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Gamma spectrometry of spent nuclear fuel using a miniature CdTe detector

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Abstract

The application of a planar miniature CdTe detector for gamma spectrometry has been tested by analysing standard radioactive sources and spent nuclear fuel. The detector was connected to a charge loss corrector amplifier and yielded an energy resolution (FWHM) of 7.2 keV at 662 keV. The application of the CdTe detector to spent fuels resulted in gamma spectra of well resolved photopeaks for the fission products ^{134}Cs and ^{137}Cs. The ratio of the two fission products, complemented by isotopic correlations, verified the burnup of the fuel to within 10% of that declared by the operator.

1. Introduction

Verification of the burnup of spent nuclear fuel is an important component of safeguards and criticality safety during their transport and storage. Non-destructive assay of the material, by measuring their neutron and gamma emissions, is one of the verification procedures pursued in safeguards [1]. High resolution gamma spectrometry based on germanium (Ge) detectors has been routinely applied; however, the need to cool the detectors results in bulky devices. In the monitoring and assaying of nuclear material there is an increasing movement towards compact devices for unattended and remote operation within an integrated monitoring system. The use of semiconductor compounds, e.g. CdTe, CdZnTe, has therefore been explored for application to gamma spectrometry [2]. Their small size, good energy resolution and, above all, operation at room temperatures, renders them portable and suitable for remote applications on nuclear material under dry or wet conditions.

This paper reports the findings from a study on a commercial miniature planar cadmium telluride (CdTe) detector. Gamma spectrometry was performed using standard sources and directly on irradiated nuclear fuel. The former were used for calibration purposes, e.g. determination of resolution and efficiency, and for optimisation of the electronics. The latter, in the form of spent fuel pins and pellets, were used to demonstrate the operation of such

detectors on samples of high gamma emission and the subsequent verification of the declared burnup of this material. The studies were performed in the laboratories and hot cell facilities of the Institute for Transuranium Elements in Karlsruhe.

2. The CdTe crystal in gamma spectrometry

The suitability of CdTe for gamma spectrometry is twofold. Firstly, the high atomic numbers of cadmium ($Z_{Cd} = 48$) and tellurium ($Z_{Te} = 52$) ensure high stopping power for gamma rays. The photoelectric absorption coefficient for CdTe as a function of energy is shown in Fig. 1 in the range from 10 keV to 1 MeV. The corresponding values for the Ge are included for comparison. CdTe possesses a photoelectric absorption coefficient a factor of 1.6 to 4.8 higher than that of the Ge, for energies in the range beyond the Cd K-edge (26.7 keV) to 1 MeV. Secondly, the density of CdTe is favourable: 6.5 g/cm^3 as opposed to 5.5 g/cm^3 for Ge. The favourable average atomic number and density of CdTe make it suitable for small volume detectors: in principle, 2 mm thickness of CdTe are equivalent to 11 mm of Ge at the gamma energy of 662 keV.

The large bandgap energy of CdTe (1.47 eV), in comparison to Ge at 0.64 eV, allows stable operation of the CdTe at room temperature and immediate use. This is an advantage over the use of Ge, which has to be cooled, if at room temperature, for several hours prior to being employed.

