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Total neutron emission generation and characterization for a Next Generation Safeguards Initiative spent fuel library

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ABSTRACT

In March of 2009 the Next Generation Safeguards Initiative of the U.S. Department of Energy began a nominal five year spent fuel research effort with the goal of: (1) quantifying plutonium content in spent nuclear fuel through the use of non-destructive assay (NDA) techniques; (2) quantifying the capability of these NDA techniques to quantify burnup, cooling time, and initial enrichment, as well as detect pin diversions, for spent fuel assemblies. These NDA techniques were first scoped in computation space in order to understand detection limitations and down select for further testing. Multiple Monte Carlo based spent fuel libraries (SFL's) were developed to be used as source terms for assessing the detection limitations of each of these techniques. The characterization of the SFL's provide an analysis of different burn strategies used for spent fuel, functional fits for state point analysis, and the components of the neutron signal.

This paper details both a methodology for generating the spatially dependent gross neutron emission (GNE) as well as a characterization of the GNE for a particular NGSI SFL for two different shuffle patterns. The GNE was characterized by examining three major components: (1) the total neutron emission; (2) the (α, n) and spontaneous fission emissions; and (3) the main contributors to the spontaneous fission and (α, n) emission signals. The total neutron emission was characterized by looking at two different shuffle patterns and using three different pin analysis techniques. The pin analysis techniques used were: (1) sum all the pins in the assembly; (2) create four pin zones to be summed; and (3) examine specific pins. These characterizations provide a large set of data with integral values (cooling time vs emission and burnup vs emission) and derivative values (isotopic percent composition) with visual depiction and detailed explanation. The characterizations also provide a comparison of the effects of shuffle schemes on the neutron emission signal for two different shuffle patterns. This data can be used in the development of time and burnup dependent normalized functions to describe the trends displayed in the data. This study also shows that the information which can be obtained from the GNE depends upon how the information is grouped and analyzed. Summing all the pins of the assembly or assuming a uniform burnup results in the loss of valuable neutron emission information. By splitting the pins into zones, trends such as the total emissions across the assembly become evident. By evaluating individual pins trends such as the effect of water rods on the neutron emission become evident. This type of analysis can be of great use to safeguards. Evaluating the GNE on a pin by pin basis leads to a much clearer picture of the constituents of the assembly.

1. Introduction

According to the Information Circular (INFCIRC) 153 (IAEA), the technical objective of International Nuclear Safeguards is "… the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection." In March of 2009 the Next Generation Safeguards Initiative of the U.S. Department of Energy began a nominal five year spent fuel research effort with the technical goals of: (1) quantifying plutonium content in spent nuclear fuel through the use of non-destructive assay (NDA) techniques; (2) assessing the capability of these NDA techniques to verify burnup, cooling time,

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and initial enrichment; and (3) determining their ability to detect pin diversions in spent fuel assemblies (Galloway et al., 2011). Before the development of any hardware, these NDA techniques were first scoped in computation space to understand detection limitations and down select systems of interest for further testing. Multiple Monte Carlo based spent fuel libraries (SFL) were developed to be used as source terms for assessing the detection limitations of each of these techniques (Fensin et al., 2009; Galloway et al., 2012). In the context of this paper, a Monte Carlo SFL is composed of the pin by pin signatures of a fuel assembly that was either infinitely reflected or burned in an eighth core geometry.

The radioactive nuclides in spent fuel can emit several different types of radiation upon decaying to stable isotopes. The more common radioactive decay mechanisms are by the emission of neutrons, photons, beta particles, and alpha particles. Because the spent fuel is solid, the latter two mechanisms have extremely short path lengths, resulting in minimum direct emission out of the spent fuel assembly, and thus cannot be easily detected. However, neutrons and photons are electrically neutral, resulting in longer path lengths than charged particles. Because of this, these particles can be used as a mechanism for characterizing the nuclide composition. Fission product/activation photons do provide a rich history of information regarding how the fuel was burned (Fensin et al. 2010). The major problem with photon detection is that due to attenuation, only the outer pins of a fuel assembly are seen by detection equipment (Phillips and Bosler, 1994). Furthermore, the background caused by activation of other components in the fuel assembly further complicate the sole use of photons for spent fuel characterization.

The gross neutron signal (GNS) is preferable to other types of radiation signatures such as gamma or X-ray for use in assessing spent fuel assemblies because there is less attenuation of the neutron signal within a fuel assembly, and the signal origin is solely from fuel materials rather than from cladding or other structural materials. Due to the typical structure of the neutron absorption cross section, and the propensity to create charged particles as a function of neutron absorption energy, measuring the neutron spectra is actually quite difficult therefore gross neutron counting is one practical method for examining the spent fuel emission signature. In spent fuel, the GNS has three main components: (α, n), spontaneous fission, and the resulting multiplication. The components of the GNS vary as a function of initial enrichment, burnup, and cooling time because the concentration of the radioactive emitters, resulting in a detectable signature, varies as a function of both the nuclides creation and destruction during burnup, and radioactive decay during cooling time.

