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Characterization of the non-uniqueness of used nuclear fuel burnup signatures through a Mesh-Adaptive Direct Search

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ABSTRACT

The use of passive gamma and neutron signatures from fission indicators is a common means of estimating used fuel burnup, enrichment, and cooling time. However, while characteristic fission product signatures such as ^{134}Cs, ^{137}Cs, ^{154}Eu, and others are generally reliable estimators for used fuel burnup within the context where the assembly initial enrichment and the discharge time are known, in the absence of initial enrichment and/or cooling time information (such as when applying NDA measurements in a safeguards/verification context), these fission product indicators no longer yield a unique solution for assembly enrichment, burnup, and cooling time after discharge. Through the use of a new Mesh-Adaptive Direct Search (MADS) algorithm, it is possible to directly probe the shape of this "degeneracy space" characteristic of individual nuclides (and combinations thereof), both as a function of constrained parameters (such as the assembly irradiation history) and unconstrained parameters (e.g., the cooling time before measurement and the measurement precision for particular indicator nuclides). In doing so, this affords the identification of potential means of narrowing the uncertainty space of potential assembly enrichment, burnup, and cooling time combinations, thereby bounding estimates of assembly plutonium content. In particular, combinations of gamma-emitting nuclides with distinct half-lives (e.g., ^{134}Cs with ^{137}Cs and ^{154}Eu) in conjunction with gross neutron counting (via ^{244}Cm) are able to reasonably constrain the degeneracy space of possible solutions to a space small enough to perform useful discrimination and verification of fuel assemblies based on their irradiation history.

1. Introduction

The use of passive gamma-ray signatures from characteristic fission products is a staple for non-destructive burnup analysis of used nuclear fuel, both for burnup credit applications (for used nuclear fuel storage and management) as well as for safeguards and material accountancy applications. In this latter case, passive gamma-ray measurements are typically used as either a gross estimator of nuclear fuel burnup [1–3] (i.e., to reconstruct burnup gradients across assemblies) or are combined with other techniques to verify operator declarations of the assembly irradiation history with the objective of establishing total assembly plutonium content via reconstruction of the assembly isotopic content through depletion simulations with the estimated burnup, such as through the use of depletion codes like ORIGEN (part of SCALE) [4–6]. In this latter case, calculations of the assembly plutonium content rely on estimates of the fuel burnup, enrichment, and cooling time following its last irradiation cycle [7,8].

Passive gamma measurements of prominent gamma signatures are typically used as burnup and cooling time indicators, such as ^{137}Cs, ^{154}Eu, or ratios of gamma lines such as the ratio of ^{134}Cs to ^{137}Cs intensity [1–3,9,10]. These nuclides are used due to both their well-established relationship with assembly burnup (and in certain cases, cooling time) as well as their relatively prominent gamma signatures capable of being distinguished within the complex spectrum of spent fuel assemblies [9]. Passive non-destructive analysis (NDA) techniques (including both passive gamma spectroscopy and passive measurements of gross neutron counts [1,11,12]) offer a preferred pathway for estimating used fuel inventories given that they can be performed relatively quickly and inexpensively compared to destructive analysis techniques and require minimal instrument complexity [13]. As a result, passive gamma signatures analysis continues to serve as a foundation for safeguards technology development efforts such as the Next Generation Safeguards Initiative [10,14,15].

Beyond characterization of spent fuel plutonium content, passive gamma NDA indicators are likewise frequently cited as a means of establishing a unique "fingerprint" for assemblies, including for cases such as re-establishing continuity-of-knowledge upon a loss of on-site power [8] or for termination of

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safeguards at a geological repository [12]. In these types of applications, measurements would ideally be able to uniquely verify operator declarations on the basis of passive signatures; however, as a practical matter, such systems are typically oriented around the ability to verify (or reject) operator declarations (such as cycle when of an assembly was discharged or its discharge burnup). For example, assuming typical cycle lengths on the order of 12–18 months, an uncertainty of less than ± 1 year would be expected to discriminate between discharge cycles. Similarly, NGSI has expressed a goal of characterization of plutonium within assemblies within $\pm 5\%$ [14,15], which roughly corresponds to the same level of uncertainty in discharge burnup.

Unique determination of assembly initial enrichment is more challenging and is typically considered beyond the means of passive NDA techniques alone [16]; however other researchers have claimed to make unique discrimination of the initial enrichment by employing semi-empirical relationships between burnup and initial enrichment based on the assumption that nuclear plant operators would seek to minimize operating margins [17]. While relying on estimates of cooling time and discharge burnup would allow safeguards inspectors to narrow down an assembly to within a discharged batch of assemblies (given a sufficiently tight tolerance on these parameters), the ability to independently establish an assembly's initial enrichment (or at the very least to discriminate between different potential fuel enrichments within a single batch, where differences can range on the order of 1–3% ^{235}U) is still potentially required to provide unique identification of assemblies.

