

## SAFEGUARDS VERIFICATION OF SHORT COOLING TIME KANUPP IRRADIATED FUEL BUNDLES USING ROOM TEMPERATURE SEMICONDUCTOR DETECTORS

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### Abstract

*The IAEA has over a decade used non-cryogenic miniature cadmium telluride (CdTe) detectors for the verification of CANDU spent fuel bundles stored under water on stacked trays. On account of very high gamma ray count rates, the method in spite of employing heavy lead / tungsten shield and narrow collimator besides advanced pulse processing electronics, was limited to fuel bundles with a cooling time of over one year. Recent developments in semi conductor materials evolving into cadmium zinc telluride (CdZnTe) detector reflecting in its enhanced energy resolution and yield has allowed exploitation of intermediate half-life fission products and selective counting in the energy band of interest. Moreover, less sensitive silicon solid state detectors have also in the mean time become available. Together, these new and improved detectors were studied at KANUPP (137 MWe CANDU PHWR) to have the necessary potential for safeguards verification in the cooling time range that had remained elusive until recently.*

*The test verification measurements were carried out employing the CANDU Bundle Verifier for Stacks (CBVS) and differential counting in pre-selected ROIs around the characteristic Zr-95/Nb-95 and Cs-137 gamma ray lines. The measurement results indicated that super grade CdZnTe detectors are very promising as they dramatically reduce the verification threshold to barely ~ 1 month. Further development in room temperature semi conductor detectors, taking place at a fast pace together with the use of depleted uranium shield / collimators might allow the entire spent fuel storage pond inventory to become verifiable irrespective of the cooling time constraint.*

### 1. INTRODUCTION

This paper is based on an extended collaborative work involving a NPP operator (KANUPP), a detector manufacturer (Ritec) and the implementer of international safeguards (IAEA) to test, evaluate, and to stimulate the development of room temperature semi-conductor detectors for the verification of short cooling time (< 1 year) KANUPP irradiated fuel bundles (19 element type) stored on stacked trays in close packed arrangement. Besides CdZnTe (CZT), Silicon semiconductor detectors were also investigated for this purpose.

The silicon detector appeared attractive in view of its very low sensitivity and moderate response to high intensity gamma ray fields typical of short cooling time fuel bundles. Owing to its low Z-number ( $Z = 14$  compared with 48-52 of CdTe), it has insignificant photoelectric cross section. Its pulse height response comprises therefore of just the Compton continuum, devoid of any photo peaks. This necessitated integral rather than differential or selective counting in an energy band of interest. The fact that the silicon detector is less sensitive and can tolerate gamma dose rates in the ultra high range also helps laying lesser demand on pulse processing spectroscopy amplifiers. Such requirements as base line restoration (BLR) and pole zero (p/z) cancellation, necessary for high count rate work assume all of a sudden lesser importance.

In spite of the advantages described in the fore, the silicon detectors cannot however provide a spent fuel specific attribute test. This discrepancy is overcome in CZT detectors which are now available with much improved resolution (1-3 % for Cs-<sup>137</sup>). Compared to NaI detectors their resolution is ~ 3 to 10 times better depending on factors such as detector type, size, and energy. However compared to HPGe detectors the resolution of CZT's is about 2-4 time lower [1]. Since they do not require liquid nitrogen cooling, these are particularly suited for remote monitoring applications. The CZT detectors have now been developed to an extent that coupled with much improved pulse processing electronics they can provide adequate fission product signatures even at short cooling times. Compared to silicon the CdTe based detectors have good intrinsic efficiency on account of its high density and high mean value atomic number (Z). This characteristic is important for spent fuel verification where small well shielded detectors with good spectral performance are required to obtain spectra in the presence of a significant high energy scattered background. The CZT is forerunner of CdTe detector which had until recently been used for the verification of moderate to long cooling time fuel bundles. It is superior on account of its higher resistivity and wider band gap (1.6 eV for CZT compared to 1.47 eV for CdTe).

In this work hemispheric miniature detection probes (SDP310Z S) with integrated preamplifier made by Ritec Ltd. (Riga, Latvia) were used [2]. These, make use of CdZnTe raw material supplied by eV Products (USA). The cooperative effort between the material developers and the detector fabricators has led to considerably improved reliability and performance. The detector/preamplifier assembly has small dimensions (diameter 8.2 mm, length 90 mm) and weight, high-energy resolution, room temperature operation, small consumption of power and high counts rate capability. It may be mentioned that small size of the detector was not the only requirement from the view point of particular application (in-situ spent fuel verification) but also to minimize the characteristic large amount of charge trapping and transport problems intrinsic to the material poor hole transport, contributing heavily to the low energy tailing problem. This further led to near gaussian shape of the photo peak and much improved resolution between lines of closely lying gamma ray energies. In accordance this detector can now approach the performance criteria that were previously the exclusive domain of the liquid nitrogen cooled HpGe detectors. With the help of automatic scanning mechanism and appropriate data processing software it has moreover allowed much improved verification speed.

