ELSEVIER

Nuclear Instruments and Methods in Physics Research A 405 (1998) 153–158

NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A

Use of miniature CdZnTe X/γ detector in nuclear safeguards: characterisation of spent nuclear fuel and uranium enrichment determination

K. Abbas[a], J. Morel[b], M. Etcheverry[b], G. Nicolaou[a,*]

[a] *European Commission, Joint Research Centre Institute for Transuranium Elements Postfach 2340, D-76125 Karlsruhe, Germany*
[b] *Laboratoire Primaire des Rayonnements Ionisants, CE-Saclay B.P. 52, F-91193 Gif-sur-Yvette Cedex, France*

Received 15 August 1997; received in revised form 1 October 1997

Abstract

The feasibility to perform X/γ-ray spectroscopy, on spent nuclear fuel and uranium solution and pellets, using a Peltier-cooled CdZnTe crystal (9 mm^2 sensitive area, 2 mm thickness) coupled with a dedicated preamplifier and amplifier has been studied. In the case of spent fuel, an energy resolution of 5.6 keV at 662 keV is achieved and characterisation of the fuel in terms of burnup and cooling time performed. The good energy resolution at low energies (1.9 keV at 122 keV) has allowed application of the URADOS code which, on the basis of the uranium XK_γ-rays, determined the ^{235}U enrichment in the uranium solutions and pellets within 10% of that declared.

Keywords: X/γ spectroscopy; Nuclear material; CdZnTe detectors

1. Introduction

X/γ-ray spectroscopy is widely used for nuclear safeguards purposes. High-resolution X/γ spectrometry based on germanium detectors has been routinely applied. However, the need to cool such detectors results in bulky devices not easily transportable and their use becomes problematic under space restriction. In the monitoring and assaying of nuclear material there is an increasing call for compact and portable devices for unattended and remote operation. In this context, the high stopping power and ability to operate at room temperature or with a miniature thermoelectric cooler renders CdTe/CdZnTe crystals suitable for X/γ spectroscopy for safeguards purposes [1].

This paper reports the results obtained using a commercial CdZnTe detector in the spent nuclear fuel characterisation and in uranium-enrichment determination.

2. Experimental aspect

The detector used in this work is based on a $Cd_{0.9}Zn_{0.1}Te$ crystal of 9 mm^2 sensitive area and

* Corresponding author. Tel.: + 49 7247 951490; fax: + 49 7247 951595.

0168-9002/98/$19.00
PII S0168-9002(97)01143-1

2 mm thickness (AMPTEK, USA). It is cooled by a thermoelectric circuit at a temperature of $-30°C$ and has an entry window of 10 μm thickness of beryllium. The detector is associated with an electronic module comprising a power supply, amplification and pulse shaping (constant of triangular shape of 3 μs) with rise-time discrimination resulting on the more symmetric peak with improved energy resolution and peak-to-noise ratio. The high voltage applied to the crystal is 400 V. Standard radioactive sources were used for calibration purposes and energy resolutions of 1.1 keV at 60 keV (^{241}Am), 1.9 keV at 122 keV (^{57}Co) and 5.6 keV at 662 keV (^{137}Cs) were obtained.

3. Spent nuclear fuel characterisation

Verification of the burnup, cooling time and the fissile content of spent fuel is an important component in nuclear safety. In this context, HPGe γ spectroscopy has been an established non-destructive assay technique. One goal of the present work is to discuss the performance of a CdZnTe detector for γ spectroscopy on spent fuel for energies greater than 400 keV. This range corresponds to that of γ-radiation emitted by most of the fission products present in the spent fuel.

The studies were performed on single spent fuel rods stored inside a β–γ hot cell at the Institute for Transuranium Elements in Karlsruhe. 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 γ spectrometry of rods with a detector situated outside the cell. Rectangular apertures 10 mm in height and between 0.6 and 1.2 mm wide were available. The resulting γ spectrum of spent fuel, in the energy region 450 keV–1 MeV, is shown in Fig. 1. The fission products ^{134}Cs and ^{137}Cs and ^{106}Ru are visible in the γ spectrum. The 605 and 662 keV lines of ^{134}Cs and ^{137}Cs are well resolved. The 796 and 802 keV lines of ^{134}Cs are not resolved, which is also the case for the 512 keV of ^{106}Ru and the 511 keV annihilation radiation. The energy lines of 605 and 662 keV are thus the only ones suitable for interpretation purposes on spent fuel. The isotopic ratio ^{134}Cs/^{137}Cs (based on the 605 and 662 keV γ lines)

