Nuclear Inst. and Methods in Physics Research, A 897 (2018) 85–91

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Estimating irradiated nuclear fuel characteristics by nonlinear multivariate regression of simulated gamma-ray emissions

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ARTICLE INFO

Keywords:
Nuclear safeguards
Spent nuclear fuel
Gamma-ray
Multivariate analysis
Nonlinear regression

ABSTRACT

In addition to verifying operator declared parameters of spent nuclear fuel, the ability to experimentally infer such parameters with a minimum of intrusiveness is of great interest and has been long-sought after in the nuclear safeguards community. It can also be anticipated that such ability would be of interest for quality assurance in e.g. recycling facilities in future Generation IV nuclear fuel cycles.

One way to obtain information regarding spent nuclear fuel is to measure various gamma-ray intensities using high-resolution gamma-ray spectroscopy. While intensities from a few isotopes obtained from such measurements have traditionally been used pairwise, the approach in this work is to simultaneously analyze correlations between all available isotopes, using multivariate analysis techniques. Based on this approach, a methodology for inferring burnup, cooling time, and initial fissile content of PWR fuels using passive gamma-ray spectroscopy data has been investigated. PWR nuclear fuels, of UOX and MOX type, and their gamma-ray emissions, were simulated using the Monte Carlo code Serpent. Data comprising relative isotope activities was analyzed with decision trees and support vector machines, for predicting fuel parameters and their associated uncertainties.

From this work it may be concluded that up to a cooling time of twenty years, the 95% prediction intervals of burnup, cooling time and initial fissile content could be inferred to within approximately 7 MWd/kgHM, 8 months, and 1.4 percentage points, respectively.

An attempt aiming to estimate the plutonium content in spent UOX fuel, using the developed multivariate analysis model, is also presented. The results for Pu mass estimation are promising and call for further studies.

1. Introduction

1.1. Background

Organizations such as the International Atomic Energy Agency (IAEA) review nuclear facility operation records and other information provided by a State to verify that it acts in compliance with its commitments. As a means of verification, inspections of nuclear facilities and measurements of nuclear materials are conducted. Such activities allow for safeguards inspectors to independently draw conclusions about the authenticity and accuracy of the operator declared information.

The quest to develop measuring instruments with the ability to infer parameters for verifying parameters of spent nuclear fuel has been a long-term undertaking by the safeguards community; see e.g. [1–3] and [4]. In particular, in a case where declared information about spent nuclear fuel is lost or deficient, such ability is of great importance. However, while some concepts such as high-resolution gamma-ray spectroscopy provide data that may be suited for this purpose, the analysis of this data has thus far been performed by using rather basic methods. As is emphasized in this paper, the strong and complex correlations between isotope concentrations and various fuel parameters call for an advanced analysis methodology in order to extract the potentially large amounts of information from the measurements. Multivariate analysis techniques, with their ability to reveal underlying structures in seemingly unstructured data, could thus be of interest in this context.

1.2. Scope of this study

The aim of this work was to theoretically investigate the feasibility of using multivariate analysis methods to estimate burnup, cooling time and initial fissile content of irradiated PWR fuel without reliance on operator declarations.

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https://doi.org/10.1016/j.nima.2018.04.034
Received 10 December 2017; Received in revised form 15 April 2018; Accepted 17 April 2018
Available online 22 April 2018
0168-9002/

Table 1
List of the selected isotopes and their correspondence to the cooling times (CT) used in this work.

Isotope	Half-life	Group 1 CT: 0–1 y	Group 2 CT: 1–10 y	Group 3 CT: 10–20 y
^{141}Ce	32.5 d	✓		
^{95}Nb	35.0 d	✓		
^{91}Y	58.5 d	✓		
^{95}Zr	64.0 d	✓		
^{144}Ce	285 d	✓	✓	
^{106}Ru	372 d	✓	✓	
^{134}Cs	2.07 y	✓	✓	✓
^{154}Eu	8.60 y	✓	✓	✓
^{137}Cs	30.1 y	✓	✓	✓

The work presented here rests upon findings from two previous papers [5] and [6]. In [5], simulated gamma-ray intensities were analyzed using linear multivariate regression models for estimating the three fuel parameters; burnup, cooling time and initial fissile content. This paper presents a similar approach as in [5], but the analysis has been extended to include more complex nonlinear regression models with the presumption that even higher accuracy would be attainable due to the nonlinear nature of the underlying data examined in [5] and [6]. This paper in addition studies the possibilities of estimating the total plutonium content of irradiated fuel. This capability is explicitly targeted also in other safeguards development projects such as the Next Generation Safeguards Initiative (NGSI) [7,8].