The reported assessment work was performed at the KANUPP spent fuel storage pool where the fuel bundles are stored on stacked trays (18 trays / stack and 11 bundles / tray). The bundles (diameter: 81 mm) are separated by a water gap of 20 mm in the vertical direction, in which they are scanned. The verification method utilized is specific to CANDU bundles stored on stacked trays in a dense geometry under water. A well-collimated and shielded detector is moved vertically along the end-faces of the bundles. For verification based on the use of CZT detectors Cs-137 peak area is recorded as a function of the scanning coordinate. This method works quite well when cooling time of the bundles is longer than one year [3]. With short cooling time (< 1 yr.) and the associated intense gamma field, the Cs-137 peak disappears in the heavy Compton background. It has however been studied that the gamma rays of short-lived isotopes themselves can be used for the verification purposes in this time domain. This could have been difficult, but recent developments in the field of CZT detectors, as mentioned above have made it possible to exploit the fission product gamma lines characteristic of such assemblies. In this paper the use of the gamma lines of Nb/Zr-95 for the verification of CANDU bundles with cooling time of less then one year is also investigated. They have an energy of 724 and 765 keV respectively, which is only a little higher than the 662 keV gamma ray of Cs-137.

## 2. MEASUREMENT EQUIPMENT

### 2.1 Shield/Collimator Assembly

Shield	Tungsten (surrounding the detector ) and upper shield of Lead
Dimensions	90 mm × 145 mm
Collimator	Tungsten Insert Tube (5 mm dia)

The above specified shield collimator assembly is described in an accompanying paper being presented at this symposium [4]. It was designed and fabricated at Seibersdorf laboratories of IAEA.

### 2.2 CANDU Bundle Verifier for Stacks (CBVS)

The CANDU Bundle Verifier for Stacks (CBVS) is standard verification equipment used by the IAEA inspectorate. The equipment was developed in collaboration with Seibersdorf workshop of the IAEA at KANUPP under a separate research contract and is described elsewhere. For use at KANUPP, it is mounted on the bridge of spent fuel storage bay. It comprises the support structure, the electronic controller and the shield-collimator assembly. Making use of a stepping motor the shield-collimator assembly housing the detector is moved with a constant speed in front of tray stacks. If the collimator hole points towards a bundle, the gamma signal is high, if it faces the gap between two bundles, the signal drops to a lower value. Besides scanning in the vertical direction, the CBVS also provides lateral movement for adjusting the position of collimator. The scanning speeds from 5mm/s to 0.5 mm/s are available.

### 2.3 Pulse Processing and Data Acquisition Equipment

Amplifier	TENNELEC (TC-244)
MMCA	MCA-166
Computer Hardware	HP 100LX Palmtop /IBM Thinkpad Notebook Computer
Computer Software	WinMCA (Ver 0012) with SCANDU option or WinSCAN

The output of preamplifier was connected to a Tennelec TC 244 spectroscopy amplifier and on to the MMCA (Mini MCA, model MCA 166) developed at Forschungszentrum, Rossendorf for the International Atomic Energy Agency and the Euratom inspectorate. It is a battery powered (operates for 8 hours on re-chargeable Li-ion battery) high performance 4 K MCA/MCS module, provided with the detector high voltage supply and an internal amplifier. For working with shorter than 1  $\mu$ s pulse shaping time using CZT detector, the internal amplifier could be readily by-passed. The MMCA is small (155  $\times$  9.5  $\times$  50 mm). Its shape and size having been optimized for use with a HP 100/200 palmtop. It could however, also be used with any other laptop or standard PC with a parallel printer port (RS-232 C). Since the lowest computer platform was the HP 100LX palmtop computer, the software (MCA) used was the one, which worked under MS-DOS 5.0 or higher. For Windows 95, the program WinMCA (Ver. 0012 with SCANDU option) was employed which had somewhat upgraded features compared to the software MCA. It was written in Visual Basic in 32-bit code and had WINSPEC (pulse height spectrum measurement) MCS (multi channel scaling) and SCANDU (verification of CANDU fuel bundles with simultaneous capability of pulse height analysis and multi channel scaling) operation modes.

