Experimental Comparison between High Purity Germanium and Scintillator Detectors for Determining Burnup, Cooling Time and Decay Heat of Used Nuclear Fuel - 14488

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ABSTRACT

A experimental study of the gamma-ray energy spectra from used nuclear fuel has been performed. Four types of detectors were used to measure spectra from three PWR used fuel assemblies stored at the interim storage for used fuel in Sweden, CLAB: HPGe, LaBr3, NaI and BGO.

The study was performed in the context of used fuel characterization for the back end of the fuel cycle in Sweden. Specifically, the purpose was to evaluate the behaviour of the different scintillator detectors (LaBr3, NaI and BGO) and their ability to be used instead of HPGe detectors when determining spent fuel parameters such as burnup, cooling time and decay heat of the used fuel.

This paper presents results from the experimental study and an analysis of the capability of the detectors for used fuel characterization. The results shown are important when designing systems for used fuel characterization, e.g. for determining decay heat or fuel parameters concerning safeguards.

INTRODUCTION AND PURPOSE

The interim storage facility for used fuel (CLAB) in Oskarshamn, Sweden, is managed by the Swedish Nuclear Fuel and Waste Management Company (SKB AB). In CLAB, all used fuel generated from the nuclear power plants in Sweden are stored, awaiting future encapsulation and storage in a geological facility about 500 meters deep in the Swedish bedrock [1].

The used nuclear fuel assemblies will be encapsulated in copper canisters with a steel insert, providing a barrier between the used fuel and the clay that will act as a buffer between the canister and the surrounding bedrock. Safety requirements limit the maximum decay heat of the used fuel that can be put in the copper canister to about 1700 Watt. Therefore, it is important to assess the decay heat of fuel assemblies to be put in the canister. A method to measure the decay heat using gamma-ray spectroscopy has been developed earlier [2,3]. It is based on measuring the intensity of gamma radiation emanating from Cs-137 in the used fuel.

For safeguard purposes, it is important to verify the integrity the fuel assemblies before they is stored in a difficult to access storage, e.g. a deep geological repository. The fuel parameters burnup (BU) and cooling time (CT) of the used fuel is one set of parameters that can be measured and compared to operator declarations, thus enabling a verification of the used fuel. By measuring the gamma radiation emitted from Cs-137, Cs-134 or Eu-154 in the used fuel, BU and CT can be inferred [4,5].

In August 2013, the intensity of gamma radiation from three used pressurized water reactor

(PWR) fuel assemblies were measured in the pool of the CLAB facility. Four types of detectors were used: Three scintillator detectors (Lanthanum bromide - LaBr3, Sodium iodine - NaI and Bismuth germanate - BGO) and one high purity germanium (HPGe) detector. The purpose was to evaluate how well the passive gamma intensities from the fuel assemblies could be determined using different scintillator detectors (LaBr3, NaI and BGO) as compared to using a standard HPGe detector.

METHODS AND EQUIPMENT

Spectroscopic Methods Used To Determine Burnup, Cooling Time and Decay Heat

Using gamma-ray spectroscopy, it is possible to determine both the safeguards related parameters burnup and cooling time as well as the decay heat of the spent fuel.

Determining BU is based on measuring only Cs-137 or, e.g., both Cs-137 and Cs-134. For fuel assemblies with long cooling time, Cs-137 with its long half-life of 30 years is most widely used for passive gamma measurements. For younger fuel assemblies, Cs-134 is still available for assay (with a half-life of 2 years). The half-life of Eu-154 is about 8.6 years, implying that it is available for assay for some decades.

The activity of Cs-137 in the fuel assembly is nearly proportional to its BU and so is the ratio of the activities of Cs-134 and Cs-137, see reference [7]. The measurement of the intensity ratio is approximately independent of, e.g., detection efficiency. However, directly using these proportionalities required knowledge of the CT of the used fuel.

Reference [5] shows that BU can be determined independently of CT using measurements of the intensities from two isotopes, see equation (1).

$$BU = exp\left[\frac{ln\left(\left(\frac{k_2}{I_2}\right)^{\lambda_1}\cdot\left(\frac{I_1}{k_1}\right)^{\lambda_2}\right)}{\lambda_2\kappa_1-\lambda_1\kappa_2}\right] \qquad \text{(Eq. 1)}$$

Where

- I_1 and I_2 are two measured intensities of, e.g., Cs-137 and Cs-134.
- k_i and κ_i are fitting parameters for a calibration curve on the form $I = k\cdot BU^{\kappa}\cdot e^{-\lambda\cdot CT}$. For Cs-137 and Cs-134, κ is about 1 and 2, respectively.
- λ_i is the decay constant of isotope i.