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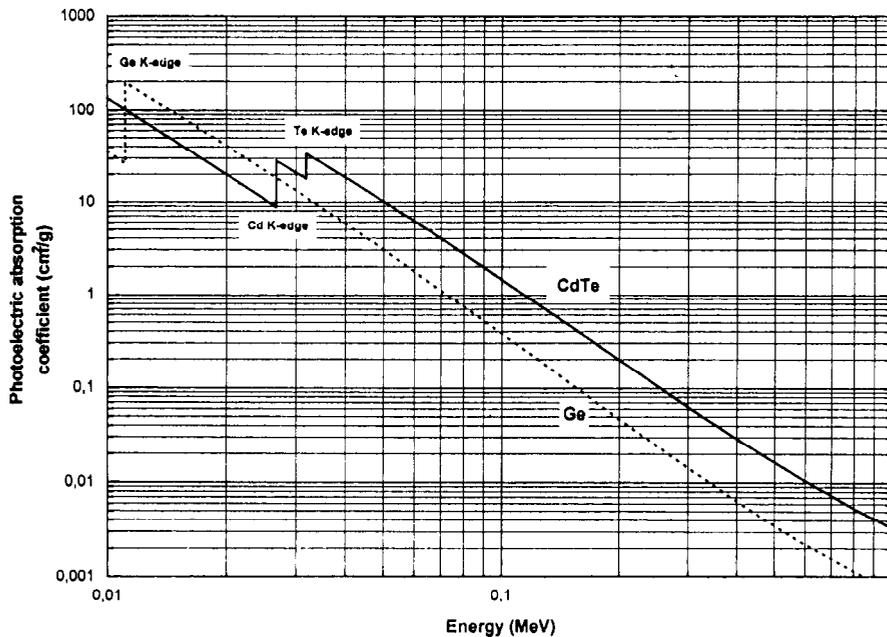
Fig. 1. The photoelectric absorption coefficient of Ge and CdTe as a function of energy.

The improvements in semiconductor technology over the past ten years have made it possible to construct CdTe crystals for gamma spectrometry with an adequate energy resolution. The growth of crystals of the order of a few mm thickness and several mm^2 sensitive area is feasible through several methods [3]. Although already suitable for gamma spectrometry, CdTe requires further improvement in its crystallographic qualities. Presently, it is difficult to reproduce crystals of the same characteristics. The operation of CdTe at low voltage (150 V) renders it safer than the Ge which operates at several thousand volts. Danger during handling and damage to the crystal in the case of power failure are minimised considerably. The presence of impurities and the low operating voltage result in energy resolutions inferior to that of Ge but considerably better than that of NaI scintillator.

3. Material and methods

3.1. Instrumentation

A planar CdTe crystal (2 mm depth, 4 mm^2 sensitive area) was used in this work (Fig. 2). The crystal and preamplification circuitry are incorporated inside a cylindrical probe housing made of aluminium alloy (9 mm diameter and 90 mm length). The crystal is operated at room temperature without the use of cooling. The detector probe was purchased from EURORAD [Strasbourg, France].

The nuclear electronics used comprised a standard NIM high voltage supply, a specially developed amplifier, an ultra high speed analog-to-digital converter (0.75 μs fixed conversion time for high resolution and count rate spectrometry) and a PC-based MCA. The spectrometry amplifier was manufactured by EURORAD especially for use with the CdTe crystal. This amplifier combines the normal amplification operation with an electronic pulse processing unit (charge loss corrector CLC) resulting in improved energy resolution, peak-to-Compton and peak-to-valley ratios. The improvement is achieved through the compensation of the charges lost due to the recombining or trapping of the holes [4]. It should be noted that the use of the CLC improves the resolution at the expense of efficiency. Nevertheless, the CLC is an essential unit in high count-rate gamma spectrometry.

The pulses from the preamplifier were fed to the CLC unit through a cable of 10 m in length, resembling the
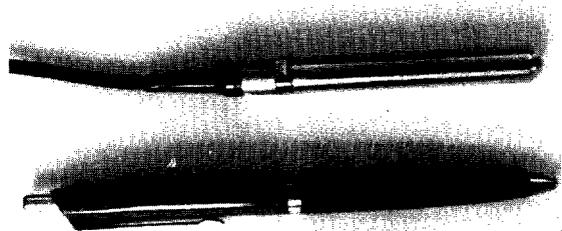

Fig. 2. The miniature CdTe detector used in this work.

remote operating conditions for the study of spent fuel. Great care was required in the optimisation of the gamma spectra obtained using the CLC amplifier: not only gain adjustments but also adjustments on the charges loss correction cause an energy shift in the spectrum. Optimisation, on the basis of the energy resolution, was performed on a particular gamma energy line and is strictly valid only over a small energy range around this line. The optimisation for the present study was made on the basis of the 662 keV (^{137}Cs) line, for the range 500 to 800 keV where the fission products of interest emit strong gamma lines.