### 3. TEST MEASUREMENTS

The reported upon test measurements, comprised (1) analyzing the pulse height response of the detector using both the single gamma energy as well as composite gamma ray energy source (2) determining the relationship between the age of spent fuel bundles and the integral count rate (for silicon detector) and (3) analyzing if the spent fuel bundles of cooling time < 1 year could be adequately detected through the modulation of multi channel scaling response in the SCANDU (Scan for CANDU Bundle Verification) mode.

#### 3.1 Tests With Silicon Semiconductor Detector

The specifications of the silicon semiconductor detector used for the reported investigations are given in Table-I

**TABLE-I**

**Specifications of Silicon Semiconductor Detector**

Manufacturer	EURORAD
Model	S08Wsid
Serial Number	01
Housing	Cylindrical shaped Diameter: 8 mm; length 91 mm Material: aluminum alloy
Detector Bias Voltage	12 V
Detector Sensitivity	3 cps/rad/hr
<b>Preamplifier:</b>	
Noise:	1.4 keV Si at 0 pf
Sensitivity:	31 mV / MeV Si
Output Polarity:	Positive
Pulse Rise Time:	20 ns
Feedback: 1 pf / 500	1 pf / 500 M $\Omega$
Power Supply:	+ 12 V (9 mA) -12 V (4 mA)

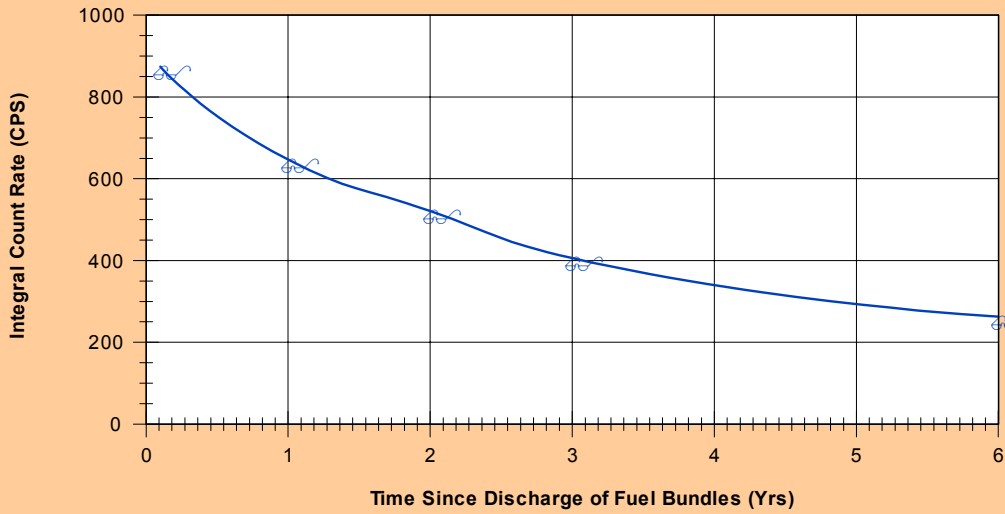
#### 3.11 Relationship Between Age of Spent Fuel Bundle and Integral C/R Response of Silicon Detector

The analysis of the detector pulse height response indicated that the entire information related to the irradiation and cooling history of a fuel bundle was contained in its integral Count rate distribution. The relationship between the cooling time and integral count rate is graphically represented in Fig. 1.

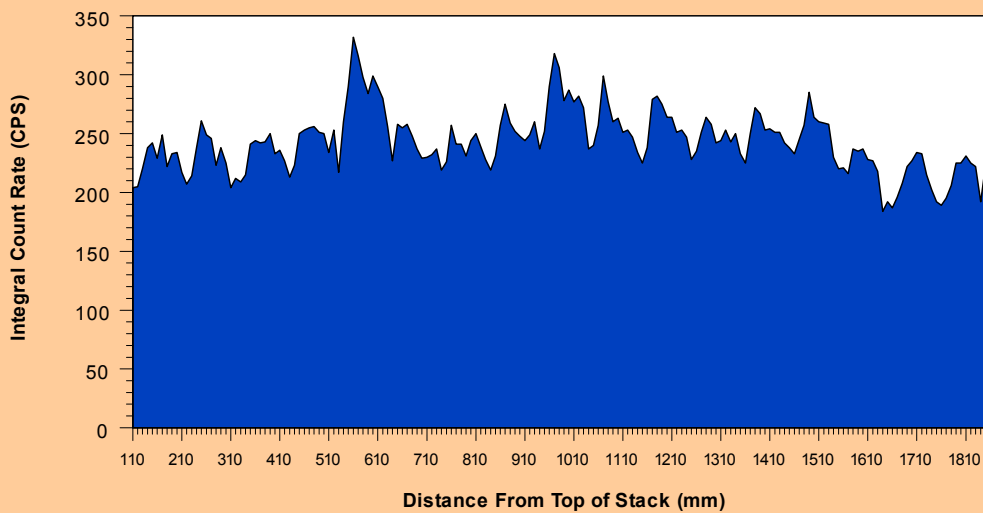
#### 3.12 Verification Measurements Using Silicon Detector

