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Nuclear spent fuel parameter determination using multivariate analysis of fission product gamma spectra

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ABSTRACT

In this paper, we investigate the application of multivariate data analysis methods to the analysis of gamma spectroscopy measurements of spent nuclear fuel (SNF). Using a simulated irradiation and cooling of nuclear fuel over a wide range of cooling times (CT), total burnup at discharge (BU) and initial enrichments (IE) we investigate the possibilities of using a multivariate data analysis of the gamma ray emission signatures from the fuel to determine these fuel parameters. This is accomplished by training a multivariate analysis method on simulated data and then applying the method to simulated, but perturbed, data.

We find that for SNF with CT less than about 20 years, a single gamma spectrum from a high resolution gamma spectrometer, such as a high-purity germanium spectrometer, allows for the determination of the above mentioned fuel parameters.

Further, using measured gamma spectra from real SNF from Swedish pressurized light water reactors we were able to confirm the operator declared fuel parameters. In this case, a multivariate analysis trained on simulated data and applied to real data was used.

1. Introduction

In accordance with nuclear safeguards treaties and regulations, e.g. (IAEA, 1968), nuclear material such as spent nuclear fuel (SNF) needs to be verified. This includes independent inspections by regulatory bodies and the use of non-destructive assay in order to verify the presence of the material and its properties.

Of interest can be the verification or determination of fuel parameters such as initial enrichment (IE), burnup (BU) and cooling time (CT). For nuclear safeguards, BU is important for several reasons; i) it can be used to support a conclusion that the fuel has been operated in accordance with the expectations of civil nuclear fuel cycle, ii) together with IE it gives an indication of the total fissile content of the fuel and iii) in itself it is a good proxy of the plutonium content in the fuel. In the case of fuels being transferred to a difficult to access storage, such as a final repository, it is particularly important that all measurements are done with as great detailed as possible since re-measurements are typically not foreseen. Further, in a final repository, detailed knowledge of IE and CT is also important as these are safety related parameters that connects to the boundary conditions on both criticality as well as decay heat production (SKB, 2016).

Measurements on SNF are to a large extent conducted using non-destructive techniques. The purpose of these measurements can be to verify the presence of special nuclear material and operator declared information, with as little intrusion in facilities and activities as possible. There are a number of measurement techniques that can provide relevant information, and one example of a non-destructive technique is passive gamma-ray spectroscopy (GRS). Some established analysis methods of GRS data use the ratio between the contributions from two different fission products (Rinard, 1983; IAEA, 1979; Cobb, 1982; Philips, 1980; Jansson, 2002; Willman, 2006a,b). One case is the ratio between the two isotopes Cs-134 and Cs-137, which can be used to determine BU.

The method relies on the fact that the build up of Cs-137 scales linearly with BU while the build up of Cs-134 has a roughly quadratic scaling. The ratio will therefore be roughly proportional to BU. One advantage with this method is its transparency and close connection to the physical process; namely that Cs-137 is formed directly, while Cs-134 is formed in a multistep process. However, the method is limited on its own and needs a correction for CT since the two isotopes have significantly different half-lives. Typically, the discharge of nuclear fuel is witnessed by safeguards

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inspectors, and if a Continuity of Knowledge has been maintained, CT is well known. However, if the Continuity of Knowledge if lost, CT can no longer be considered as a known quantity, and a (re)verification of this parameter is desirable.

Although current verification technologies can in many cases be considered sufficiently good, there is room for improvement.

- The International Atomic Energy Agency (IAEA) is striving to more extensively make use of automated and unattended measurement and analysis tools in order to become more efficient. New instruments and analysis tools should, if possible, be able to extract more information from objects and data than today, and perform the analysis more efficiently.
- 'Most existing technologies *verify* fuel parameters against operators' declarations; they do not aim at *determining* them.
- It is preferred that fuel parameters and characteristics can be determined with a precision matching that of calculations.
- There are fuels and materials in non-conventional configurations (damaged, reassembled or dissolved fuel) and in new types of facilities (encapsulation and long-term-storage, fuel reprocessing and recycling facilities) which are not as well quantified using existing instruments and analysis techniques.

