



# $\gamma$ -Ray Spectrometry on Nuclear Material Using a CdZnTe Detector

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(Received 13 February 1996; in revised form 21 March 1996)

A planar CdZnTe detector of dimensions of  $10 \times 10 \times 5$  mm was tested for its application to  $\gamma$ -ray spectrometry on nuclear material. The outcome from the analysis of a spent nuclear fuel pin, natural uranium solutions, plutonium powder and mixed oxide pellet (MOX) are reported. Quantification could be performed on the spectra obtained from samples containing only uranium. However, on the basis of the spectra obtained, it was found possible to qualitatively discriminate between the different kinds of nuclear material analysed. Copyright © 1996 Elsevier Science Ltd

## Introduction

Non-destructive assay (NDA) of nuclear material is routinely performed through, among other techniques, high energy resolution ( $< 0.3\%$ )  $\gamma$ -ray spectroscopy using germanium detectors. However, the need for cooling results in bulky detectors and their use becomes problematic when employed in limited space under unattended conditions. Detectors based on NaI scintillation material, with high efficiency and capable of operating at room temperature have also been employed. However, their poor energy resolution of about 7% (Fig. 2) makes them unsuitable for the analysis of complex  $\gamma$ -ray spectra. Over the last 10 years, there is an increasing interest towards the use of compact  $\gamma$ -ray detectors, e.g. CdTe, CdZnTe. Their small size and the possibility to operate them at room temperature renders them portable, and their good energy resolution makes them suitable for remote applications on nuclear material.

The feasibility of performing  $\gamma$ -ray spectroscopy of different nuclear material using a CdZnTe detector was studied.  $\gamma$ -ray spectroscopy was carried out on spent nuclear fuel, fresh mixed oxide material (MOX) and natural uranium solutions as well as standard radioactive sources for calibration purposes.

## The CdZnTe Detector in $\gamma$ -ray Spectroscopy

The CdZnTe crystal ( $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ ) is produced by substituting 10% cadmium by zinc in the CdTe

compound. In principle, the high atomic number of Cd, Zn and Te (48, 30 and 52 respectively), results in CdZnTe crystals with high stopping power for  $\gamma$ -ray spectroscopy. In addition, the large bandgap energy (1.6 eV) of this material allows stable operation at room temperature. Relatively inexpensive, small and efficient crystals for  $\gamma$ -ray spectroscopy can be obtained, capable of operating at ambient temperature.

## Experimental

The planar CdZnTe detector of 5 mm thickness and  $100 \text{ mm}^2$  sensitive area used in this work was purchased from eV Products Inc. (Saxonburg, Pa, U.S.A.). The nuclear instrumentation comprised a high voltage unit (bias 1500 V), a standard amplifier (Ortec 572) with shaping time of  $0.5 \mu\text{s}$  and an ultra-high speed analog-to-digital converter ( $0.75 \mu\text{s}$  fixed conversion time for high resolution and count rate spectroscopy).

Standard radioactive sources of  $^{137}\text{Cs}$  (3.5 MBq),  $^{133}\text{Ba}$  (3.5 MBq) and  $^{241}\text{Am}$  (200 kBq) were used for calibration purposes over the energy range 60–700 keV. This range covers the  $\gamma$ -rays from uranium, plutonium and fission products used routinely in the analyses of nuclear material.  $\gamma$ -ray spectrometry was performed on a  $\text{UO}_2$  spent nuclear fuel pin,  $\text{PuO}_2$  powder MOX pellet and natural uranium solution. The studies on spent fuel were performed at the  $\beta$ - $\gamma$  hot cell facilities where the fuel is stored. The cell wall was made of concrete with a thickness of 1 m. A collimator of lead and tungsten,