For the purpose of this paper, to evaluate the detectors used, we assume an infinitely good calibration curve, i.e. which has zero uncertainty in the calibration parameters, and no correlation between the two measured intensities. It can then be shown that the relative uncertainty of BU can be written as in equation (2).

$$\frac{\Delta BU}{BU} = \frac{1}{|\lambda_2\kappa_1-\lambda_1\kappa_2|}\cdot\sqrt{\lambda_2^2\left(\frac{\Delta I_1}{I_1}\right)^2+\lambda_1^2\left(\frac{\Delta I_2}{I_2}\right)^2} \qquad \text{(Eq. 2)}$$

Reference [5] also elaborates on how CT can be determined, independently of BU, using measured intensities of, e.g., Cs-137 and Cs-134. Equation (3) is used in this case, with the

same parameters as used in equation (1).

$$CT = \frac{ln\left\{\left(\frac{\kappa_2}{I_2}\right)^{\kappa_1} \cdot \left(\frac{I_1}{\kappa_1}\right)^{\kappa_2}\right\}}{\lambda_2\kappa_1 - \lambda_1\kappa_2} \quad \text{(Eq. 3)}$$

Assuming again a good calibration and independent intensity measurements, we can write the relative uncertainty of CT as in equation (4).

$$\frac{\Delta CT}{CT} = \frac{\sqrt{(\kappa_2\Delta I_1/I_1)^2 + (\kappa_1\Delta I_2/I_2)^2}}{CT \cdot |\lambda_2\kappa_1 - \lambda_1\kappa_2|} \quad \text{(Eq. 4)}$$

The decay heat, P, can be determined using a measurement of the intensity of Cs-137. Reference [5] show that the decay heat is proportional to the Cs-137 intensity when including a correction factor for the fraction of the heat that originates from the decay of Cs-137. Therefore, the relative uncertainty of the determined decay heat is proportional to the relative uncertainty in the measured intensity, as shown in equation (5).

$$\frac{\Delta P}{P} \propto \frac{\Delta I}{I} \quad \text{(Eq. 5)}$$

It is implicitly assumed that the calibration curves established are applicable also when measuring unknown fuel assemblies, i.e. that the measuring geometry, detector system or type of fuel assembly is not changed between measurements.

The Gamma Scanning Equipment

Build into one of the pool walls at CLAB, there is a passive gamma measurement station that can be used to axially scan a nuclear fuel assembly in front of a collimator and detector. The distance between the center of the fuel assembly and the end of the collimator is about 2.5 m. An elevator in the pool allows vertical movement of the fuel assembly as well as for rotation of the fuel assembly, see figure 1. The fuel assembly is at all times kept in the pool, while the detector and data acquisition system is located in a room behind the pool wall. The detector station is not a fixed installation and the operator may change the detector of data acquisition system in the measurement room. References [5,6] reports geometrical details of the setup. The gamma-ray detector was mounted so that it allowed for easy exchange between different detectors without changing the measurement geometry.

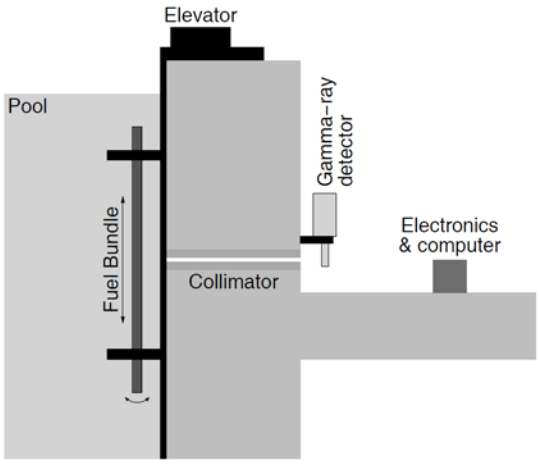

Figure 1. Schematic view of the gamma scanning equipment installed at CLAB. The arrows indicate how the nuclear fuel assembly can be translated and rotated in front of the collimator.

Data Acquisition System and Detectors

To measure the gamma radiation from the different kinds of detectors used, two data acquisition systems were used. For the HPGe detector, the LYNX system and the Genie 2000 software from Canberra Inc. was used. For the scintillator detectors, a DSPEC 50 system and the MAESTRO software from ORTEC Inc. was used. Both systems are based on digital signal analysis of the measured pulses from the detectors using trapezoidal shaping that allows for good energy resolution over a wide range of counting rates.

For the scintillator detectors, high voltage was provided by the DSPEC. For the HPGe detector, an electronic circuit that protects the detector from warm-up was used to supply high voltage. Table 1 summarizes data on the detectors and parameters for the data acquisition system. A set of attenuating sheets of different materials was positioned between the collimator and the detector: 11.93(2) mm of stainless steel, 1.29(1) mm of copper, 8.55(8) mm of lead. The detectors were all positioned horizontally at a distance of 11.83(2) cm from the collimator opening.

