This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-96SR18500 with the U. S. Department of Energy.

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COMPARISON OF CdTe AND CdZnTe DETECTORS FOR FIELD DETERMINATION OF URANIUM ISOTOPIC ENRICHMENTS

K. J. Hofstetter and D. M. Beals
Westinghouse Savannah River Company
Savannah River Technology Center
Aiken, SC 29808, USA

A performance comparison of a CdTe and a CdZnTe detector when exposed to uranium samples of various isotopic enrichments has been performed. These high-resolution detectors can assist in the rapid determination of uranium isotopic content of illicit material. Spectra were recorded from these “room temperature” semiconductor detectors with a portable multi-channel analyzer, both in the laboratory and in a field environment. Both detectors were operated below ambient temperature using the vendor supplied thermoelectric coolers. Both detectors had nominally the same active volume (18 mm^3 for the CdZnTe and 25 mm^3 for the CdTe detector) and resolution (i.e., <1.0 keV resolution FWHM at 59.5 keV). Spectra of samples of known isotopic content were recorded at fixed geometries. An evaluation of potential signature γ rays for the detection of enriched uranium was completed. Operational advantages and disadvantages of each detector are discussed.

Introduction

There is a need to improve the detection sensitivity during the interdiction of special nuclear materials (SNM)[*] for increased homeland protection. It is essential to provide additional tools to first responders and law enforcement personnel for assessing nuclear and radiological threats.

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[*] SNM includes fissile isotopes such as U-233, U-235, and Pu-239 that can be used to produce a nuclear weapon

The presence of penetrating radiation is a signature of most SNM. The passive detection of γ-rays or neutrons is presently the most effective screening tool for detecting illicit SNM. Some active interrogation techniques are also used in the field but these are usually at fixed monitoring locations. While plutonium can be detected with passive γ-ray and/or neutron techniques, the field detection of highly enriched uranium (HEU)[*] is more difficult since the γ-rays are low in energy, the γ-ray emission rate is low, and there is no appreciable spontaneous fission rate. Additional complications arise as there is an abundance of uranium bearing materials that are part of normal commerce and uranium also appears as a component in many naturally occurring radioactive materials. The ability to quickly identify uranium that has been isotopically enriched in ^{235}U using a hand-held instrument is one of the goals of this study.

Recent developments in semiconductor detector technologies have opened new possibilities for the field measurement of γ-ray and X-ray radiations in the 10-500 keV region of the spectrum. Cadmium-zinc-telluride (CZT) or cadmium-telluride (CdTe) semiconductor detectors, that can be operated without cryogenic cooling, have permitted the field measurement of energy spectra from radiation sources using hand-held instruments with greatly improved energy resolution over more traditional NaI scintillation detectors. This capability greatly improves the ability to detect, locate and identify radiation sources and discriminate between items of commerce, naturally occurring radioactive material and nuclear contraband. CZT or CdTe detectors have a significant advantage over high purity germanium (HPGe) detectors in their compact design without the need for cryogenic cooling.

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[*] HEU contains ≥20 percent by weight of U-235

The major disadvantage of these room temperature semiconductor detectors is the low radiation detection efficiency due to their small size. The selection of the best γ-ray or X-ray detector for a specific application is therefore a compromise between energy resolution and detection efficiency. A comparison of various types of commercially available radiation detectors for field applications has been reported [1]. There continues to be advancements in the CZT and CdTe semiconductor technologies to produce more material with higher resistivities and decreased leakage currents. Additional improvements have included the use of thermoelectric cooling with Peltier elements to improve the energy resolution and stability of the detectors [2] and digital pulse processing to reduce the hole trapping effects [3]. This study compares the performance of two commercially available instruments, one containing a CZT and the other a CdTe detector, when exposed to uranium samples of varying isotopic content.

Experimental

The response of the two detectors was measured using various radionuclides with γ-ray and X-ray emissions following standard spectroscopic practices. Spectra were recorded and analyzed using vendor-supplied firmware for data acquisition, multichannel analyzers and software for analyzing the spectra. Energy, efficiency and peak shape calibrations were determined using standard radioactive sources for each detector over the energy range of 50-200 keV, the region of the spectrum where most of the radiations from the uranium radioisotopes and their radioactive progeny occur. The detectors tested in this study included a 3 mm x 3mm x 2 mm CZT detector with a 0.025 mm Be entrance window and a 5 mm x 5 mm x 1 mm CdTe detector with a 0.010 mm Be entrance window.