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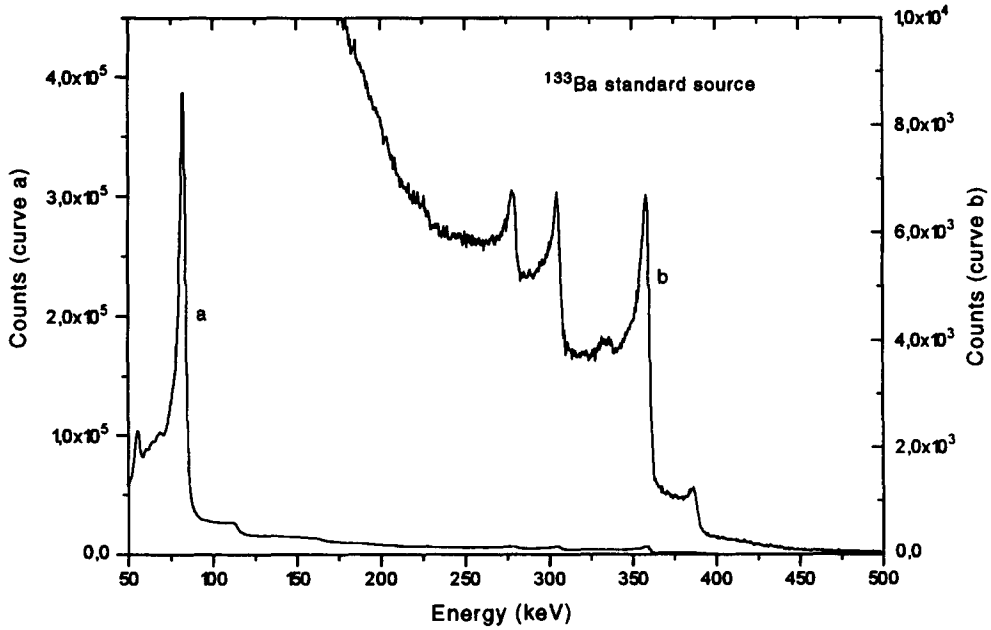


Fig. 1.  $\gamma$ -ray spectrum of  $^{133}\text{Ba}$  standard source using the CdZnTe detector.

1.2 mm diameter, was incorporated into the wall allowing  $\gamma$ -ray spectrometry of pins with a detector situated outside the cell. The powder and solution samples were analysed in contact with the CdZnTe detector.

**Results and Discussion**

$\gamma$ -ray spectra obtained with the CdZnTe detector, for a counting period of 500 s, for the standard radioactive sources  $^{133}\text{Ba}$  and  $^{137}\text{Cs}$  are shown in Figs 1 and 2. The CdZnTe detector was characterised in

terms of energy resolution, efficiency and peak-to-valley ratio using those spectra. The  $\gamma$ -ray spectrum of  $^{133}\text{Ba}$  reveals well-resolved energy lines. However, the peak-to-valley ratio deteriorates above 100 keV.  $\gamma$ -ray spectra of  $^{137}\text{Cs}$  obtained with a CdTe, HPGe and NaI detectors are included in Fig. 2 for comparison purposes from the view points of energy resolution and peak-to-valley ratio. The highest energy resolution and peak-to-valley ratio are obtained with the HPGe detector, while the NaI offers the lowest energy resolution. The CdZnTe and CdTe detectors have comparable resolution, which is