Using the integral count rate as the verification signal, the stacks of 6, 3, 2 and 1 year old as well as freshly discharged fuel bundles were scanned. The scans were carried out from top to bottom along the 6th bundle position. While the patterns for the longer cooling time bundles were well resolved, clearly identifying the presence of fuel bundles by the well pronounced and separated peaks, the short cooling time bundles of 1 year and less also gave rise to a modulation pattern but with poorer resolution. The scan involving long cooling time bundles (~ 6 years) is shown in Fig. 2

**FIG. 1 RESPONSE OF SILICON SEMICONDUCTOR DETECTOR TO IRRADIATED FUEL BUNDLES AS FUNCTION OF COOLING TIME**



**FIG. 2 SCANDU VERIFICATION PROFILE OF ~ SIX YEARS OLD BUNDLES USING SILICON SEMICONDUCTOR DETECTOR**



### 3.2 Tests With CdZnTe (CZT) Semiconductor Detector

Although only one detector was used for all the fuel characterization and verification measurements reported, several CZT detectors were utilized for the optimization of parameters. These were all of supergrade category with quoted resolution of 10 keV (FWHM) or better at 661.6 keV photo peak of Cs137. The specification of a typical detector of this type is presented in Table-II.

**TABLE-II****Specifications of CdZnTe Semiconductor Detector**

<b>Manufacturer</b>	Ritec Ltd., Riga, Latvia
<b>Model</b>	SDP 310/Z/20 S
<b>Housing</b>	Cylindrical shaped Diameter: 8 mm; length 91 mm Material: aluminum alloy Cable length: 200 mm
<b>Operating Parameters</b>	
(Measured at the 662 keV line of Cs-137 at the detector operating voltage and amplifier shaping time of 1 $\mu$ s)	
Detector Operating Voltage	+ 300 V
FWHM	7.3 keV
FWTM	20.1 keV
Peak-to-Compton Ratio	4.67
Peak-to-Valley Ratio	81.49
Recording Sensitivity	0.011 mm <sup>2</sup>
Output Signal Polarity	Negative