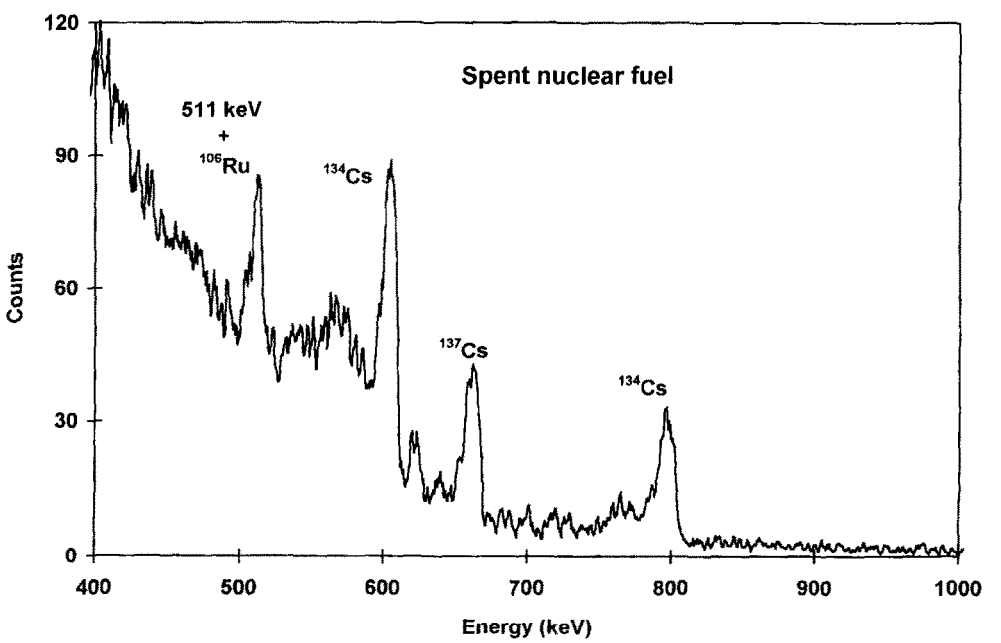

Fig. 1. γ spectrum of spent nuclear fuel obtained with the CdZnTe detector.

complemented by isotopic correlation could be used for the verification of the declared burnup and cooling time to within 10% and 3%, respectively, of the declared values.

4. Uranium-enrichment measurement

In the past few years, several methods have been developed for the determination of the uranium enrichment through X/γ spectrometry [2–7]. Most of them consist in the exploitation of some well-resolved peaks with a scintillation detector. Such systems are usually collimated and, as a rule, it is necessary to make a previous calibration of the pair detector and collimator with known references. More recent processes were aimed at removing the need for this calibration [8,9]; they are based on the analysis of the uranium spectra in the XK-rays region measured by germanium detectors. However, the use of such detectors has some drawbacks as far as field measurements are concerned because of the constraints linked to their cooling with liquid nitrogen or with a compression system. Consequently, tests were undertaken to see if it was possible to determine the enrichment of uranium samples with an uncertainty of about 10% using a CdZnTe detector.

The measured samples were either nitric acid solutions of known enrichment, or certified pellets of uranium oxide. Typical spectra obtained are shown in Figs. 2–4. In general, the measured samples were placed 5 mm away from the entry window of the detector and the counting lasted several hours (from 2 to 20 h according to the sample). An energy resolution of 2.4 keV at 186 keV (^{235}U) was obtained.

One way to avoid the constraints linked to a calibration with external references is to reduce the region of interest to be studied so that the variation of the efficiency of the detector would be insignificant. This is possible when considering the XK_γ-rays region where numerous peaks of different shape and width are present such as γ-ray peaks (Gaussian distribution) and X-ray peaks (Voigt function).

