

Advantages and Use of CdZnTe Detectors in Safeguards Measurements

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Introduction

Room temperature semiconductor detectors - in particular CdZnTe and CdTe detectors - have been used in International Safeguards for more than 10 years [1, 2, 3, 4, 5]. With their properties, they complement the classical NaI and liquid nitrogen cooled Germanium detectors. There are certain classes of measurements where the unique features of these detectors come to bear, allowing the development and implementation of verification methods which otherwise would not be possible [6, 7, 8, 9]. In many cases, the use of these detectors has helped to increase both the efficiency and effectiveness of NDA methods for safeguards. The burden on operators and inspectors was reduced by allowing access and verification of the items with an NDA detector rather than their removal from storage for verification [10, 11]. The development of such detectors is still dynamic, and new and improved models become available almost every year. In addition, new applications occur in the field of inspections under the additional protocol and to combat illicit trafficking of nuclear material and radioactive sources.

The development of improved CdZnTe detectors depends on the availability of high quality raw material (CdZnTe single crystals), on a good knowledge of the material properties, contact and electrode design and the understanding of electrical field distribution and charge collection in the detector volume. Last but not least, the associated electronics must support the applications with small size and low power consumption. The performance of the newly developed detectors must be well understood and documented in order to plan their optimal utilization [12, 13, 14]. In addition, spectrum processing software must be developed/adapted to extract the needed information from CdZnTe gamma spectra [15, 16, 17].

Safeguards applications form only a relative small commercial market. The improvement of the CdZnTe material is largely driven by other fields, such as space research, medical imaging and industrial applications. The introduction of CdZnTe material, and its replacement of CdTe in the 90's, has brought a quantum leap in the performance of these detectors. Reliability and long term stability have considerably increased, the sensitive detector volume has grown by a factor of 10-20 and the resolution has improved by about a factor of 2-4. Reliable commercial suppliers are available. They can produce large volume detectors in the needed numbers. This is illustrated in Figure 1, where the performance of our standard model large volume detector - CZT/500(s) is shown. The raw material for the detectors was made by eV Products (US). The detectors and

preamplifiers are produced by RITEC (Latvia). The detector assemblies are commercially available from both suppliers.

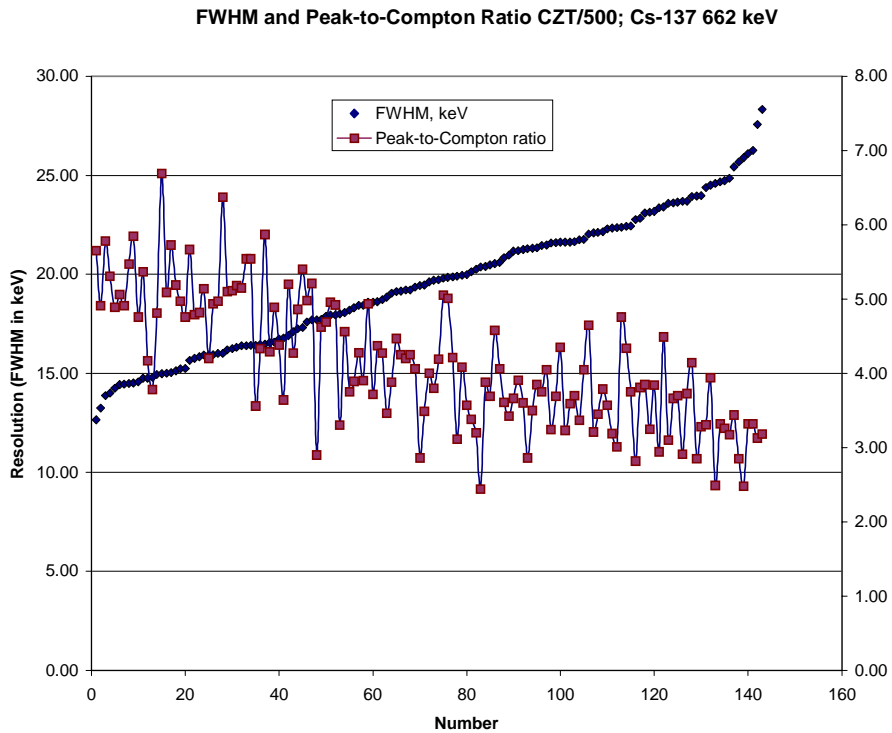


Figure 1 Resolution and peak/compton ratio of 144 CZT/500 detectors produced from 1997-2000 by RITEC using eV Products material

Stimulated by the introduction and use of these detectors by safeguards inspectors, we see new detector models appear on the market at regular intervals Their development is also driven by new requirements and applications, such as the need for high quality portable, hand-held isotope identification devices to combat illicit trafficking of nuclear material and radioactive sources. Since CdZnTe detectors have a much better resolution compared to NaI detectors, they are the only room temperature detector which allow hand-held and portable devices to provide a detection of Pu signatures behind significant lead or steel shieldings.