3.2. Experimental

Standard radioactive sources (^{137}Cs, ^{152}Eu, and ^{133}Ba) were used for calibration purposes. Gamma spectrometry was also performed on UO$_2$ spent fuel pin and pellets with higher burnup (>35 GW d/t) and short cooling time. The enrichment, burnup and cooling time of the spent fuel were known from the declaration of the operators. The studies on spent fuel were performed in the hot cell facilities where the fuel was stored. Two experimental arrangements were used.

In the first, spent fuel pins stored inside a β–γ hot cell were used. The cell wall, made of concrete, had a thickness of 1 m. A collimator, made of lead and tungsten, was incorporated into the wall allowing gamma spectrometry of pins with a detector situated outside the cell. These measurements were therefore performed in a low background environment. Rectangular apertures 10 mm in height and between 0.6 to 1.2 mm wide were available. Gamma spectrometry was also carried out using a HPGe detector for comparison.

In the second arrangement, the spent fuel pellets were stored inside an α–β–γ hot cell. The CdTe detector, specially packed to avoid contamination, was brought into the cell where it was exposed to a high radiation dose during use. Studies of a whole pellet were performed. Under this geometry, the detector to pellet distance was 50 cm.

4. Results and discussion

Standard sources of ^{137}Cs (3.5 MBq), ^{133}Ba (3.5 MBq) and ^{152}Eu (0.4 MBq) were used to obtain the characteristics of the detector and optimise the setup of the electronics.

The gamma spectrum of the ^{137}Cs standard source obtained with the CdTe crystal in the setup described above is shown in Fig. 3 (spectrum a). An energy resolution (FWHM) of 7.2 keV at 662 keV is obtained. The peak-to-Compton (P/C) and peak-to-valley (P/V) ratios are respectively 3 and 18. The same spectrum, taken without the CLC, is included in Fig. 3 for comparison purposes (spectrum b). The energy resolution is 10 keV and the P/C and P/V ratios 0.7 and 3 respectively. Both spectra a and b were collected for the same counting time. The improvement in the quality of the spectrum where the CLC is used is evident in Fig. 3. However, the improvement in energy resolution is accompanied by a 70% reduction in photopeak area (662 keV). Similar observations have been previously obtained using CdTe detectors, however with cooling of the crystal [5–7].

The gamma spectrum obtained simultaneously from the ^{137}Cs and ^{133}Ba standard sources, with use of CLC, is shown in Fig. 4. The gamma lines of ^{133}Ba are well
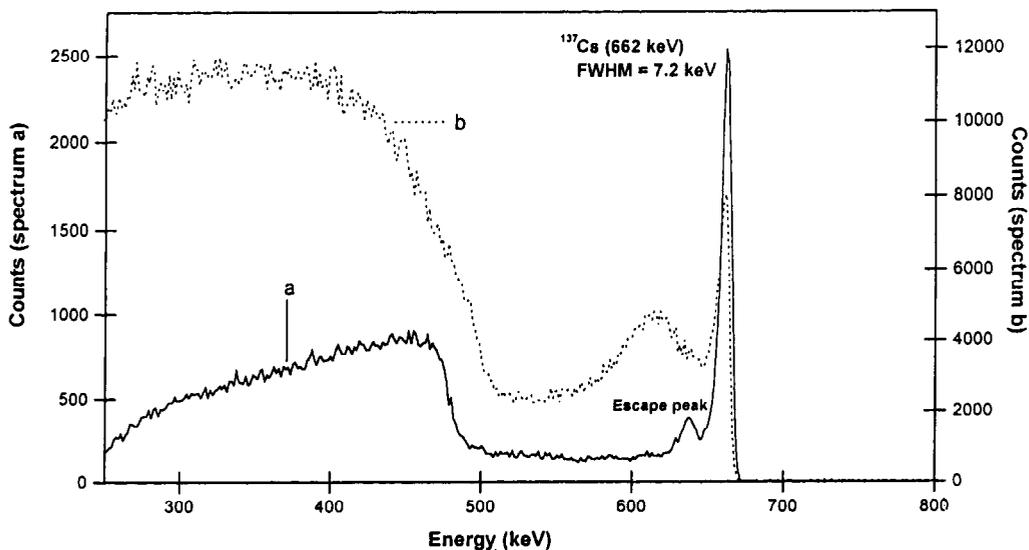