The 3 x 3 x 2 mm CZT detector and charge sensitive preamplifier were mounted on a Peltier cooling element to maintain operating temperatures near -30°C. The preamplifier was connected to an external NIM amplifier with a 0.5 μsec shaping time. The amplifier was operated in the rise time discrimination mode to exclude signals resulting from incomplete charge collection. The 5 x 5 x 1 mm CdTe detector operated with 1.5 μsec pulse shaping time and rise time discrimination. Data collection was accomplished with an internal ADC coupled to a HP200LX palmtop computer. This detector and the FET feedback preamplifier are also thermoelectrically cooled. A CZT detector without thermoelectric cooling was previously tested with results reported in reference 2.

The spectrum of natural uranium in equilibrium with all its progeny is very complex and easily identified by its higher energy γ-ray signature [4]. A typical low energy γ-ray spectrum for a small unprocessed uranium sample that is in radioactive equilibrium with all its progeny is shown in Figure 1. The most prominent feature of the spectrum, recorded with the 5 x 5 x 1 mm CdTe detector, is the Bi K_α X-rays at 74 and 77 keV. Other radiations from the decay daughters are also indicated on the spectrum including a trace of the 186 keV γ-ray from ^{235}U.

Uranium that has been chemically processed has a less complex γ-ray spectrum; ^{235}U and ^{238}U reach radioactive equilibrium with only a few radioactive progeny due to the long half-lives of the intermediary decay products ^{231}Pa and ^{234}U. Table 1 lists the principal radiations emitted from chemically processed ^{235}U and ^{238}U in radioactive equilibrium with their prompt daughters. From the data in this table, the intensity of the radiations being emitted from a uranium source of natural isotopic abundance (0.72% by weight ^{235}U) was calculated (see Table 2).

Spectra of uranium samples of various isotopic compositions were recorded for short time periods (10 minutes) with each detector. Ten minutes was chosen arbitrarily to simulate routine field screening limitations. The isotopic compositions of all the uranium samples were determined by thermal ionization mass spectrometry. A portion of the spectra of radiations from a uranium sample depleted in ^{235}U content (0.23 wt. %) and a chemically processed sample containing ^{235}U with natural isotopic abundance (0.72 wt. %) taken with the 5 x 5 x 1 mm CdTe detector is shown in Figure 2a. The prominent radiations occur at 63.3 keV and ~92 keV arising from the decay of ^{234}Th and the uranium K X-rays in the energy region 95-110 keV. A typical spectrum of a HEU sample with 68 wt. % ^{235}U taken with the same CdTe detector is shown in Figure 2b. The spectrum shown in Figure 2b is dominated by ^{235}U radiations due to the higher specific activity and greater γ-ray emission probability of ^{235}U.

The results of our measurements of the γ-ray spectra from a variety of uranium samples with varying isotopic composition have concentrated in three primary regions of interest in the energy spectrum. The very low energy radiations below 50 keV were not considered in this study as they can be easily shielded and their emission affected by self-absorption. The first region is the low energy (50-75 keV) portion of the spectrum. Figure 3a displays this region for the enriched uranium sample (68 wt. % ^{235}U) as compared to a sample of uranium with naturally occurring isotopic composition. The natural uranium sample spectrum is dominated by the 63.3 keV γ-ray from ^{234}Th. The dominant feature in the spectrum of the enriched uranium sample is a γ-ray at 53.2 keV which originates from the decay of radiogenic ^{234}U. The 53.2 keV γ-ray is not evident in the spectrum of the sample with normal ^{235}U isotopic content due to its low ^{234}U natural abundance (0.0055 wt. %). The calculated emission rate for this γ-ray is 15.6 γps/gm U for a natural uranium sample as compared to the other γ-rays given in Table 2. The isotopically enriched uranium sample contained ~0.5 weight percent ^{234}U.