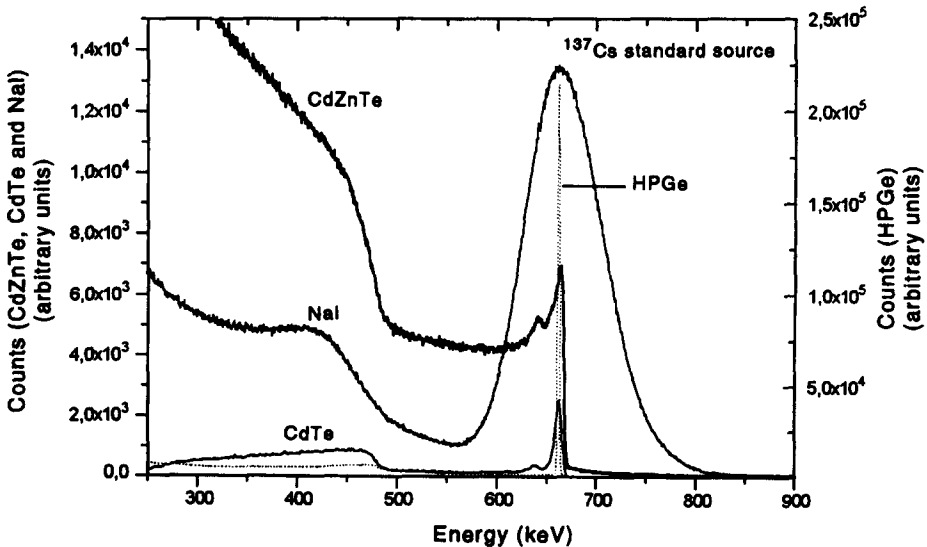


Fig. 2.  $\gamma$ -ray spectra of the  $^{137}\text{Cs}$  standard source obtained with different detectors (HPGe, CdTe, CdZnTe, NaI).

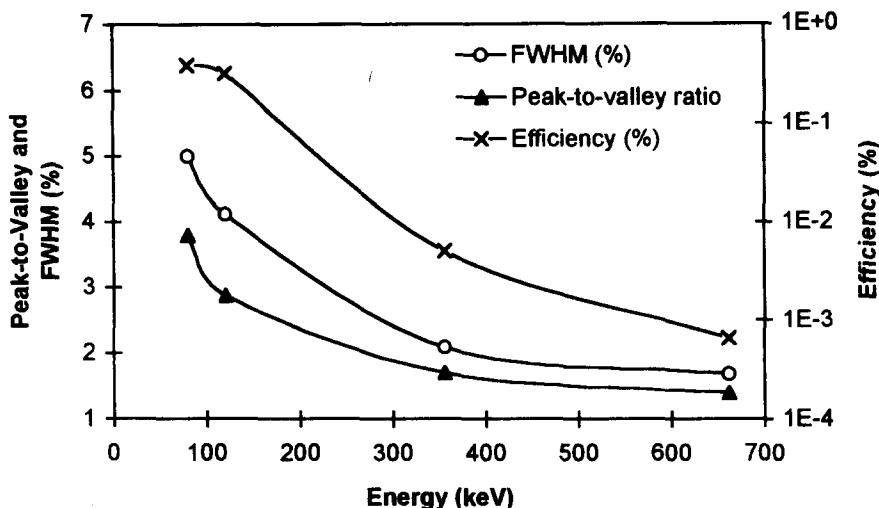


Fig. 3. Efficiency, energy resolution (FWHM) and peak-to-valley ratio vs energy for the CdZnTe detector used in this work.

about a factor of 5 lower than that of the HPGe detector. However, the CdZnTe detector is by far inferior to the others from the peak-to-valley ratio view point. This is due to charges lost from recombining or trapping of the holes.

The trend of energy resolution, efficiency and peak-to-valley ratio as a function of energy are shown in Fig. 3 for the CdZnTe detector under the experimental conditions used in this work. A drastic decrease in efficiency occurs which is up to a factor of 1000 in the energy range of 100–700 keV. In the case of the HPGe detector, this factor is less than 10. The energy resolution for the CdZnTe detector varies from 5 to 2% in the above energy range. However, the peak-to-valley ratio is much higher at low energies further supporting the suitability of the CdZnTe detector at these energies. The CdZnTe detector should thus preferentially be used for lower energies (< 200 keV).

The long-term scope of the work is the application of the CdZnTe detector in the nuclear fuel cycle. In this context,  $\gamma$ -ray spectrometry was carried out on a PWR UO<sub>2</sub> spent nuclear fuel pin (burn-up 55 GWd/t, 5 years cooling).  $\gamma$ -ray spectra obtained with the CdZnTe and HPGe detectors under identical conditions are shown in Fig. 4, for the energy range of 450–750 keV. Verification of the declared burn-up or cooling time would be carried out using the isotopic ratio (<sup>137</sup>Cs/<sup>134</sup>Cs) on the basis of the  $\gamma$  lines at 662 and 605 keV (Foggi and Koch, 1982). The low peak-to-valley ratio of the CdZnTe detector in this energy range results in small peaks just above the background. Clearly, quantification of this spectrum is difficult in this range. In contrast, the superior peak-to-valley ratio of a CdTe detector, when complemented by the Charge Loss Corrector (CLC) (Richter and Siffert, 1992), results in well-resolved  $\gamma$  lines allowing quantification of the spectrum (see inset