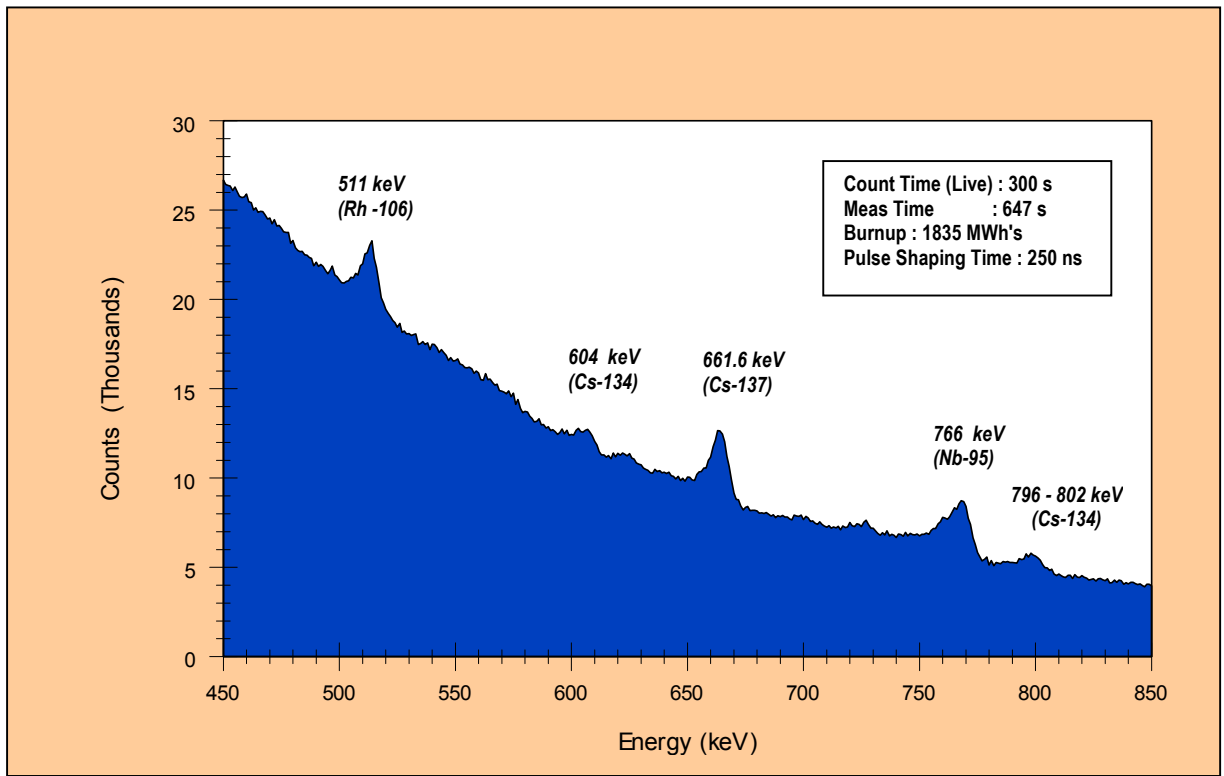
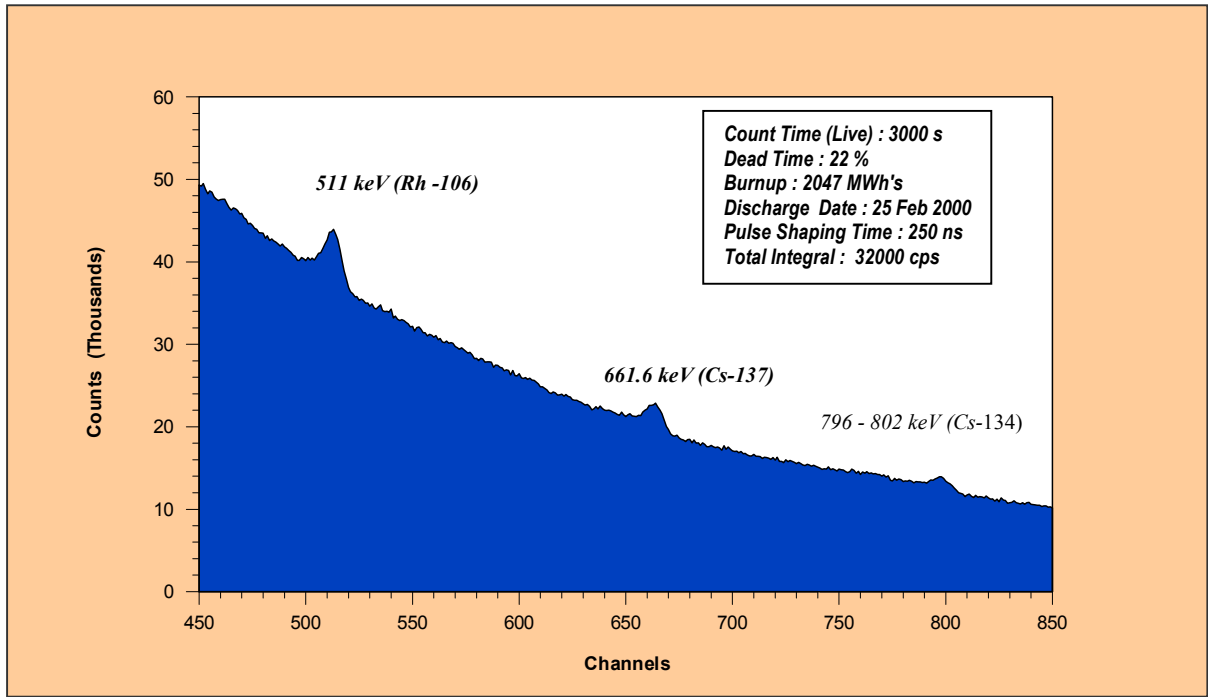
As mentioned besides the actual verification extensive optimization tests aimed at selection of operating voltage and pulse shaping time were carried out. Detector resolution (FWHM) and peak/Compton ratio, in general improve with the applied bias. The most optimum high voltage for the detectors utilized for the irradiated fuel verification was determined to be 300 V. With the increase of pulse shaping time, the amplifier PUR throughput decreases. The measurements performed on one-year-old fuel bundles indicated that the count rate decreases from 70,000 cps at 250 ns to about 54,000 cps (77%) at 750 ns. The functional dependence being linear in the entire range. The effect on detector resolution was also studied. Best resolution is not necessarily associated with the shortest (250 ns) pulse shaping time. Increasing the time constant results at first in improving the resolution from 12.5 keV (at 661.6 keV Cs-137 energy) to less than 11 keV at 350 ns. A further increase leads however to deterioration of the parameter sharply to 13.5 keV at 750 ns. Based on the measurements so conducted the pulse shaping time constant was optimized to be 350 ns for bundles of cooling time greater than a few months and 250 ns for shorter cooling time bundles.