The 53.2 keV γ-ray was present in all the enriched uranium samples investigated in this study due to small quantities of ^{234}U in the enriched ^{235}U. The ^{234}U content is typically increased with nominally the same efficiency as the fissionable isotope ^{235}U during the isotopic enrichment process but it can vary substantially depending on the enrichment method. Thus the 53.2 keV radiation cannot be used to calculate the isotopic composition of a sample but it can be used as an indicator of the presence of HEU. The 63.3 keV γ-ray is absent in the spectrum of the 68% enriched uranium shown in Figure 2b due to the nearly 100-fold increase in the ^{234}U and ^{235}U content and the corresponding 3-fold decrease in the thorium content. The 53.2 keV γ-ray is also clearly visible in spectra of low enriched (1-2 wt % ^{235}U) uranium (LEU) samples (not presented here). The emission of the 63.3 and 53.2 keV γ-rays would be equivalent for ~20% HEU if the ^{234}U and ^{235}U were comparably enriched. The intensity of the 63.3 and 53.2 keV γ-rays are decreased by self-absorption in the large uranium samples and further reduced by external shielding but these radiations are useful in detecting the presence of low uranium enrichments in small, unshielded samples.

The second region of interest in the spectra of uranium samples is the 75-125 keV energy range. This region is shown in Figure 3b for the 68% enriched uranium sample and a natural uranium sample. This region of the spectrum contains γ-rays and X-rays from the decay of the uranium isotopes and their equilibrium decay progeny. The X-rays are the $K_{\alpha1}$ and $K_{\alpha2}$ and the $K_{\beta1}$ and $K_{\beta2}$ from the various isotopes. The dominant X-rays in this energy region are the $K_{\alpha1,2}$ from U and Th (see Table 2.). From Table 2 one might expect the uranium X-ray at 98.4 keV to be a possible indicator of ^{238}U since there is no corresponding radiation in the decay of ^{235}U nor its equilibrium progeny in this energy range. Inspection of Figure 3b reveals that the spectrum of the enriched uranium sample contains a significant peak at ~98 keV. This is probably due to the X-rays produced in the bulk uranium sample as a result of the increased specific α-activity causing secondary uranium fluorescence. As can be seen in the spectrum, the 92 keV γ-rays from ^{234}Th are obscured by the K_α X-rays from uranium in the enriched uranium sample spectrum.

The third region of interest in the uranium spectra is shown in Figure 3c. The characteristic 185.7 keV γ-ray in the decay of ^{235}U dominates this energy range along with the lesser abundant 163.3 keV γ-ray. There are no corresponding radiations from ^{238}U or its decay daughters in a chemically separated sample in this energy region.

Figure 4a-4c are spectra of an enriched uranium sample taken with the 3x3x2 mm CZT detector and the 5x5x1 CdTe detector for the three regions of the spectrum discussed earlier. The slightly larger CdTe detector appears to have slightly increased detection efficiency in all spectral regions and comparable energy resolution. Our experience to date suggests the presence of HEU (^{235}U > 20 % by wt) can be detected in small samples if the intensity of the 98 keV X-ray is greater than the 93 keV complex. This is in addition to possibly observing the 53.2 and 63.3 keV γ-rays. Further experimentation is underway to confirm these observations and quantify their effect.

CONCLUSIONS

A comparison of the responses of two non-cryogenic solid-state semiconductor detectors have shown their ability of provide field personnel with valuable information concerning the isotopic enrichment of uranium samples seized as possible nuclear contraband. Inspection of the low energy region of the spectrum (50-75 keV) can quickly determine if a sample contains enriched uranium by observing the presence or absence of the 53.2 keV γ-ray from ^{234}U without extensive modeling and self-absorption corrections. Initial evidence also suggests the presence of HEU can be determined by inspection of the X-ray energy region of the spectrum taken with these detectors by noting the relative intensity of the K_α X-rays (93 and 98 keV) characteristic of uranium.

The primary advantage of the CdTe detector over the other CZT detectors tested during this study was its packaging. The CdTe is part of a total man-portable instrument for field use (mfg. by AmpTek, Inc.). The 3x3x2 mm CZT detector used in this study (mfg. by eV Products) required external amplification, an ADC, a MCA, and a source of power. Both instruments utilize semiconductor detectors that are cooled below ambient conditions which provides improved detector resolution and stability. As other manufacturers begin to incorporate thermoelectrically cooled CZT or CdTe detectors into their hand-held portable spectrometers, these instruments should become a useful tool for first responders and law enforcement personnel to detect nuclear contraband and protect themselves and the general public from potential acts of nuclear terrorism.