In this paper we briefly summarize their features and describe the verification methods used by the Department of Safeguards of the International Atomic Energy Agency, illustrating why CdZnTe detectors are unique in some of the applications. In the second part of the paper we describe new detectors and applications which are under development.

Comparison of Detector Properties

| Property: | Bandgap (eV) 300 K | Energy per e-h pair (eV) | Mobility.lifetime(e) ($\mu\text{m}^2/\text{Vs}$) | Mobility.lifetime(h) (μs) | Charge collection (nsec) | Atomic number | Density g/cm^3 | Maximal volume(cm^3) | Resolution FWHM (keV) |
|---|-------------------------------|-----------------------------|---|---|-----------------------------|------------------------|-----------------------------------|------------------------------------|--------------------------|
| Influences: | Leakage current Resolution | Resolution | Resolution | Resolution Low energy tailing | High rate operation | Photo peak/ Compton | Efficiency | Efficiency | Separation of lines |
| Ge | 0.67 | 2.96 (90 K) | high | high | 100 | 32 | 5.35 | 100 | 0.4-2 |
| Si | 1.12 | 3.61 | high | high | 10 | 14 | 2.33 | 0.1 | 0.2-1 |
| CdTe | 1.5 | 4.43 | medium | low | 100 | 50 | 6.2 | 0.1 | 0.2-20 |
| $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ | 1.57 | 4.64 | medium | low | 100 | 49.1 | 5.78 | 3.4 | 0.2-20 |
| Hgl ₂ | 2.1 | 4.15 | very low | very low | > 1000 | 62 | 6.4 | 4 | 0.2-30 |
| GaAs | 1.43 | 4.3 | very high | medium | <10 | 32 | 5.3 | 0.1 | 3 |
| NaI | | | | | 230* | | 3.67 | > 100 | 15-50 |

Decay constant of the light pulse

Table 1 Properties of common gamma detectors

In Table 1, the main properties of CdZnTe detectors are compared with other semiconductor detectors and a NaI(Tl) scintillation detector. What makes them unique is their wide band gap and the sufficiently low amount of energy needed to create an electron/hole pair. The wide band gap allows their use at room temperature and the energy per electron/hole pair offers much better resolution compared to other gamma detectors which can be operated at room temperatures, such as the widely used NaI detectors. The high value of the atomic number (Z) of CdZnTe leads to a high intrinsic photopeak efficiency and a favorable photopeak/compton ratio, even when the detector volume is relatively small. From the table, however, we see also problems associated with the use of these detectors. Both the mobility and lifetime of the electrons and the holes are quite different. Due to their low mobility and short lifetime, holes are trapped very quickly and cannot contribute to the formation of a full energy signal. Consequentially, in a gamma-ray spectrum, the corresponding pulses contribute to a useless continuum below the photopeak or degrade the photopeak resolution by contributing to the low-energy tailing. As explained below, significant effort is needed to compensate this deficiency and to obtain gamma spectra with well pronounced gamma peaks and good peak/compton ratio. A second disadvantage of CdZnTe is the difficulty of obtaining large, homogenous single crystals - a precondition for making large volume detectors. The maximum volume of a single element detector is presently limited to about 2.3 cubic cm. However, we expect significant improvements in the future as the quality of the as-grown material is improved.