in Fig. 4) (Abbas *et al.*, 1996). It must be pointed out that CdTe (2 × 2 × 2 mm) and CdZnTe (10 × 10 × 5 mm) crystals used in this work were grown with two different methods, namely Travelling Heat Method (THM) and Bridgman High Pressure (BHP) respectively (Mathy *et al.*, 1993). The two growth methods result in different transport properties and hence spectroscopic performances. The THM gives the best material for spectroscopy whereas the BHP provides larger crystals. In addition, the lack of CLC for BHP CdZnTe detector prevents any further improvement of the  $\gamma$ -ray spectrum.

The performance of the CdZnTe detector in the  $\gamma$ -ray energy range below 200 keV was studied by counting unirradiated samples of uranium and plutonium. In this energy range, the peak-to-valley ratio is high, while the energy resolution, although low, is sufficient for  $\gamma$ -ray spectroscopy.

$\gamma$ -ray spectra of PuO<sub>2</sub> powder obtained with the CdZnTe and a HPGe detector are shown in Fig. 5. The only  $\gamma$  lines well resolved in the spectrum of the CdZnTe are those of <sup>241</sup>Pu–<sup>237</sup>U at 148 and 208 keV. The energy range around 100 keV cannot be explored because of the complexity of the spectrum of plutonium. The  $\gamma$ -ray spectrum of a MOX PWR pellet, using the CdZnTe detector, was dominated by the plutonium  $\gamma$  lines. A differentiation of the MOX pellet and PuO<sub>2</sub> powder would therefore be difficult. However, the spectra obtained allows the presence of plutonium to be identified in the samples analysed.

$\gamma$ -ray spectroscopy was also performed on standard reference materials consisting of solutions of various concentrations of natural uranium (50–400 g U/L) in nitric acid. This range corresponds to that encountered in a commercial reprocessing plant. A typical  $\gamma$ -ray spectrum of these samples, obtained with the CdZnTe detector, for a counting period of 1800 s, is