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Table 1. The γ-ray emission rate* calculated for 1 gram of the isotopes ^{235}U and ^{238}U.

Energy (keV)	γps/gm ^{235}U	Isotope	Energy	γps/gm ^{238}U	Isotope
90.0	2853	^{235}U	63.3	598	^{234}Th
93.4	4656	^{235}U	83.3	10	^{234}Th
105.0	2156	^{235}U	92.4	349	^{234}Th
109.2	1234	^{235}U	92.8	349	^{234}Th
143.8	8783	^{235}U	94.7	18	^{234m}Pa
163.3	4071	^{235}U	98.4	29	^{234m}Pa
185.7	45,324	^{235}U	111.0	13	^{234m}Pa
205.3	4015	^{235}U	112.8	35	^{234}Th

Table 2. The calculated γ-ray emission rate from 1 gram of natural uranium.

Energy (keV)	γps/gm U	Isotope
63.3	598	^{234}Th
83.3	10	^{234}Th
90.0	21	^{235}U (Th X-ray)
92.4	349	^{234}Th
92.8	349	^{234}Th
93.4	34	^{235}U (Th X-ray)
94.7	18	^{234m}Pa (U X-ray)
98.4	29	^{234m}Pa (U X-ray)
105.0	16	^{235}U (Th X-ray)
109.2	9	^{235}U (Th X-ray)
111.0	13	^{234m}Pa (U X-ray)
112.8	35	^{234}Th
143.8	63	^{235}U
163.3	29	^{235}U
185.7	330	^{235}U
205.3	29	^{235}U

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[*] The decay scheme data were taken from the PCNUDAT Nuclear data file produced by the National Nuclear Data Center at Brookhaven National Laboratory, Upton NY

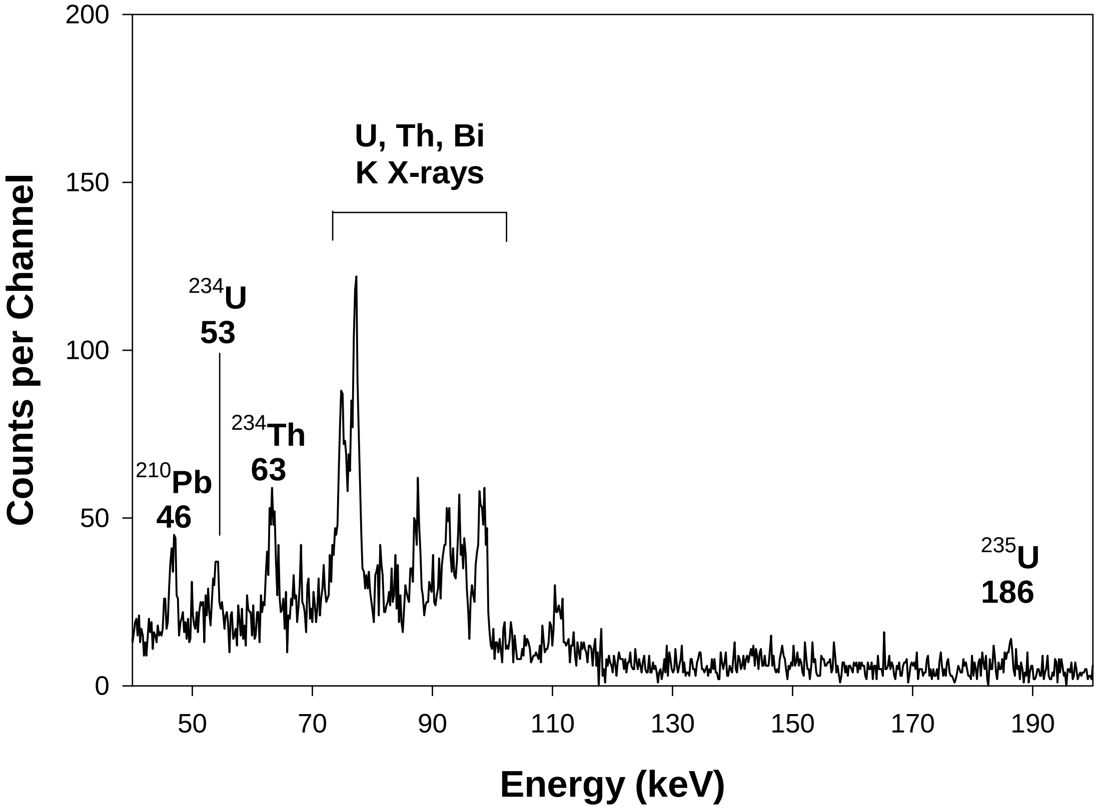
LIST OF FIGURES

Figure 1. Spectrum of uranium in secular equilibrium with its decay progeny taken for 10 minutes with the CdTe detector.