Fig. 3. Gamma spectrum of the ^{137}Cs standard source obtained with (spectrum a) and without (spectrum b) charge loss correction.
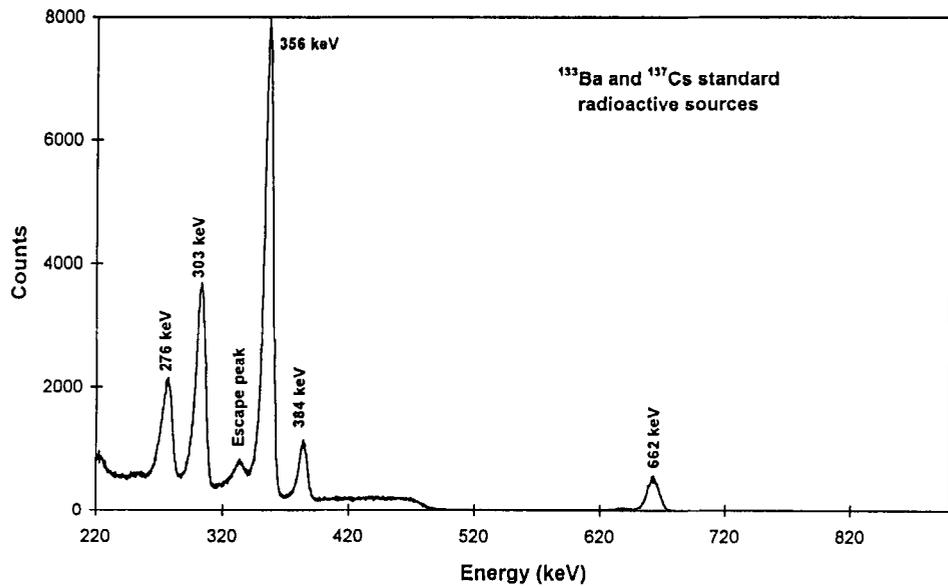
Fig. 4. Gamma spectrum of both ^{133}Ba and ^{137}Cs standard sources.

resolved and visible on the Compton continuum corresponding to the 662 keV line. Fig. 5 shows the gamma spectrum of the ^{152}Eu standard source. Gamma lines corresponding to energies up to 1 MeV are visible.

The CdTe detector was used to detect the gamma radiation emitted by a spent fuel pin in the arrangement described under Section 3.2. The resulting gamma spectrum, in the energy region 450 keV to 1 MeV, is shown in Fig. 6 (spectrum b). The corresponding spectrum with the HPGe detector is included for comparison purposes (spectrum a). The fission products ^{134}Cs and ^{137}Cs and ^{106}Ru

are visible in the spectrum obtained with the CdTe detector. The 605 and 662 keV lines of ^{134}Cs and ^{137}Cs are well resolved and free from interferences. The 796 and 802 keV lines of ^{134}Cs are not resolved, which is also the case between the 512 keV of ^{106}Ru and the 511 keV annihilation radiation. The energy lines at 605 and 662 keV are thus the only ones suitable for interpretation purposes on spent fuel.