In [1], a methodology based on multivariate analysis for discrimination between UOX and MOX fuels was investigated. The results from [1] suggest that it is possible to successfully discriminate between UOX and MOX fuels. Hence, in this work it is considered known whether the PWR-fuel under investigation is of UOX or MOX type. This knowledge is crucial since two models, one for UOX and one for MOX fuel, can be used separately in the analysis. Further, in [1] it was concluded that the gamma emissions from fuel assemblies with cooling times longer than 20 years do not provide adequate information for the analysis to be successful. In this work, 20 years was therefore the upper limit for the cooling times investigated.

2. Isotopic contents and gamma-ray emissions of irradiated fuel

The nine isotopes selected in this work and shown in Table 1 are frequently recommended in the literature relating to fuel characterization, see e.g. [9,10] and [11]. The isotopes were selected because:

- They cover a wide range of half-lives, such that at least a few of them are detectable up to a cooling time of 20 years (for more details regarding these isotopes see for example [6]). Table 1 also shows which isotopes are used in the three groups of cooling times considered in this work.
- They exhibit prominent gamma energies that can, in principle, be identified using passive high-resolution gamma-ray spectroscopy [5]. In practice, however, such measurements are by no means trivial to conduct due to various experimental constraints such as high counting rates and therefore large dead times, large background levels due to e.g. Compton scattering. Although not part of this work, such experimental issues must be thoroughly addressed. This subject was discussed in [6].

The source activity of each isotope was calculated in the way as described in [6]. Relative rather than absolute activities were used, with the sum of activities for each fuel assembly normalized. To simulate the effect of various sources of experimental uncertainty, including statistical fluctuations, a uniformly distributed noise of 3% was added to the simulated gamma activities. The reasoning, based on a semi-empirical approach using uncertainty estimations from previous gamma-scanning measurements on spent nuclear fuel [2,12], is more elaborately presented in [6].

3. Fuel simulations

The concentration of each chosen isotope was generated by simulating irradiation histories of the fuel, and the concentrations were used to calculate isotope-specific total gamma activities. Table 2 shows some final preprocessed data used for the multivariate analysis performed on fuels with cooling times between 1 and 10 years, therefore consisting of five isotopes. Note that the sum of total activities for each fuel item has been normalized to 1.

Three separate data sets were generated for specific purposes: a training set was used to train the multivariate model to predict the fuel parameters, an optimization set was used to fine-tune the multivariate model itself, and an evaluation set was generated for assessing the overall performance of the methodology. In the following sections, these sets are further discussed.

3.1. The training data set

For the training set, approximately 110,000 combinations or fuel configurations of burnup, cooling time and initial fissile content were modeled in the Monte Carlo code Serpent, version 2.1.28 [13]. The simulations considered PWR fuel assemblies only and of these, half were of MOX type and half of UOX type. The fuel parameters used were distributed among the fuel assemblies such that each assembly had an individual configuration of fuel parameters, while all regions of the parameter space were represented in the data set. The ranges of the fuel parameters were:

- initial fissile content: 2%–5%,
- burnup: 1–60 MWd/kgHM, and
- cooling time: 0–20 years.

The number of irradiation cycles for the fuel varied between one and six, since a maximum burnup reached during one irradiation cycle was set to 10 MWd/kgHM.

Shuffling of fuel between the irradiation cycles, which would add more realism to the simulations, has been omitted in this study. Moreover, operational conditions such as fuel temperature, moderator temperature, and moderator density have not been varied. Several such sources of realism have been included in spent fuel libraries produced within the NGSI effort [14]. The libraries contain, however, relatively few fixed combinations of burnup, cooling time, and enrichment.

The parameter space was divided into three-dimensional subspaces, $V_{\beta\varepsilon CT}$, each spanned by burnup (β), initial fissile content (ε) and cooling time (CT) according to:

$$V_{\beta\varepsilon CT} = A_\beta \times B_\varepsilon \times C_{CT} \tag{1}$$

where $A_\beta = 1$ MWd/kgHM, $B_\varepsilon = 0.1\%$ and $C_{CT} = 1$ year (for CT < 1 year: $C_{CT} = 1$ month). This gives rise to 60 × 31×31 rectangular subspaces. The fuel parameter values for a specific fuel assembly were defined by uniformly at random selecting one location within each subspace.

Because high burnup levels are not achievable for fuels with low initial fissile contents, the empirical relationship between initial enrichment (ε, in percent), and maximal reachable burnup (BU, in MWd/kgHM) used in [6] and shown in Eq. (2) was applied to the data. Data points not satisfying this constraint were not included in generated data sets.

$$\varepsilon = l \cdot BU^k \tag{2}$$

with $l = 0.31$ and $k = 0.65$ [15].