Figure 2a-2b. Spectra of chemically processed uranium oxide powder samples in secular equilibrium with the prompt decay products taken with a CdTe detector.

Figure 3a-3c. Selected regions of interest comparing the CdTe spectra of enriched uranium samples (enr.-U) with chemically separated natural uranium (nat-U).

Figure 4a-4c. Selected regions of interest comparing the performance of the CZT detector with the CdTe detector.



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150
100
50
0
Counts per Channel
50
70
90
110
130
150
170
190
Energy (keV)
U, Th, Bi
K X-rays
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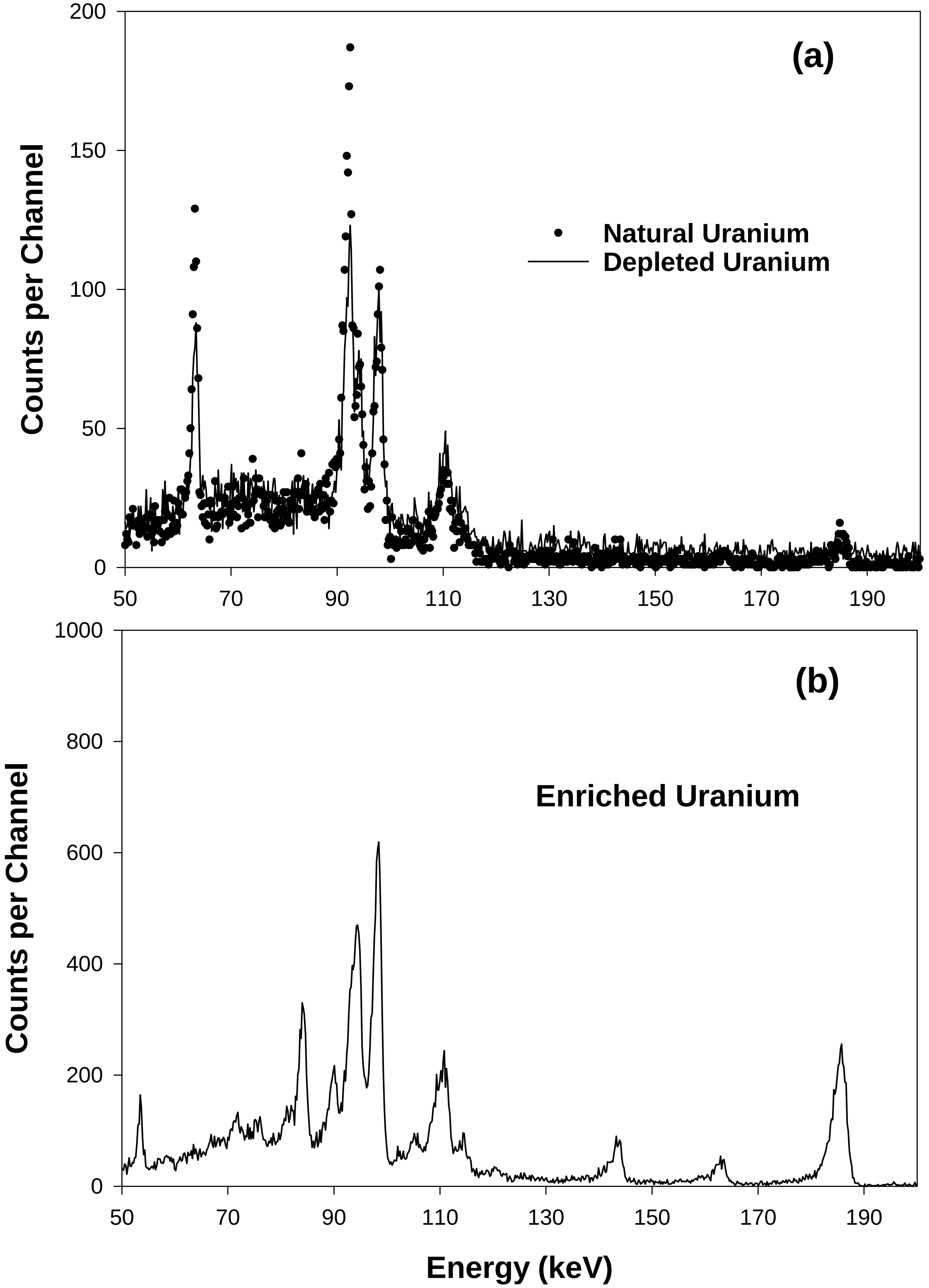
53
^{234}Th
63
^{210}Pb
46
^{235}U
186