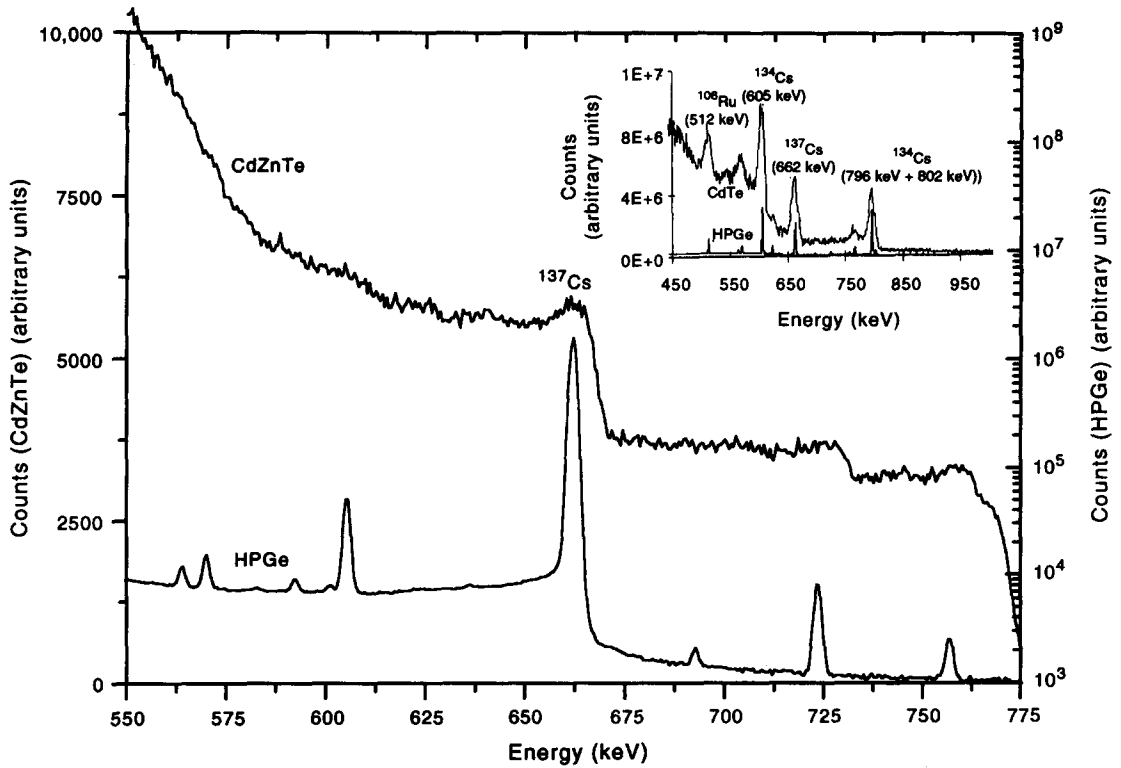


Fig. 4.  $\gamma$ -ray spectra of a spent nuclear fuel pin obtained with CdZnTe and HPGe detectors. Inset:  $\gamma$ -ray spectra of spent nuclear fuel pin obtained with CdTe ( $2 \times 2 \times 2$  mm) and HPGe detectors.

shown in Fig. 6. The corresponding spectrum with a HPGe detector is included for comparison. Each sample contained 5 mL solution in a Plexiglas container ( $2 \times 4.5 \times 1.2$  cm). The  $\gamma$  lines of  $^{235}\text{U}$  at 186, 163 and 144 keV are well resolved by the

CdZnTe detector. The measurement of the  $^{235}\text{U}$  concentration is carried out by determining the 186 keV peak area. The CdZnTe detector can be successfully used for this task as is shown from the experimental calibration curve relating the 186 keV

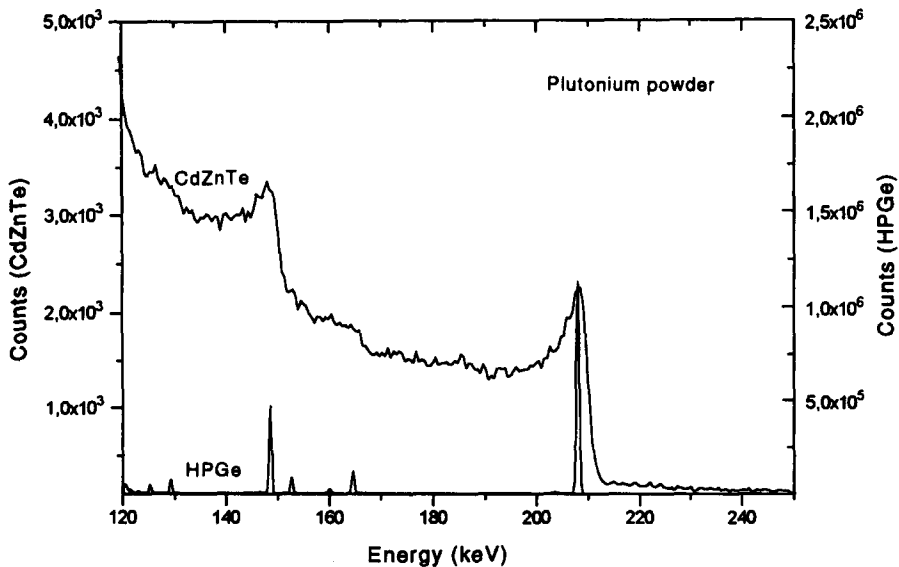


Fig. 5.  $\gamma$ -ray spectra of a plutonium powder sample obtained with the HPGe detector and with the CdZnTe detector.