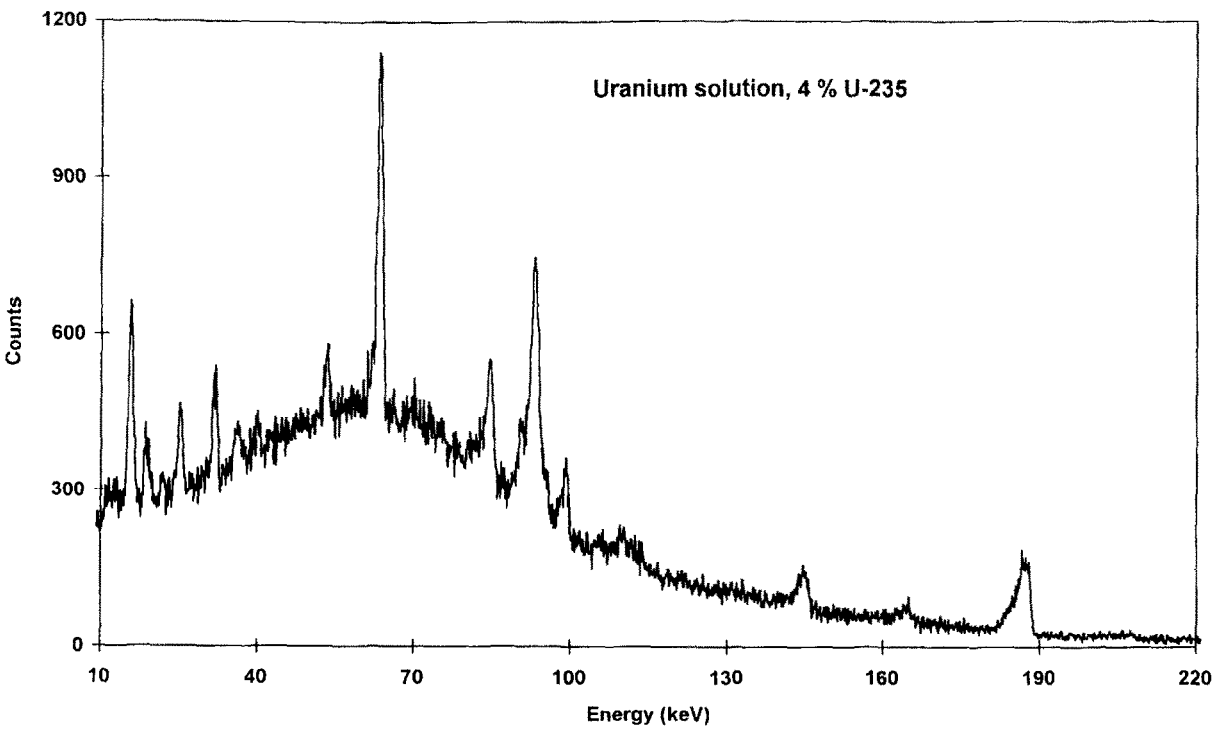

Fig. 2. γ spectrum of nitric acid solution of uranium of 4% enrichment obtained with the CdZnTe detector.