**3.21 Response of CdZnTe (CZT) Detector to Short Cooling Time Fuel Bundles - ROI Optimization**

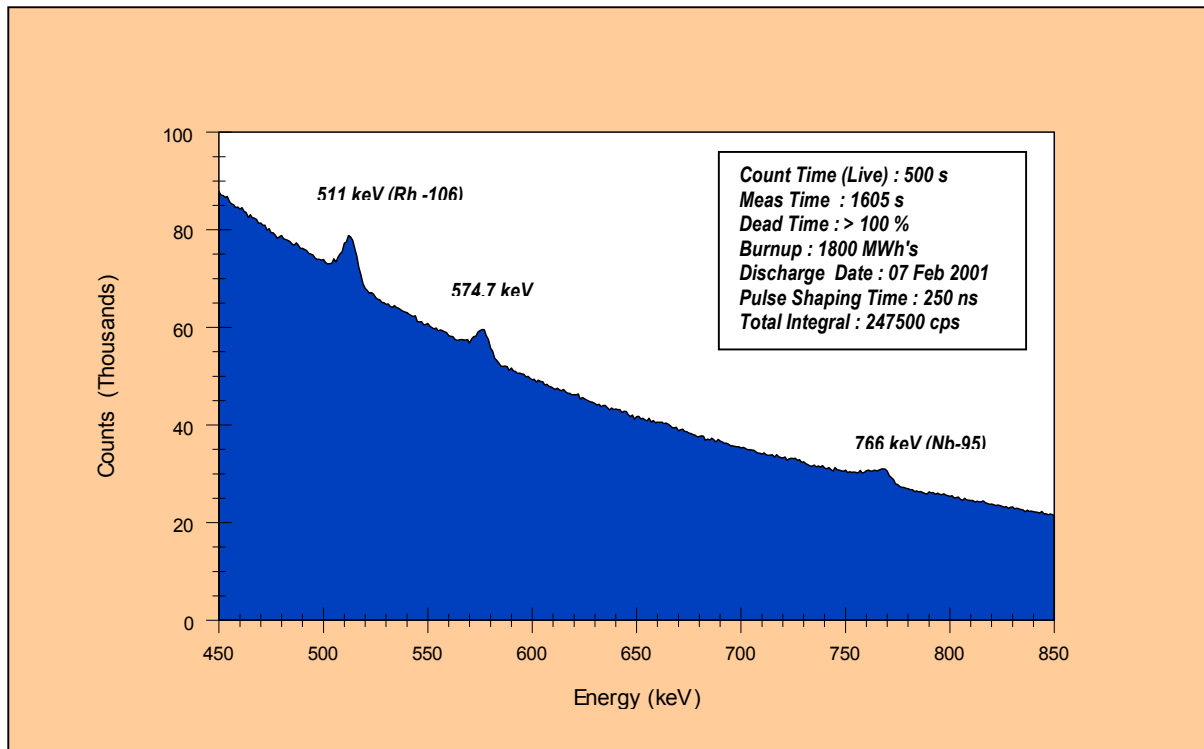
Although characterization measurements were carried out on bundles of long as well as short cooling times ranging from tens of years to barely a few days, only those measurements are presented here which pertain to the cooling time period in which fission product gamma ray spectra undergo maximum changes. This period relates to cooling time of one year and less. The representative spectra are therefore chosen to be those of one year, six months and one week old fuel bundles. For all the measurements discussed hereunder only one detector was used and all instrumentation settings were optimized. These included: (a) detector bias: +300 V (b) amplifier gain:  $\sim$  1keV/channel such that 661.6 keV Cs-137 peak could be positioned in channel 662 and (c) pulse shaping time: 250 ns (with PUR on).