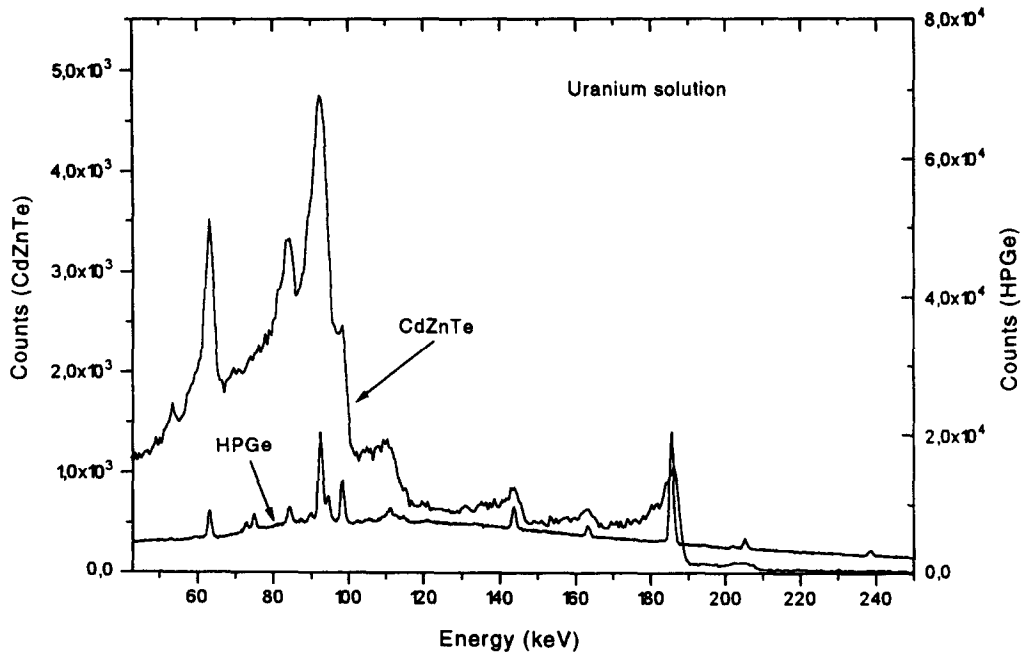


Fig. 6.  $\gamma$ -ray spectra of uranium solution obtained with the HPGe detector and with the CdZnTe detector.

peak area to concentration of natural uranium in the standards (Fig. 7).

### Conclusion

The CdZnTe detector yielded spectra of adequate resolution and peak-to valley ratio at energies less than 200 keV. In this range, complex spectra from nuclear material containing uranium or plutonium were obtained. Quantification of the spectra obtained from samples containing only uranium could be performed. The plutonium in the MOX or PuO<sub>2</sub>

samples could only be qualitatively identified. In the energy range between 500 and 800 keV, the poor spectra quality prevents any verification of the declared burn-up or cooling time of the spent fuel studied. Nevertheless, sufficient information is obtained to identify it as being spent fuel. It is evident that, on qualitative basis, the different types of nuclear material analysed could be discriminated. The small size, portability, ambient operation and performance render CdZnTe detectors capable of preliminary *in situ* identification of unknown nuclear material.

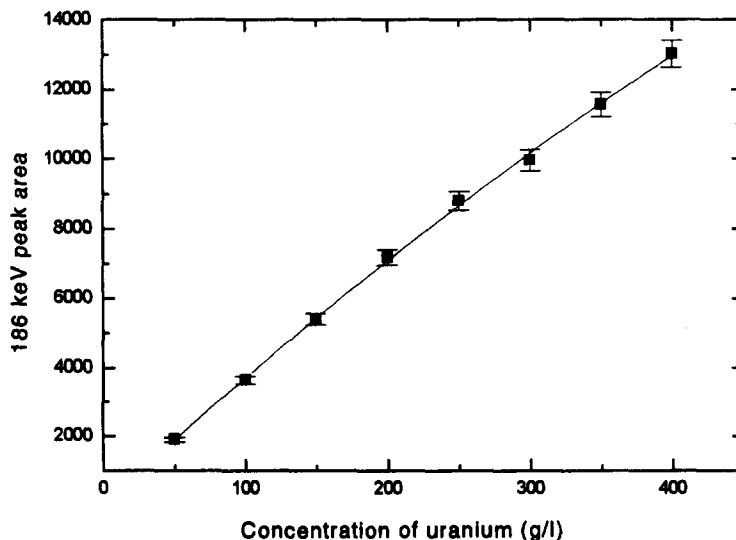


Fig. 7. Calibration curve of peak area of uranium (186 keV) vs concentration of uranium.

*Acknowledgements*—The authors gratefully acknowledge the technical support of P. Lajarge and A. Logreco.

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