With high-resolution gamma spectroscopy measurements of SNF, up to a dozen different fission products can be detected, and many of them have significantly different yields depending on the fissioning isotope (e.g. U-235 and Pu-239). Therefore, if the total gamma spectrum is considered, more information about the underlying fuel parameters can be extracted compared to if only the ratio of two isotopes is used. However, the complete data is high-dimensional, and a simple model connecting the data to the fuel parameters of interest is not always straightforward to obtain. In such cases, multivariate analysis (MVA) methods are often employed,

MVA methods have been considered previously for monitoring of nuclear material. One example is (Orton, 2011) where the applicability of MVA methods to monitor the operation and the use of process monitoring data to possibly detect diversion or manipulation of the process. Within the Next Generation Safeguards Initiative Spent Fuel project (Veal, 2010; Humphrey, 2012), a large number of gamma-ray and neutron measurement techniques have been studied with the purpose of combing the data from several instruments with MVA methods to probe the plutonium content (Burr, 2015).

The question we address in this paper is to which extent we can obtain information on the fuel parameters CT, BU and IE from the combined analysis of all measurable fission product signals in the gamma spectrum. We also investigate what limitations there are, e.g. when fuels with long cooling times are measured and the number of measurable fission products decrease. The analysis in this paper is limited to data from passive gamma spectroscopy, but we note that multivariate analysis methods are well suited for combining data from different measurements, e.g. gamma spectroscopy and different neutron measurements.

In this study, we apply multivariate analysis techniques to high-resolution gamma-ray spectroscopy data from SNF. Specifically, we discuss the application of two methods: Principal Component Analysis (PCA) and Partial Least Squares (PLS) regression, see (Pearson, 1901; Wold, 2001). PCA is used in a first exploratory stage to find the internal structures of the data and to identify cases that are suitable for further analysis. PLS regression is then used to estimate the fuel parameters from the information provided by the fission products. We build a PLS regression model using simulated synthetic data. The accuracy of the PLS regression is tested by introducing perturbations and model errors on a second simulated data set. Finally, we apply the PLS regression models to data from gamma scanning of real SNF to demonstrate the applicability of the analysis model.

We consider only the relative contributions to the signal from different fission products. No measurements and calibrations on absolute scales are therefore necessary. This makes the analysis very robust as many experimental factors that could bias absolute methods cancel out in relative measurements.

2. Gamma-ray emitting isotopes

Passive gamma spectroscopy is a non-destructive assay (NDA) technique, which has been used for decades in order to verify spent nuclear fuel and radioactive material in general (IAEA, 1991; IAEA, 2011; LANL, 2013). High-resolution gamma spectrometers that are able to resolve closely lying isotope-specific gamma energies are typically used. This includes e.g. high purity germanium spectrometers (HPGe). Depending on the initial enrichment (IE), burnup (BU), cooling time (CT) as well as the irradiation history of the spent nuclear fuel, the relative contribution from different fission product isotopes will vary. This is the basis for the analyses in this paper.

The gamma-emitting isotopes are characterized by their half-lives, the emitted gamma energies and the branching ratio. The half-lives of measurable isotopes in SNF vary from seconds to decades. For an isotope to be of interest in this work it needs to fulfill the following criteria:

i) The fission yield associated with the fission product should be large enough to produce a measurable quantity in the fuel.
ii) The half-life should be long enough to allow for a measurable quantity to remain in the fuel at the time of measurement.
iii) The energy of the emitted gamma rays should be high enough in order to be detectable.
iv) The intensity (probability of emission) of the detectable gamma rays should be high enough for them to be visible in the spectrum. In particular when considering the Compton continuum.
v) The isotope should not be gaseous and remain in the fuel matrix.

Except for point v), the above criteria are not fixed, but vary depending on which fuel is measured, and at what point in time it is measured. For example, what is considered a long enough half-life depends on the cooling time of the fuel. Further, the required yield of a fission product, as well as the required intensity of its gamma rays, depends on the energy of the gamma rays. High-energy gamma rays are more likely to escape the fuel matrix and be detectable. In addition, the Compton background decreases with energy, and for higher gamma ray energies, weaker signals can therefore be detected.

As a starting point we have selected 12 gamma-emitting fission products, which are summarized in Table 1. The starting point for our selection is a compilation in (SIPRI, 2015) of isotopes that have previously been successfully measured and are of interest for nuclear safeguards. We selected only isotopes with half-lives longer than 1 month, implying that we have limited the study to fuels with more than about 0.5 years cooling times. In addition to the compilation in (SIPRI, 2015) we also include Eu-154, which is commonly used for fuel verification.

Examples of two experimental gamma ray spectra from spent PWR fuel that were readily available to the authors at the time of writing (Jansson, 2014; Jansson, 2016) are shown in Fig. 1. The data were collected with a high purity germanium (HPGe) detector