CdZnTe Detectors Assemblies in Routine Use

In Table 2 an overview is given on the detector models which are routinely used by the Department of Safeguards and the verification methods associated with them. The SDP310ZXX family (Fig. 2) is based on a miniature preamplifier and detector which is placed in a 8 mm wide stainless steel housing, 40-60 mm long. The detector cable is integrated and can be extended, depending on application, up to 20 meters and more. The sensitive volume in these probes ranges from a few mm^3 (just large enough to produce a

photopeaks up to 1 MeV) to 60 mm³ - the largest detector volume which fits into the slim steel housing.

| Detector type | Description of application |
|-----------------------------------|---|
| SDP310Z20s | Verification of short living spent fuel assemblies in the accessible core of research reactors (SFAT) |
| SDP310Z60s | In-situ verification of WWER-440 spent fuel (SFAT) |
| CZT/500s | In-situ verification of PWR and BWR spent fuel (SFAT) |
| SDP310Z60 | Verification of CANDU bundles with long cooling time stored in stacks under water (CBVB) |
| SDP310Z20s | Verification of CANDU bundles with short cooling time stored in stacks under water (CBVB) |
| SDP310Z20 or Z60 | Verification of CANDU dry storage canisters filled with baskets or modules of spent CANDU bundles (CANDU Canister Verifier) |
| CZT/500 or CZT/500s and SDP310Z60 | Attribute verification of non-irradiated U and Pu samples; in-situ verification of non- irradiated WWER 440/1000 assemblies |
| SDP310Z60 | Unattended verification of irradiated items in a hot cell (MMCA in unattended mode of operation) |
| SZT/500s | Verification of non-irradiated LWR MOX assemblies stored under water (FMAT) |
| SDP310Z20s | Attribute test of isolated spent fuel assemblies and non-fuel items (IRAT) |

Table 2 Overview on detector types and verification methods routinely used in the Department of Safeguards

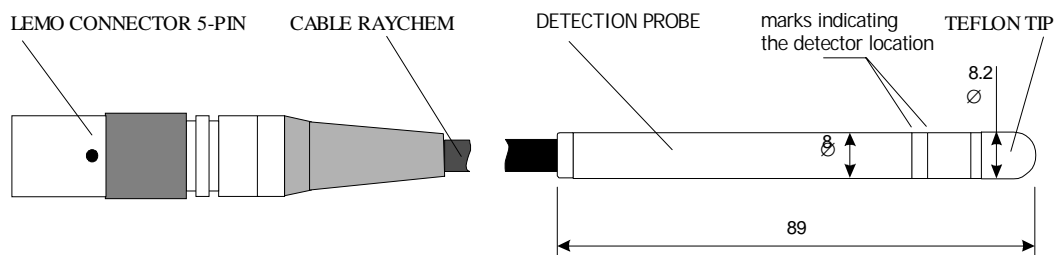


Figure 2 Schematic drawing of a SDP310Z20,60 detector assembly with cable connection (all dimensions in mm)

The CZT/XXXX family (Fig. 3) covers detectors with the largest volume presently routinely used. The standard model is the CZT/500 (10X10X5 mm³), while the largest commercial detectors (CZT/1500) have a geometric volume of 1687 mm³ (15x15x7.5 mm³).

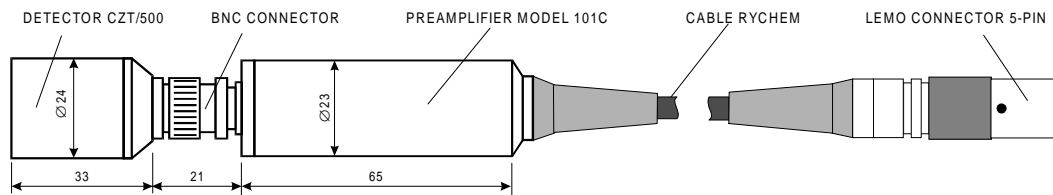


Figure 3 Schematic drawing of a CZT/500 detection probe with detachable preamplifier

All the described detectors are of hemispheric design to achieve single charge collection. Their hemispherical geometry (see Fig. 4) compensates for the much inferior hole collection efficiency found in today's CdZnTe by modifying the internal field in a manner that emphasizes the collection of electrons. The extension of the cathode over the vertical edges of the detectors, along with one of the major horizontal surfaces, and the use of a small area anode results in a concentration of the electric field lines in the region of the anode. This results in a corresponding increase of the weighting potential in this volume. Electrons generated in the bulk of the detector volume can, due to their relatively long lifetime, travel to the high field region. The induced signal due to this motion is small, but increases dramatically in proportion to the increased electric field when the electron approaches the anode [18]. The holes travel into the opposite direction, but contribute much less to the spectrum. Because of their low mobility and short lifetime, they are trapped more quickly. In addition they move in the peripheral region of the detector where the weighing potential and therefore the induced signal are low. These effects lead to a much peak/compton ratio compared to a planar detector if standard Gaussian shaping is used.