The gamma spectrometry on spent fuel pellets was carried out to examine the performance of the CdTe detector in a high background. Integrated count rates, over
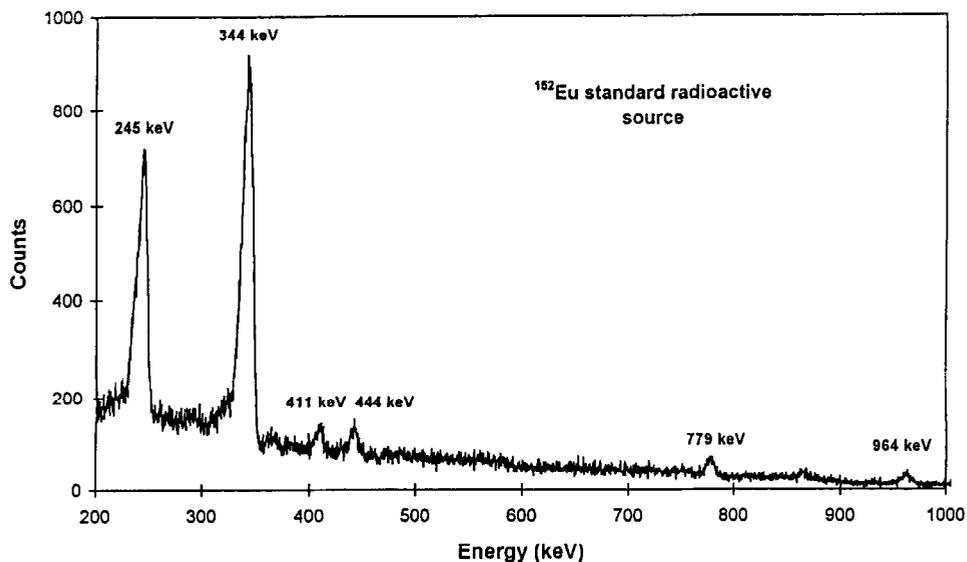

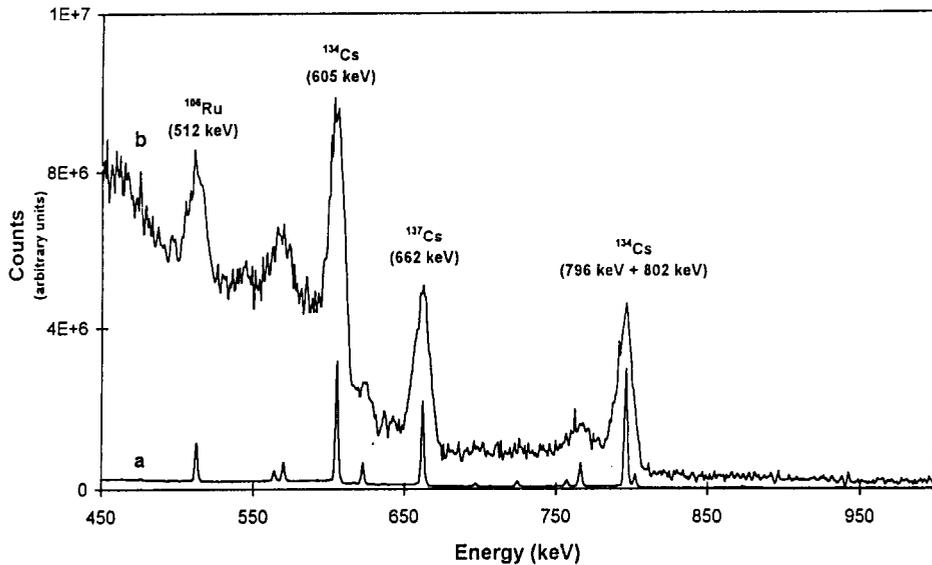Fig. 5. Gamma spectrum of the ^{152}Eu standard source.

Fig. 6. Gamma spectra of spent nuclear fuel pin obtained with the HPGe detector (spectrum a) and with the CdTe detector (spectrum b).

the spectra, of the order of 2000 counts/s were achieved with little influence on the dead time. The gamma spectrum of a spent fuel pellet obtained with few minutes counting is shown in Fig. 7 (spectrum a). This shows a similar profile to spectrum b in Fig. 6. The 605 and 662 keV lines of ^{134}Cs and ^{137}Cs are again the only lines well resolved. The high background and scattering in this hot cell results in a 50% deterioration of the energy resolution at 662 keV. The effect on the gamma spectrum when the CLC function of the amplifier is not utilised is demonstrated in Fig. 7 (spectrum b). There is a loss of resolution and signal-to-noise ratio, and it is evident that gamma spectrometry with CdTe requires the CLC function. A HPGe detector used under the same experimental conditions was completely saturated due to the high gamma radiation field.