Nuclear fuel assemblies measured

Three PWR 17x17 used nuclear fuel assemblies were measured in this study. They represent a relatively large span of parameters of the used PWR assemblies stored in CLAB. Table 2 summarizes the data on the fuel assemblies.

Table 1. Parameters for the data acquisition system and data on the detectors.

Detector	Parameter	Value
HPGe detector	Type	N
	Relative efficiency @ 1332 keV	44 %
	Energy resolution @ 1332 keV	2.0 keV
	Trapezoidal rise time	3.0 µs
	Trapezoidal flat top	0.6 µs
LaBr3 detector	Size (diameter x length) [mm]	25.4 x 50.8
	Energy resolution @ 662 keV	2.6 %
	High voltage	+691 V
	Trapezoidal rise time	0.8 µs
	Trapezoidal flat top	0.3 µs
BGO detector	Size (X x Y x Z) [cm]	2 x 3 x 5
	High voltage	-800 V
	Trapezoidal rise time	0.8 µs
	Trapezoidal flat top	0.3 µs
NaI detector	Size (diameter x length) [mm]	50.8 x 50.8
	Energy resolution @ 662 keV	7.2 %
	High voltage	+560 V
	Trapezoidal rise time	0.8 µs
	Trapezoidal flat top	0.3 µs

Table 2. Data on the used fuel assemblies studied in this work.

Assembly Id	Type	BU [MWd/tU]	CT [years]	Initial enrichment [%]
PWR 5	17×17	46866	5.2	3.94
PWR 19	15×15	35027	28.3	3.20
PWR 24	17×17	23151	18.1	2.10

Spectra evaluation

The measured gamma-ray energy spectra are displayed in figure 2. As can be seen in figure 2, the gamma energy spectrum is dominated by the Cs-137 peak at 662 keV, especially for cooling times beyond 18 years. For a cooling time of about 5 years, the spectrum also shows strong Cs-134 peaks at 605 keV and 796 keV and a Eu-154 peak at 1274 keV. The HPGE detector delivers the best energy resolution and the worst resolution is obtained from the BGO detector.

The identified peaks are resolved for the HPGe and for the LaBr3 detector for all cooling times in the measured set of assemblies. At a cooling time of about 5 years, for the NaI detector, the Cs-134 peak at 605 keV interferes with the Cs-137 peak at 662 keV and for the BGO detector, the energy resolution is not enough to differentiate between the peaks.

For the HpGe and LaBr3 detector, a relatively simple peak area evaluation method can be used. This holds true also for the BGO detector but only at long cooling times. For the NaI detector, more advanced peak area evaluation using peak fitting with overlapping peaks may be needed to determine the individual intensities, especially at short cooling times.

Considering a fuel assembly with unknown parameters, the use of a detector that can not resolve the peaks of interest might not be the best approach. Therefore, we focus the evaluation on HPGe and LaBr.

Using the built-in capabilities of the Maestro software (from ORTEC Inc, for the LaBr3 detector) and the Genie-2000 software (from Canberra Inc, for the HPGe detector), the Cs-137 peak was quantified for all measured spectra. The results are given in table 3. The measured intensity from Cs-137 using the LaBr3 detector was consistently about a factor of 3 lower than for the HPGe detector.

Table 3. Values on the Cs-137 peak in all measured spectra for the HPGe and LaBr3 detectors. Intensities are given in counts per second. Energy resolutions (FWHM) are given in percent. Real acquisition times were about 10 minutes for all measurements except for the HPGe measurement on PWR 19 which acquired data for about 40 minutes.

Assembly Id:	**PWR 5**		**PWR 19**		**PWR 24**	
Detector	**Intensity**	**FWHM**	**Intensity**	**FWHM**	**Intensity**	**FWHM**
HPGe	2755 (4)	0.37	1112 (1)	0.28	817 (2)	0.27
LaBr3	838 (11)	3.5	332 (6)	3.7	292 (5)	3.6
Ratio (HPGe/LaBr3):	3.3		3.3		2.8	