Eq. (1) describes the total signal counted in a detector.

$$\mathrm{DS} = S_o e^{-\sigma x} \varepsilon \frac{\Omega}{4\pi} M \tag{1}$$

where DS is the detector signal, S_o is the emission source strength, σ is the attenuation coefficient (absorption cross section), x is the distance from the source to the detector, Ω is the solid angle, ε is the intrinsic efficiency, and M is the contribution from the neutron multiplication. The attenuation term depends on the fuel assembly geometry and the counting geometry (i.e. the signal must first attenuate through the spent fuel assembly, and the spent fuel will then attenuate through the transport geometry between the detector and source such as a pool of water or air). The solid angle term depends on the relative distance between the source to the detector as well as the detector size. The intrinsic efficiency depends on the detector size, material composition, electronics and processing algorithms, and is defined as (Knoll, 2000)

$$\varepsilon_{\text{int}} = \frac{\text{number of pulses recorded}}{\text{number of radiation quanta incident on detector}} \tag{2}$$

In this paper we look at the gross neutron emission (GNE). The GNE is defined as the emission of neutrons from each of the individual pins in an assembly before transport has taken place. Therefore, we are concerned only with characterizing S_o from Eq. (1). This study eliminates the complexity of the terms related to transport in Eq. (1) and focuses on emission so that the work can more easily be applied to other counting geometries (spent fuel reprocessing, counting in water vs. air, using a variety of detectors, etc.).

In this paper we describe the generation and characterization of the GNE for a SFL described in Fensin et al. (2009). The SFL represents a typical Westinghouse pressurized water reactor spent fuel assembly irradiated as part of a model using 1/8 core symmetry and different assembly shuffling patterns (Galloway et al., 2012). It contains state-point information for burnups at 15, 30, and 45 GW d/MTU; cooling times of 1, 5, 20, 40, and 80 years; and an initial enrichment of 4 weight percent. Due to the ability to use detailed continuous energy cross sections for extended burnup calculations and to easily develop models of complex geometries, the SFL was developed using the Monteburns (Trellue and Poston, 1999) burnup code, which links MCNP for transport calculations to an isotope generation and depletion code such as ORIGEN or CINDER90 (CINDER90 was used to generate this library) (Croff, 1980). The fuel assembly of interest was modeled with one radial and one axial region per fuel pin. Because of the 6.67 eV resonance in U-238, resulting in a very short mean free path for thermalizing neutrons, the plutonium production in the outer 100 μm of the fuel pin is about twice that of the rest of the fuel pin, resulting in a plutonium production falloff from the outer radius to the center of the fuel pin. Using only 1 radial region per fuel pin averages this effect. Because the total volume of the outer 100 μm of the fuel pin is only a fractional volume percent of the whole fuel pin (~4.8% assuming a typical PWR fuel pin radius of 4.1 mm), for total neutron counting at burnups less than 70 GW d/MTU, averaging this effect is not an unreasonable approximation (Rondinell and Wiss, 2010). For a fuel pin, with finite axial dimension, and uniform moderator temperature (i.e. uniform moderator density), the power profile is cosine shaped (Lamarsh, 1983). But, if you consider moderator heat up, then the power shape at Beginning of Life looks more like a nose that is peaked towards the bottom of the core. As the reactor burnup increases, Xe will be created faster in the region where the power is peaked, and as a result that region will become less reactive and more power will come from the top of the core. This causes more Xe to buildup at the top of the core and results in a temporal Xe oscillation, which constantly changes the axial power shape as a function of time (Olsson, 1969). Because the power is always changing shape and is never constant, assuming 1 axial region, as opposed to several, is a poorer approximation of the 3-D burnup and isotopic buildup as will be discussed below.

It should be noted that the GNE from spent fuel has been studied extensively for decades and the neutron production mechanisms are well understood (Bosler et al., 1982). Bosler et al. (1982) is one such study describing calculations for examining the GNE and establishing correlations between the GNE, state points of the spent fuel, and fissile content assuming uniform burnup in the radial direction of a PWR. In this paper we expand on these types of studies in two ways: (1) by studying the effect of two different shuffle schemes on the spent fuel emissions (i.e. the effect of the leakage flux); and (2) by studying not only the GNE when averaged over the entire assembly, but also the GNE from four different zones of the assembly, and the GNE of individual pins from different locations in the assembly. The SFL makes (2) possible since the spent