The gamma ray spectrum of one-year-old fuel bundle is depicted in Fig. 3. At this point in time total integral counts are  $\sim$  30,000 cps. In the region of interest (450 – 850 keV) three gamma ray lines i.e., 511 keV (Rh-106), 661.6 keV (Cs-137) and 796-802 keV (Cs-134) stand out. Any of these can in principal be used for the purpose of verification but 661.6 keV is normally used for selective counting and gives good results.

With decreasing cooling times, changes in the spectrum start appearing and in addition to the above mentioned three prominent sources an additional source i.e., Zr95/Nb95 starts appearing. This could be seen in the spectrum taken at the cooling time of 6 months in Fig. 4. An additional photo peak at 765 keV is clearly observable. Both the 661.6 keV and 765 keV lines could be utilized for the verification purposes.

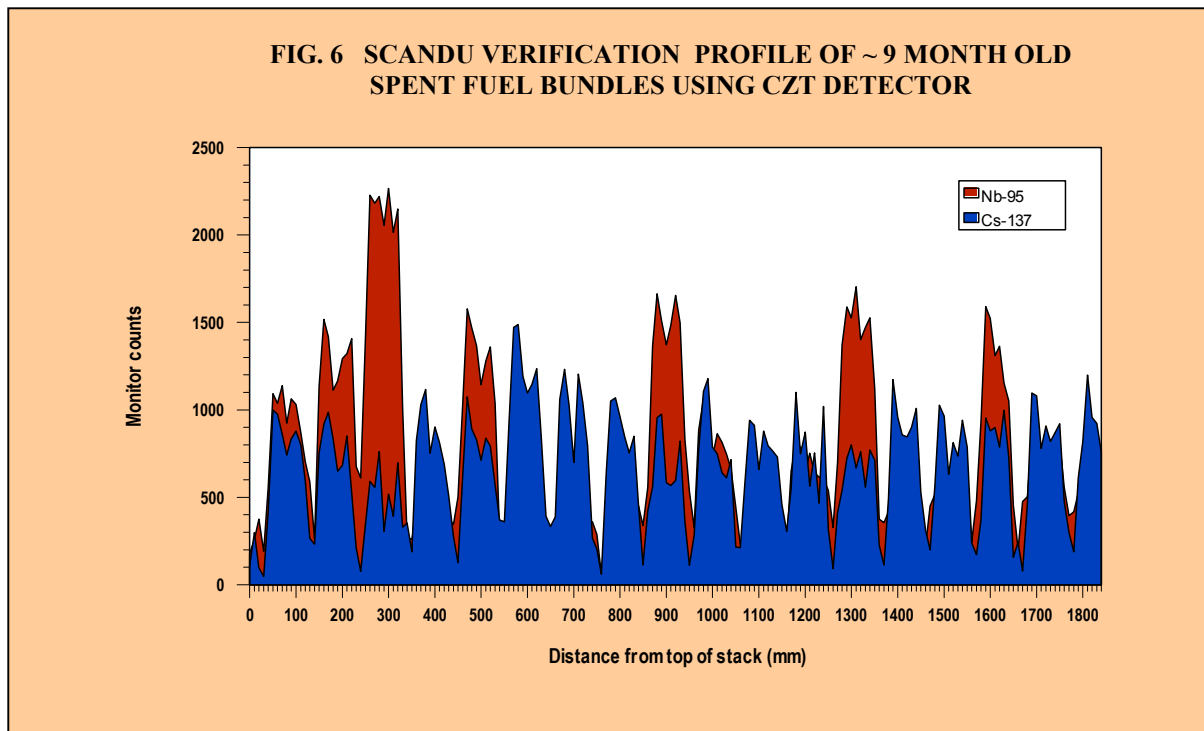


At cooling times of less than one month the count rate drastically increases. On account of high intensity of emitted radiation from short lived fission products, the signatures originating in Cs-134 and Cs-137 get lost and in only the 511 keV, 574.7 keV and 766 keV (Nb-95) remain visible. A spectrum taken at the cooling time of one week as shown in Fig. 6 clearly depicts this. The count rate at such cooling time is ~ 250,000 cps. The Nb-95 peak at 766 keV is used and results in quite adequate verification of bundles at such short cooling times.