(a)
Counts per Channel
0
50
100
150
200
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70
90
110
130
150
170
190
● Natural Uranium
Depleted Uranium
(b)
Counts per Channel
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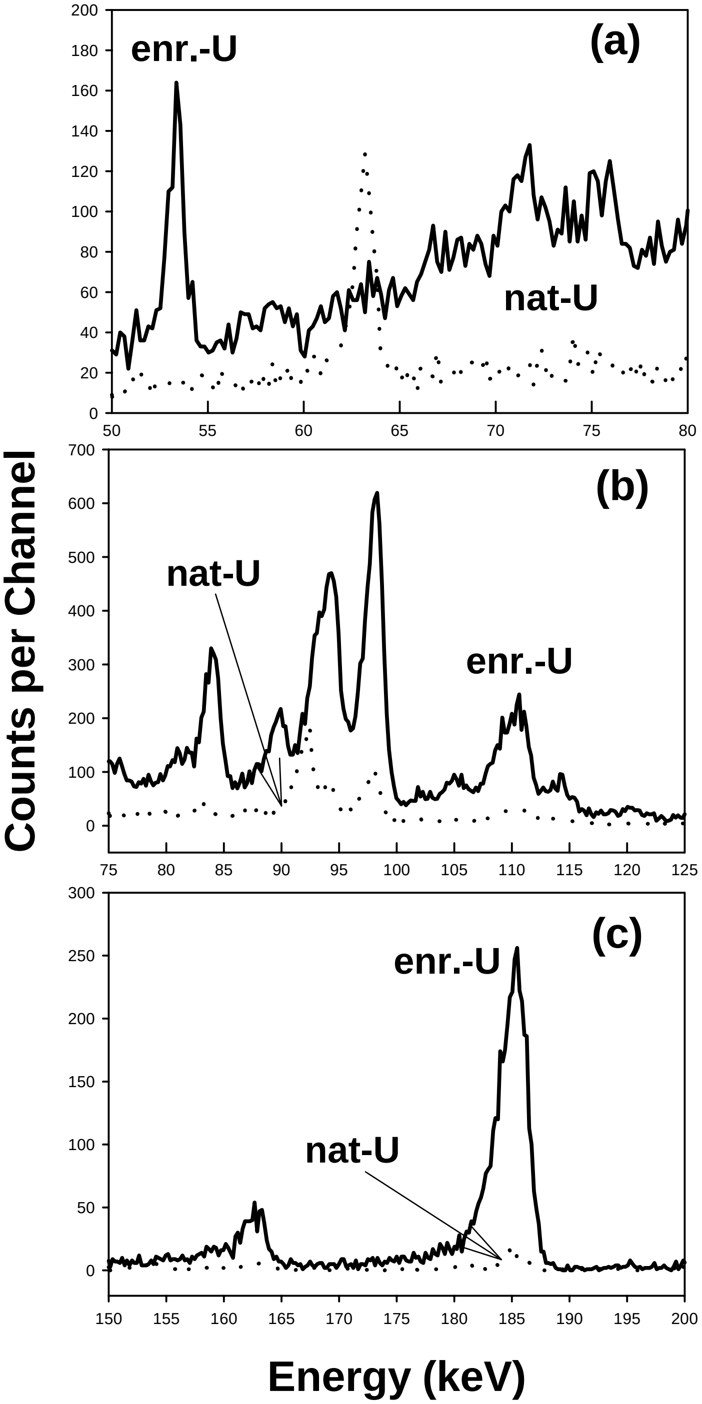
400
600
800
1000
Enriched Uranium
50
70
90
110
130
150
170
190
Energy (keV)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nat-U
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160
140
120
100
80
60
40
20
0
50
55
60
65
70
75
80
(b)
nat-U
enr.-U
700
600
500
400
300
200
100
0
75
80
85
90
95
100
105
110
115
120
125
(c)
enr.-U
nat-U
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185