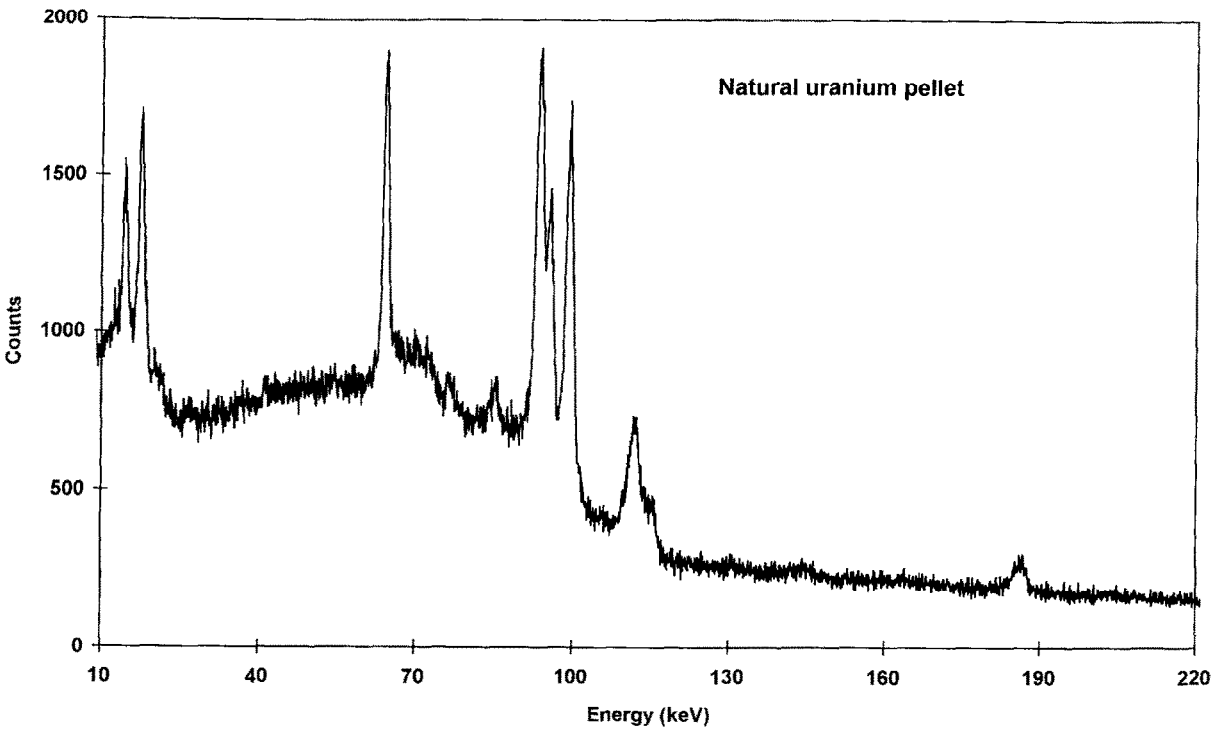

Fig. 3. γ spectrum of fuel pellet of natural uranium obtained with the CdZnTe detector.

The treatment of this region of interest is delicate and requires to take into account the following three contributions:

1. The first corresponding to the presence of ^{235}U and its daughters: Rays of Th-$XK_{\alpha 2}$ of 90.0 keV, Pa-$XK_{\alpha 2}$ of 92.3 keV, Th-$XK_{\alpha 1}$ of 93.3 keV, Pa-$XK_{\alpha 1}$ of 95.9 keV, and γ-rays of 89.9 and 99.3 keV.
2. The second corresponding to the presence of ^{238}U and its daughters: Rays of Pa-$XK_{\alpha 2}$ of 92.3 keV, Pa-$XK_{\alpha 1}$ of 95.9 keV, and γ-rays of 92.4 and 92.8 keV.
3. The third corresponding to the spectrum of self-induced X-ray fluorescence: Rays of U-$XK_{\alpha 2}$ of 94.6 keV and U-$XK_{\alpha 1}$ of 98.4 keV.

With the "URADOS" code [9] used in the present work, the contribution of these components is estimated according to the principle of the least-squares method. Beforehand, several isolated peaks or peaks considered as such, are exploited to establish responses in energy and width of peaks. These responses are then refined in order to refine the adjustment.

The results obtained are presented in Table 1 with an uncertainty value for a level of confidence of 68%. The calculated enrichments are within 10% of those declared, except for the natural uranium. This limitation for natural uranium is due to the low ^{235}U component, in relation to those of ^{238}U and uranium self-induced X-ray fluorescence, in the analysed spectrum. A similar difficulty is anticipated for high ^{235}U enrichments (> 90%) which would be due to the ^{238}U component being lower than those of ^{235}U and uranium self-induced X-ray fluorescence in the spectrum.

5. Conclusions

X/γ-ray spectroscopy on spent nuclear fuel and uranium solution and pellets was carried out using