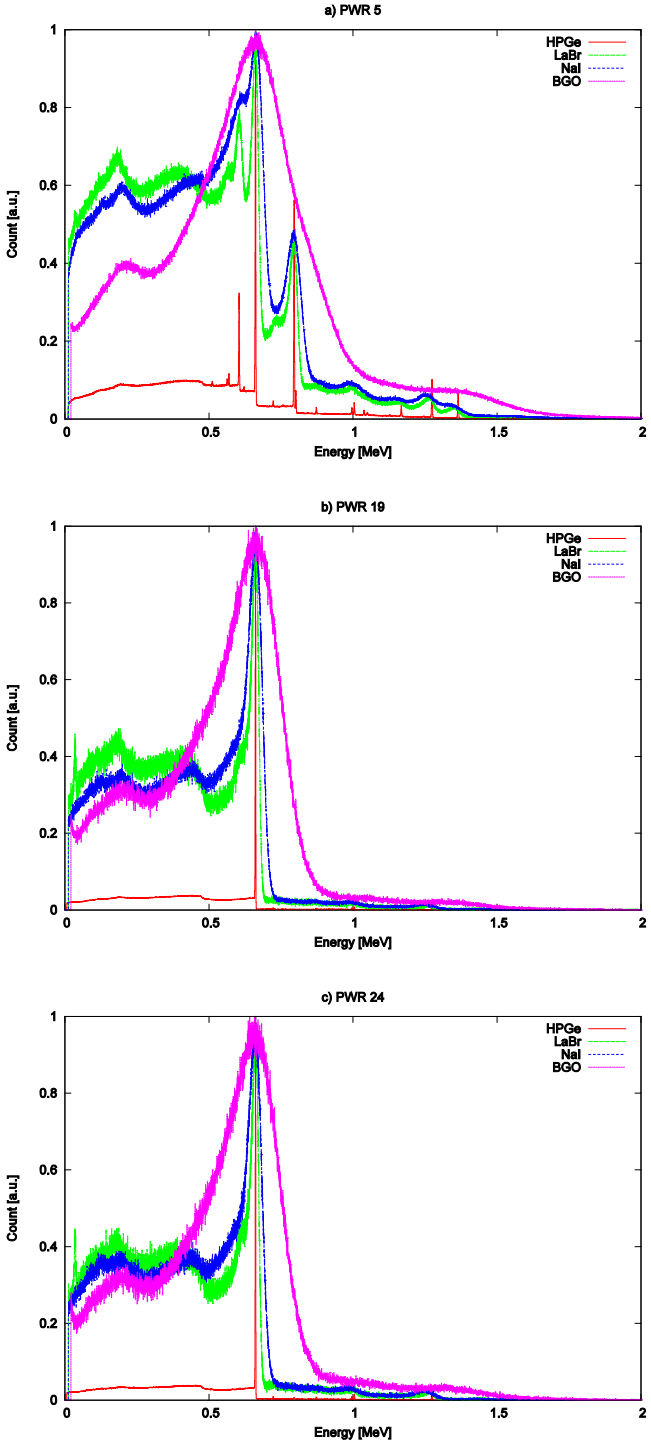

Figure 2. Measured gamma-ray energy spectra, normalized so that the maximum height of the Cs-137 peak is one. The parameters of the measured fuel assemblies are shown in table 2.

Discussion

As noted above, the LaBr3 detector is usable also for short cooling times. The evaluated intensity of the Cs-137 peak is proportional to the intensity evaluated for the HPGe detector.

Using equations (2), (4) and (5), uncertainties in determined BU, CT and decay heat can be evaluated. Since this study is focused on a viability study, we leave a full uncertainty analysis for future studies. However, it can be stated that the relative uncertainty in these parameters is in general proportional to the relative uncertainty of the measured intensity. For determination of CT using equation (3), the uncertainty is lower at small cooling times.

For long cooling times, more than 18 years, all detectors tested in this study can be used to evaluate the peak area of the dominant Cs-137 peak at 662 keV. To be able to determine BU at these long cooling times when not much more than Cs-137 is visible in the gamma-ray energy spectrum, knowledge of CT is needed to correct the measured intensities to a common reference time (usually at zero CT).

At shorter cooling times, the energy resolution of NaI and BGO becomes a limiting factor to be able to determine reliably the peak intensities.

CONCLUSIONS

It is concluded that LaBr3 can be used for passive gamma assay of used fuel, with its energy resolution of about 3% being good enough to resolve the most dominant gamma peaks in the energy spectrum even at short cooling times (in the order of 5 years).

For long cooling times even NaI or BGO detector can be used to evaluate the intensity of the dominant Cs-137 peak at 662 keV. According to the data measured in this study, they are usable for cooling times beyond 18 years. Further studies are needed to determine at which cooling times (earlier than 18 years) that NaI and BGO detectors become a usable option.

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ACKNOWLEDGEMENTS

This work was performed in the framework of the collaboration agreement between the EU and the US-DOE-NNSA in the field of Nuclear Material Safeguards and Security Research and Development. The personnel at the interim storage facility for used fuel (CLAB) in Oskarshamn, Sweden, are acknowledged for their work in supporting this effort.