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195
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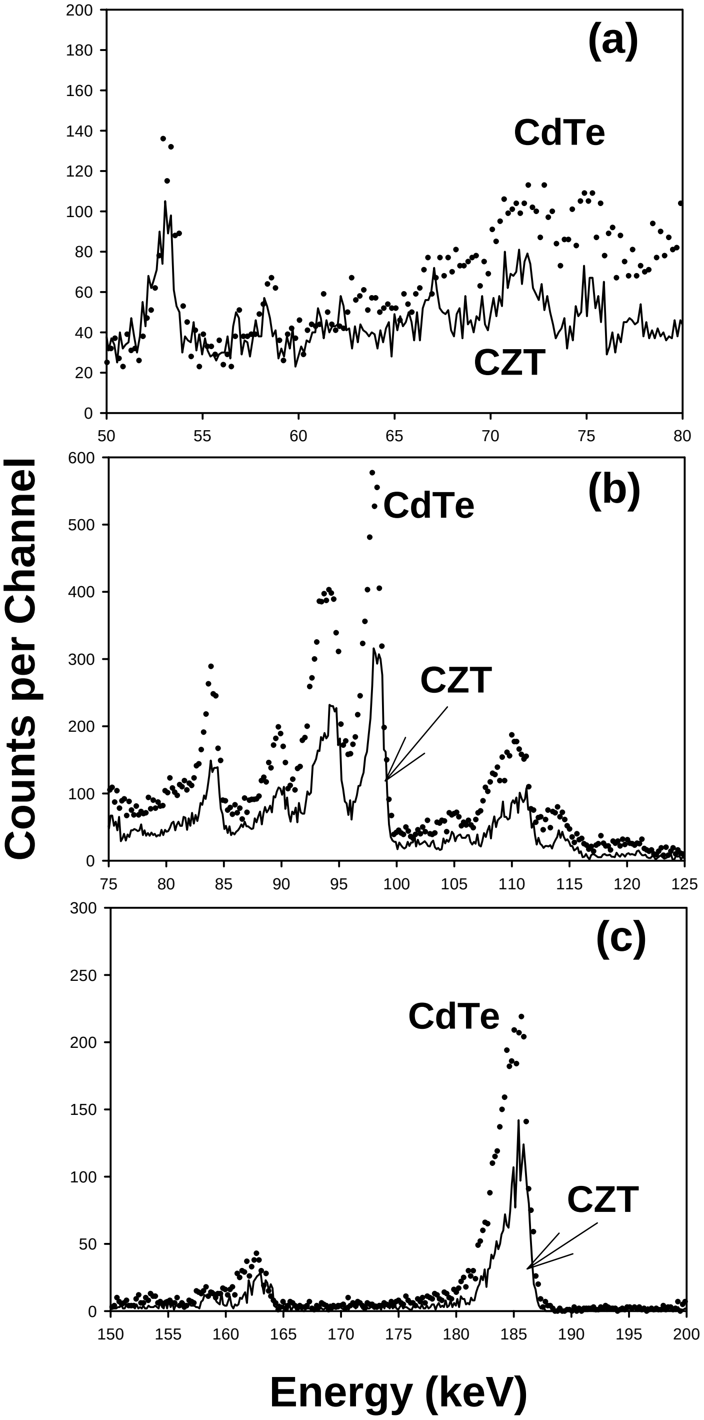
Counts per Channel
Energy (keV)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CdTe
CZT
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140
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100
80
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65
70
75
80
(b)
CdTe
CZT
600
500
400
300
200
100
0
75
80
85
90
95
100
105
110
115
120
125
(c)
CdTe
CZT
300
250
200
150
100
50
0
150
155
160
165
170
175
180
185
190
195
200
Counts per Channel
Energy (keV)