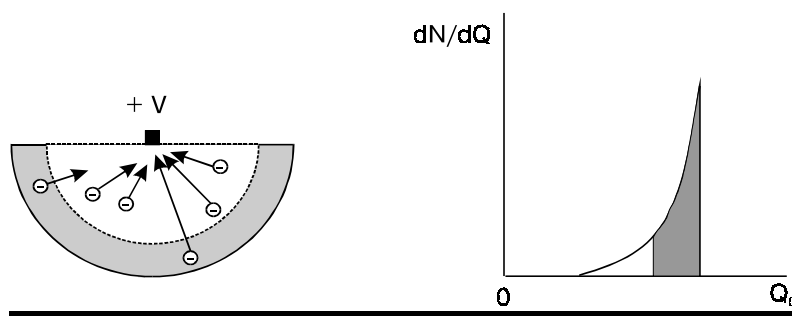


Figure 4 Schematic drawing of a CdZnTe detector with hemispheric geometry

Verification Methods in Routine Use

The methods summarized with the detectors in Table 2 can be classified into 4 groups. In the first group, CdZnTe detectors replace NaI detectors because they are more convenient to use (smaller size of the collimator, better resolution and temperature stability, access to space limited locations).

The second group consists of spent fuel verifiers for light water reactor (Spent Fuel Attribute Tester, SFAT) and Irradiated Item Attribute Tester (IRAT). In this group CdZnTe detectors are used for several reasons. Firstly, they permit the design of smaller and lighter detection systems which is essential for equipment used in spent fuel ponds above stored assemblies. Secondly they reveal - due to their improved resolution - more details in the gamma spectrum which can be used to assess qualitatively cooling time and burnup of spent fuel assemblies and to distinguish them from irradiated non-fuel items. The level of details which can be seen in the CdZnTe spent fuel spectrum is shown in Fig. 5. Gamma lines of La-140, Zr/-Nb-95, Cs-134 and Cs-137 can clearly be identified. The presence of La-140 tells us that the assembly was irradiated in the core during the last 3 months. The measurements can be performed in-situ, without moving the item using the SFAT device [10].

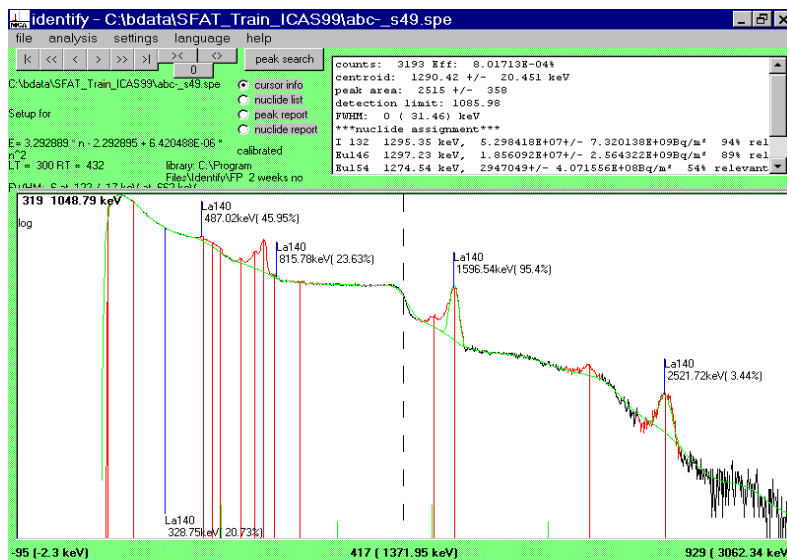


Figure 5 Gamma spectrum of MTR spent fuel assembly with short cooling time measured with Spent Fuel Attribute Tester (SFAT) and SDP310/Z/20s detector

The third group of applications is related to the verification of spent CANDU bundles stored in stacks under water or in storage canisters. In this application, CdZnTe detectors are the only gamma detectors that can be used successfully. The principle of the method is explained in ref. [7, 8, 19].