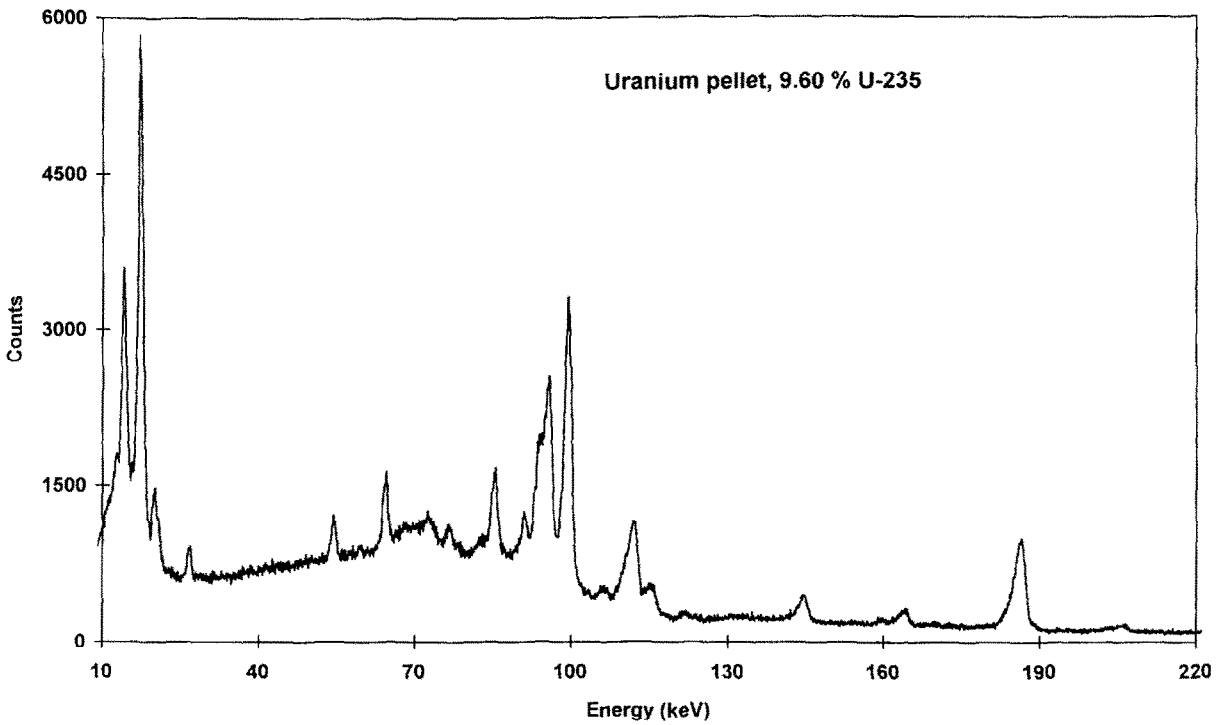

Fig. 4. γ spectrum of fuel pellet of uranium of 9.60% enrichment obtained with the CdZnTe detector.

Table 1
Uranium enrichment (% ^{235}U) measurement using X/γ spectra of CdZnTe detector and calculation code URADOS

Type of sample	Counting time (s)	Declared enrichment	Measured enrichment
UO_2 pellet	50000	0.711	0.84 ± 0.17
U solution	14000	1.65	1.72 ± 0.47
UO_2 pellet	50000	2.78	2.50 ± 0.30
U solution	7800	4.00	4.18 ± 0.58
U solution	70000	5.08	4.99 ± 0.50
UO_2 pellet	50000	9.55	9.30 ± 0.70

a Peltier-cooled CdZnTe miniature crystal. In the case of spent fuel, the achieved energy resolution of 5.6 keV at 662 keV enabled burnup and cooling time verifications to within 10% and 3%, respectively, of declarations. The good energy resolution at low energies (1.9 keV at 122 keV) has allowed application of the URADOS code, on the basis of the uranium XK_γ-rays. Using the improved resolution, the ^{235}U enrichment in the uranium solutions and pellets analysed has been determined within 10% of that declared.

References

[1] K. Abbas, G. Nicolaou, D. Pellottiero, P. Schwalbach, L. Koch, Nucl. Instr. and Meth. A 376 (1996) 248.
[2] T.D. Reily, R.B. Walton, J.L. Parker, Report LA-4605-MS, Los Alamos National Laboratory, 1970.

[3] L.A. Kull, R.O. Ginaven, G.E. Glancy, Atom. Energy Rev. 144 (1976) 681.
[4] J. Morel, H. Goenvec, J. Dalmazzone, G. Malet, Proc. 1st Ann. Symp. on Safeguards and Nuclear Material Management, Brussels, ESARDA-10, 1979, p. 356.
[5] P. Matussek, Report KfK 3752, Kernforchungszentrum Karlsruhe, 1985.
[6] T. Dragnev, J. Safar, F. Grinevich, V. Ruklo, Report IAEA STR 263, 1990.
[7] W. Nagel, F. Quik, EUR 14659 EN, CEC-CBNM-Geel, 1993.
[8] R. Gunning, W. D. Ruther, P. Miller, J. Goerten, M. Swinhoe, H. Wagner, J. Verplancke, M. Bickel, S. Abousahl, MGAU: A New Analysis Code for Measuring U-235 Enrichment in Arbitrary Samples, UCRL-JC-114713, 1994.
[9] J. Morel, G. Riazuelo, Uranium Enrichment Measurement by X- and γ-ray Spectrometry with the URADOS Process, Note technique LPRI/96/011, CEA, 1996.