The long term scope of this work is to demonstrate the use of the CdTe crystal as a gamma detector in the nuclear fuel cycle and apply it for the verification of the declared burnup and cooling time of spent fuels. The isotopic ratio ^{134}Cs/^{137}Cs was used for this purpose (605 keV for the fission product ^{134}Cs and 662 keV for ^{137}Cs). The results obtained with the CdTe detector were then checked through comparisons with other independent measurements.

In this context, this ratio as obtained for a spent fuel pin using the CdTe detector was within 10% of that obtained with the HPGe detector (Fig. 6). The spectra from the pellets using the CdTe detector were used to verify the declared burnup and cooling time of the pellets through isotopic correlations [8]. Correlations suitable for interpretation of the gamma spectrometry information were gener-
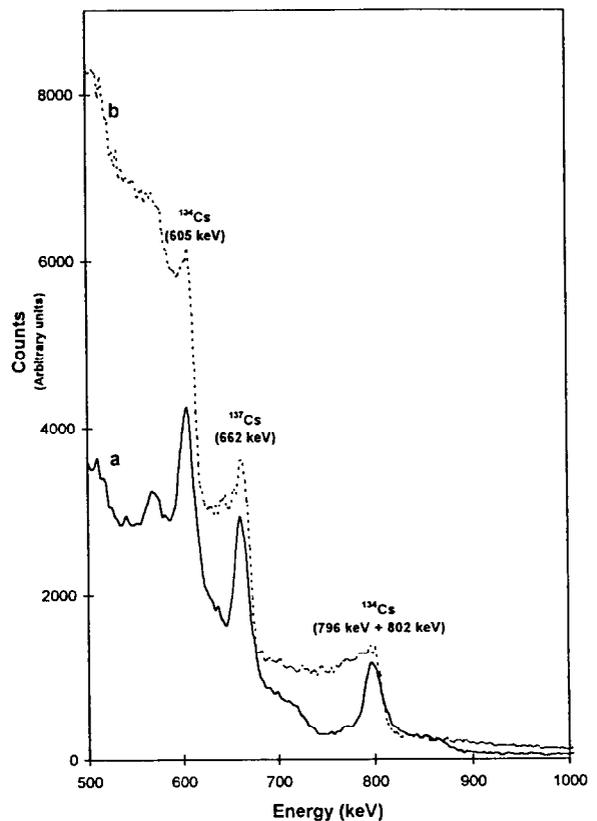

Fig. 7. Gamma spectra of nuclear spent fuel pellet with (spectrum a) and without (spectrum b) charge loss correction.

ated using the point depletion code KORIGEN [9]. The predicted parameters were within 10% of those declared by the operator. The uncertainties in the nuclear data in KORIGEN is a major contributor to the discrepancy in the predictions by the CdTe detector.

5. Conclusions

This work has shown that a CdTe gamma detector, complemented by a charge loss corrector amplifier, can yield spectra of adequate resolution for verification of burnup. Whether using standard gamma sources or spent fuel, a resolution of 7.2 keV at 662 keV is obtained. However, the use of the correction function of the amplifier is essential in order for valid spectra to be obtained at all. The ratio $^{134}Cs/^{137}Cs$, based on the 605 and 662 keV gamma lines, was used to verify the declared burnup of the spent fuel studied by means of calculated isotopic correlations. The burnup was verified within 10% of the declared one. The capability to operate this detector without cooling and its small size, render it suitable for remote operation within a compact and transportable device. In this respect, the CdTe detector can be incorporated within a compact measurement device to complement passive neutron interrogation of spent fuel inside a hot cell. Although not replacing the HPGe, the CdTe can examine spent fuel under conditions where the HPGe would be bulky and prone to saturation.

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