The fourth emerging group of applications is their use in unattended radiation monitoring systems [9]. Compared to HPGe detectors they do not require liquid nitrogen cooling and compared to NaI detectors they exhibit a better resolution.

The total inventory of hemispheric CdZnTe detectors on stock and in use in the Department of Safeguards is presently more than 300.

New Developments

In Table 3 an overview on new detector developments and associated verification methods is given.

| Detector type | Description of application |
|--|---|
| Large volume CZT detector with co-planar electrodes | Various detector geometries (cylindrical and cubical) are under evaluation for U and Pu attribute tests and for use in portable isotope verifiers, eV Products and Los Alamos (USA) |
| CZT/500 pluggable detector module | Under evaluation for use in the fieldSPEC hand held gamma spectrometer – isotope identifier (RITEC, Target/BICRON) |
| Pluggable detector module with co-planar electrodes | In the design phase, for use in portable isotope identifiers, Los Alamos (USA) |
| High resolution PIN CdTe detectors with Peltier cooling | Prototype available, detector size to be enlarged, resolution comparable to HPGE detectors, verification of U-235 without calibration |
| SDP310Z01 and Z05 | Verification of CANDU bundles with cooling time < 1/2 year |
| Slim detector assembly with low detector sensitivity | U fluorescence X-ray detection, for use between bundles |
| Backshielded, compact CZT/500s hand held detector module | Under evaluation to replace presently used CZT/500 with detachable preamplifier |
| Peltier cooled Z60s detector (SDP400) | For use SFAT use in spent fuel ponds with water temperatures up to 70 degrees C, under development (RITEC) |
| Multi element detector 4XCZT/500 | Under evaluation for attribute verification of non-irradiated U and Pu samples |
| Large area HgI-2 detector | Under development (Constellation Technology Corporation USA) |
| Large area Pixel array detector | Under development (Technion, Israel) |
| Miniature detection probe with CAP detectors | Commercially available, eV Products (USA); under evaluation |

Table 3 New detectors and applications

The table shows that the development of new detectors for safeguards applications and prevention of illicit trafficking is mainly focused on the following directions:

- i) Detectors with larger volume/area also configured as plug-in modules for hand held gamma spectrometers [20, 21].
- ii) Detectors with improved energy resolution [22].
- iii) Detectors with extremely low sensitivity to operate in contact with spent fuel assemblies [21].
- iv) Specialized electrically cooled detector assemblies for high temperature operation [21].

The most important direction is the development of large volume detectors. These are of strategic importance for various applications. There are several ways to achieve detectors with large volume/areas:

- i) Large "single slab" detectors
- ii) Multi detectors elements
- iii) Detectors consisting of many small detector pixels

Large single slab detectors can be manufactured by either scaling up the hemispheric detector design, or by using a new technology - detectors with co-planar electrodes. Scaling up the hemispheric detector design is presently limited to detector volumes of about 2.3 cubic cm. For larger volumes it become increasingly difficult to maintain the desired level of performance with this design concept, due to the increased probability of electron trapping. In addition, a detector bias of 2 kV or more is needed, which is difficult to implement in small detection probes. A different approach uses coplanar electrodes on the surface of a planar CdZnTe detector. The Co-Planar Grid (CPG) approach is based on a similar effect to the hemispherical detector, namely a modification of the electric field to result in the major part of the signal being generated close to the anode. However, this is achieved in the CPG by the use of 2 interleaved electrodes held at a differential voltage (as shown in Fig. 6)

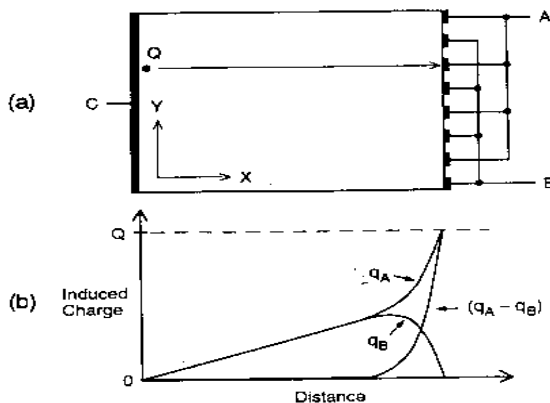


Fig. 2. (a) Basic structure of a coplanar grid detector. (b) Induced signals at the two electrodes and the subtracted signal.