In an ideal circumstance, a truly accurate reconstruction of isotopic inventories would rely on information provided directly from the reactor operator. However, given that a goal of safeguards measurements is to independently verify operator declarations, measurements from the fuel must serve to act as a proxy for the fuel parameters required to accurately reconstruct the assembly isotopic content. Assuming that the concentrations of burnup indicator nuclides are unique to the specific combination of fuel enrichment, burnup, and cooling time, the total plutonium inventory within the assembly is therefore also unique. Similarly, given a specified limiting measurement precision σ_N, it follows that the space of plutonium inventories would likewise show some statistical uncertainty σ_{Pu}, proportional to the measurement uncertainty in burnup.

However, while characteristic fission product signatures such as ^{134}Cs, ^{137}Cs, ^{154}Eu, and others are generally reliable estimators for used fuel burnup within the context where the assembly irradiation history is well-known, prior work by Cheatham and Francis has indicated that the space of solutions based on burnup indicators is in fact not unique for the space of initial fuel enrichment, burnup, and cooling time indicators [18]. Rather, they observed that a phase space of non-unique combinations of reactor parameters exist, wherein the inventories of burnup indicator nuclides are effectively indistinguishable from one another. Put another way, there exists a non-trivial space in the enrichment, burnup, and cooling time domains that yield the same inventories of burnup indicator nuclides within some measurement tolerance σ_N. Therefore, the same measured burnup indicator species yields a range of potential plutonium concentrations in the fuel. (Note that while NDA measurements would still uncover gross operator misrepresentations of an assembly's irradiation history, such as short-cycling intended to favorably manipulate the ^{239}Pu-to-^{240}Pu ratio, smaller uncertainties in total assembly Pu content are still relevant to contexts such as front-end accountancy measurements for reprocessing facilities.)

A useful consideration for passive burnup signatures analysis for used fuel burnup information in such contexts is therefore the extent of the non-uniqueness of this signature space, i.e. the size of the phase space made up of potential alternative assembly irradiation history characteristics (initial enrichment, burnup, and cooling time) which yield similar gamma signatures and in particular the influence of unconstrained parameters such as the time after discharge before measurement and the achievable measurement uncertainty on key signatures on the size of this phase space.

In this paper, we propose a new method for characterizing the shape of this degenerate signature space through the use of a Mesh Adaptive Direct Search (MADS) algorithm. By coupling the MADS algorithm directly with the latest ORIGEN application program interface (API) [19], it is thus possible to automate the exploration of the phase space shape characteristic of individual nuclides both as a function of a constrained parameters (such as the assembly's initial enrichment and irradiation history) as well as its unconstrained parameters (i.e., time before measurement and measurement uncertainty of individual nuclides). The goal of this work is to evaluate how this "degeneracy space" evolves with particular characteristics such as the nuclide identifier species, cooling time, and potential combinations of nuclide measurements that can be used to constrain the shape of the space (thereby limiting the uncertainty in calculated plutonium content).

2. Theory and methods

The objective of this method to determine the potential size of a group of ambiguous solutions (phase space), within which the concentrations of all indicator nuclides vary within a given tolerance ($\pm \sigma_N$). For example, if the only indicator nuclide is ^{137}Cs and $\sigma_{137} = \pm 5\%$, the phase space will be the group of solutions which contain a ^{137}Cs concentration within 95–105% of the ^{137}Cs concentration in the nominal case, regardless of the concentrations of other nuclides. This tolerance accounts for the uncertainty inherent in any measurement method.

To find the phase space for arbitrary indicator nuclides and thresholds, we created a tool called OrigenDSA, or the ORIGEN Degenerate Signatures Analysis. The OrigenDSA tool builds directly upon the new ORIGEN API (to be released as part of SCALE 6.2) [19] in order to efficiently harness ORIGEN for performing depletion calculations. Here, the search for degenerate assembly history parameters is performed via a Mesh-Adaptive Direct Search algorithm (MADS), which treats the group of all possible solutions as a three-dimensional space, refining the interesting solutions until the phase space appears as a solid shape embedded in the search space.

2.1. Estimation of used fuel burnup from passive gamma/neutron signatures

Gamma rays emitted from used fuel assemblies are the product of specific fission product decays. By counting the number of photons emitted (and making the appropriate efficiency corrections), the gamma ray intensity can be correlated to the inventory of the fission product species in question as follows (Eq. (1)) [9]:

$$I = \epsilon \kappa S N \lambda e^{-\lambda t} \tag{1}$$

where

- $I =$ gamma ray count rate (cps)
- $\epsilon =$ absolute detection efficiency (including self-attenuation of gammas within the fuel, detector solid angle, and detector intrinsic efficiency)
- $\kappa =$ decay line branching ratio (γ/decay)
- $N =$ number of fission product nuclei (atoms)
- $\lambda =$ fission product decay constant (decays/nucleus · sec)
- $t =$ cooling time before measurement (seconds)