness. As expected, efficiency increases with decreasing thickness of boron layer. At 0.1 mm, the expected photopeak efficiency is 0.042.

The response of a coated CZT diode to neutrons was observed with a moderated PuBe source and the pulse height spectrum had the same features shown in Figures 1 - 4.

PROTON RECOIL FAST NEUTRON DETECTOR

Since Pu is a spontaneous fission source of fast neutrons, investigations were carried out on a rugged fast neutron detector based on proton recoil. A small and relatively inexpensive prototype was built for operation in a high

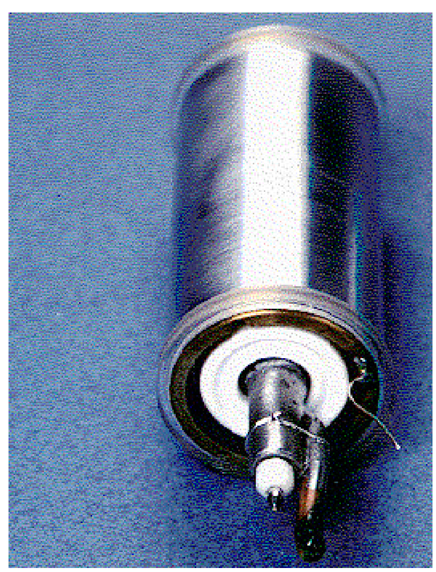

Figure 6. Proton recoil fast neutron detector

gamma-ray field and at temperatures up to 75 ºC. The prototype detector used a thin sheet of polyethylene (CH_2) mounted inside a small ion chamber. The ion chamber uses metallic indium seals for easy assembly, a small anode wire assembly, and is filled with inexpensive 90% Ar, 10% CH_4. Tests on the chamber, a year after the original gas filling, were completed to verify the stability of the gas and detector chamber integrity. The detector, shown in Figure 6, has an overall diameter of 2 cm and a length of approximately 5 cm.

Fast neutron detection is based primarily on elastic scattering of neutrons by the hydrogen nuclei. The scattering interaction transfers a portion of the neutron kinetic energy to the hydrogen nucleus resulting in a recoil proton. The recoil proton ionizes the fill gas as it passes through the counter. The electric field established by the high voltage bias sweeps the ion pairs from the active volume and generates a pulse with amplitude that is proportional to the energy deposited by the recoil proton. Preliminary tests indicated that the detector has a neutron sensitivity of approximately 0.01 cps/nv. The detector was also irradiated with gamma rays from a 2 millicurie ^{60}Co source and an aged ^{232}Th source. No pulses were observed. If any pulses were to occur due to high energy gamma photons, pulse height discrimination can easily be used for gamma rejection.

CONCLUSIONS

Neutron/gamma ray detectors based on CZT diodes, high-Z, high-density scintillators, and proton recoil have been demonstrated. The presence of small amounts of boron does not affect the gamma ray spectroscopic properties of the gamma ray detectors and it is only necessary to perform a pulse height analysis to obtain the neutron signature. In the case of designs based on the detection of one or more signature capture gamma rays, it is important that there not be a significant contribution from naturally occurring gamma rays at the same energies.

The major drawback to the use of the 478 keV gamma ray as the neutron signature is that the natural presence of gamma rays with similar energy will complicate the analysis. In the present case, the tungsten shield in the Am-Li source is activated (^{187}W) and emits a 480 keV gamma ray. A scintillator cannot distinguish the 478 keV emission from radiations separated from it by less than 7-10% except through the use of various covers that selectively attenuate gammas and neutrons. In contrast, spectroscopy-grade CZT would be expected to resolve peaks with a 4 - 5% separation.

The detectors described above are not particularly expensive; each may be fabricated for less than $500.00 (for quantity 1). Desired gamma resolution and sensitivity, as summarized in Table 2, dictates which detector should be selected for a particular application.

Detector Technology	Neutrons	Design Temperature	Neutron Efficiency	Gamma-Ray Sensitivity
Scintillator/B	Thermal	< 100 ºC	0.04	Yes
CZT/^{10}B	Thermal	< 100 ºC	0.04	Yes
Proton Recoil	Fast	< 75°C	0.01 cps/nv	No

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