These two electrodes each have their own preamplifier, and both are connecting to the inputs of a differencing amplifier. The electrons drift in the electric field in the detector volume, generating the same induced charge in each of the grids. When the output of the differencing amplifier is examined, the net signal is zero, i.e. charge movement in the bulk of the material does not result in a pulse at the output. However, when the electron approaches the grid electrodes, it starts to deviate towards the more positive grid. At this point, the signal from one amplifier goes high, and the other preamplifier mirrors this and goes low. The resultant signal from the differencing amplifier now has an amplitude corresponding to the energy deposited in the crystal, but is free from any tailing effects due to charge trapping in the bulk material (for further discussion of the CPG effect see ref. [23, 24]). The third direction to obtain large detector areas is the combination of many single detector pixels. This technology is largely used in medical and cosmic X-ray imaging and a R&D effort is under way to use adapt this technology for designing a large area detector for hand-held instruments [25].

Figure 7 shows a Ra-226 gamma spectrum taken with a detector with coplanar electrodes. It is evident that the detector with coplanar electrodes exhibits a very good peak shape (low energy tailing) which improves processing by spectrum evaluation software to extract peak energy and area.

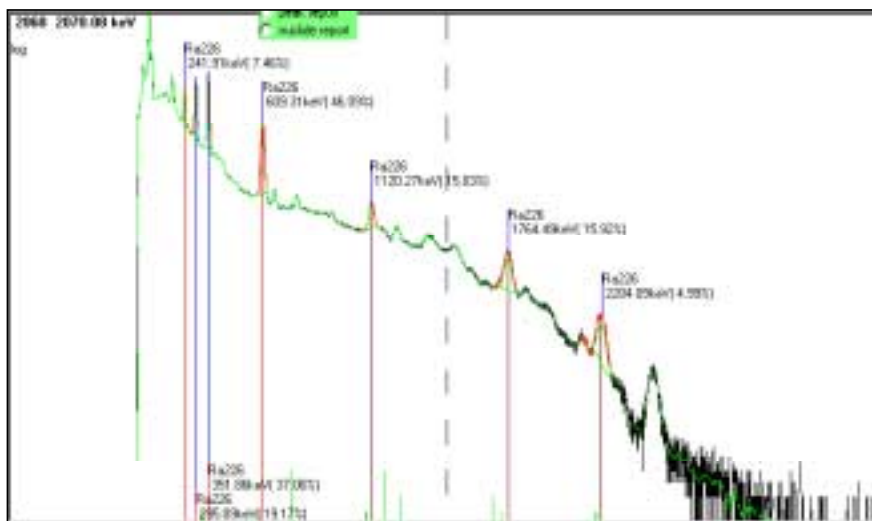


Figure 7 Ra-226 spectrum taken with cylindrical CGP detector (diameter 10mm, thickness 7.5 mm). Model spectrum and line identification was done with the Identify programme [16].

We are convinced that one of the most important applications of large volume/area detectors is their use in portable, hand-held isotope identification tools. The present generation of commercially available equipment is based on scintillation detectors. These detectors have significant deficiencies (moderate resolution, non-linear

energy scale, temperature and countrate sensitivity, etc.) which make it very difficult or even impossible to satisfy the requirements of the various users. CdZnTe detectors do not have these problems. Their energy calibration curve is linear, the temperature dependence is small (<0.1% peak position shift/°C,) and the energy resolution is better over a wide range of energies. These detectors can work without stabilization and show a constant performance over long periods.

First test results have shown that the improved resolution of these detectors even allows the detection of typical radiation signatures of Pu sources when shielded with several cm of steel or lead (see Figures 8 and 9).