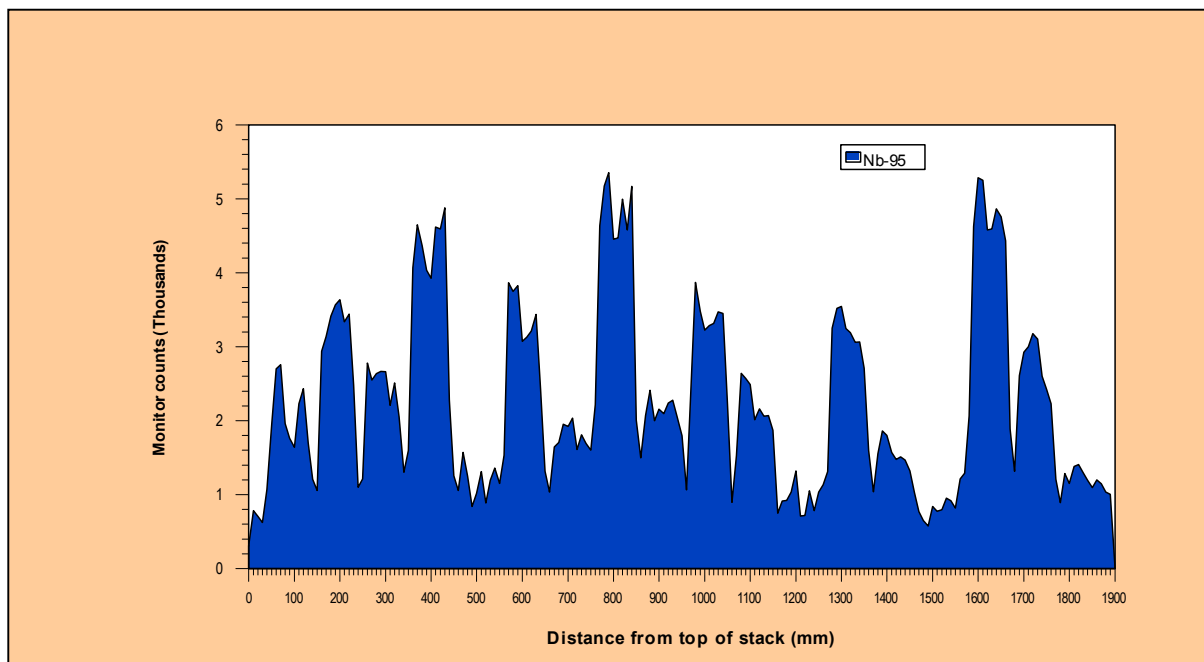


#### 4. VERIFICATION MEASUREMENTS USING CdZnTe DETECTOR

The verification measurements were conducted both for the higher and lower end of cooling time under investigation. The SCANDU verification profile of 9-month-old fuel bundles is shown in Fig. 7. The scanning was carried out from top to bottom of the stack along the 6th bundle position on each tray, at a speed of 1mm/s, which allowed counting time of 10 s. All the bundles were quite clearly verified employing either the ROI:1 (724 keV of Zr-95 / Nb-95) or ROI:2 (661.6 keV of Cs-137).



The verification profile of one-month-old fuel bundles using only 765 keV gamma ray line of Nb-95 is shown in Fig.7. All the bundles scanned are seen to be clearly verified – either by Cs-137 or through Nb-95 depending on the cooling time



## 5. CONCLUSIONS

The availability of high performance room temperature CdZnTe semiconductor detectors and the matching pulse processing equipment did contribute in achieving the objective to a very considerable degree. While for moderate cooling time ( $1 \text{ Yr} \geq T_c \geq 1 \text{ m}$ ) the verification was carried out making use of either or both the 722 keV Zr-95 / Nb-95 and the 662 keV Cs-137 gamma rays, for very short cooling time ( $< 1 \text{ month}$ ) only 722 keV gamma ray emission from Zr-95/ Nb-95 could be made use of. Depending upon the intensity of emitted radiation the signal-to-noise ratio for the conducted verification measurements ranged from 4-7 for 9-month-old fuel bundles to 2-4 for 1-month-old fuel bundles. In parallel with the testing carried and reported in this paper, the super grade CZT detectors have continued to be used for actual verification by the IAEA safeguards inspectorate with ever increasing experience and confidence.

The assessment of very low sensitivity silicon detectors has shown that due to its low sensitivity, it has a potential for use in the high count rate work. The encountered count rates are manageable and the pulse pile up effect is non existent. The fact that the % DT even in the immediate vicinity of short cooling time spent fuel bundles is insignificantly small, there are less stringent pulse processing requirements on the amplifier. Due to the fact that its pulse height response doesn't contain any specific region of interest, its integral gamma response alone could be used for the verification purposes. This detector has, however, provides a lower safeguards effectiveness since no spent fuel specific signatures are used for the verification.

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