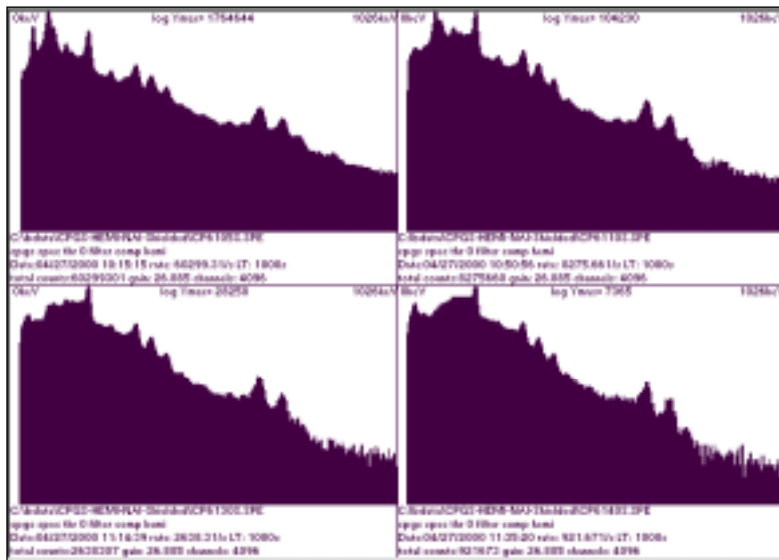
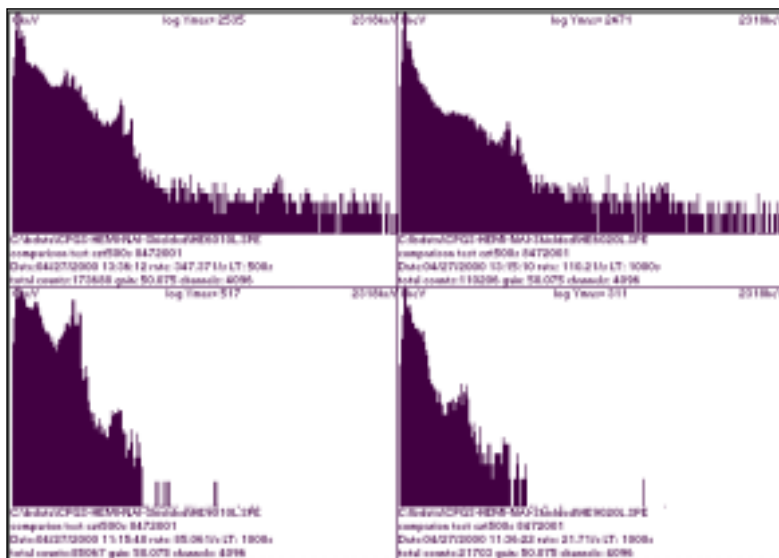


Figure 8 Gamma signatures of high burnup Pu sample taken with CPG CdZnTe detector (15X15X7.5 mm³) behind 5, 10, 30 and 40 mm steel shielding

Figure 9 Gamma signatures of high and low burnup Pu samples taken with hemispheric CZT/500s detector behind 10 and 20 mm lead shieldings



A second new direction is the design of electrically cooled CdTe and CdZnTe detection systems. There are two primary applications for this type of detectors –

- i) CdZnTe detection probes which can be operated at high temperatures (up to 70 degrees C). This is needed since in some spent fuel storage ponds the temperature is so high that standard detection probes do not work anymore.
- ii) CdTe detection systems with energy resolution close to that of the first Li-drifted Germanium detectors. To achieve this resolution, a commercial prototype detection system is nearing completion [22].

A third new development refers to detection of U X rays with a low sensitivity detector in contact with irradiated CANDU bundles. In some storage geometries, the only way to verify the bundles would be to insert a flat detection probe in the 16 mm gap between adjacent bundles. This requires the design of a detector with very low sensitivity and which detects the fluorescence X-rays of the adjacent bundle when lowered into the gap. Preliminary tests have shown that the U X-rays can easily be detected using a CdZnTe detector in close proximity to the bundles [19]. The problem, however, is to reduce the detector efficiency to such a level that the detector is not overloaded.

Summary, Outlook, Conclusions

CdZnTe and CdTe detectors have a proven record in safeguards verification measurements and related applications. They have become for the most versatile room

temperature gamma detectors, covering a wide range of applications. Their properties are ideal for field measurements and for the design of small detection probes which can be brought close to the items to verify even if there are space restrictions. A new, important usage is envisaged in the use of large volume detectors in portable and hand-held isotope identification devices needed to in the set of technical measures to combat illicit trafficking of nuclear material and radioactive sources and other related applications such as waste characterization and health physics. These detectors have significant advantage compared to the presently used NaI detectors. Their sensitivity is still smaller compared to NaI and HPGe detectors, but already sufficient for many applications. Detectors with larger detectors will become commercially available in the near future. Further support should be provided to enhance R&D to improve the quality of the detector raw material and the detector supply.

Acknowledgment

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