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Passive measurements of mixed-oxide fuel for nuclear nonproliferation

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ABSTRACT

We present new results on passive measurements and simulations of mixed-oxide fuel-pin assemblies. Potential tools for mixed-oxide fuel pin characterization are discussed for future nuclear-nonproliferation applications. Four EJ-309 liquid scintillation detectors coupled with an accurate pulse timing and digital, offline and optimized pulse-shape discrimination method were used. Measurement analysis included pulse-height distributions to distinguish between purely fission neutron sources and alpha-n plus fission neutrons sources. Time-dependent cross-correlation functions were analyzed to measure the fission neutron contribution to the measured sample's neutron source. The use of Monte Carlo particle transport code MCNPX-PoliMi is discussed in conjunction with the measurements.

1. Introduction

The need for advanced measurement techniques to accurately characterize nuclear fuels containing plutonium is increasing in demand as the desire to utilize nuclear power as a reliable energy source increases. In this context, fuel reprocessing and advanced fuel recycling are important topics in the nuclear power industry. Mixed-oxide (MOX) fuels utilize plutonium and uranium that may be separated and recycled from used commercial nuclear reactor fuel. Re-use of both plutonium and uranium in the form of MOX fuels offers a significant increase in the amount of total energy produced from the starting fuel material.

Because mixed-oxide fuel contains a significant amount of ^{240}Pu (strong spontaneous fission source; ~1000 neutrons per second per gram) and alpha-emitting isotopes, a variety of passive neutron measurements are possible. Passive measurement of these forms of special nuclear material (SNM) is a technique that is preferred to active interrogation methods, but still requires much development. The ability to accurately identify SNM benefits areas such as nuclear nonproliferation, nuclear security, safeguards, and materials controls and accountability.

Methods for fuel characterization include the analysis of neutron energy distributions, time-of-flight distributions, cross-correlation functions, and neutron and gamma-ray multiplicity distributions. Measuring MOX fuel pins located at the Idaho National Laboratory (INL) provided the opportunity to develop faster and more robust methods for characterization of SNM, specifically MOX fuel [1]. Passive measurements were performed on a large number of fuel pins (totaling approximately 1 kg of plutonium) with varying isotopic composition. The primary objective of the measurements was to differentiate and characterize specific fuel-pin types based on the analysis of the neutron emissions.

In this paper we discuss the development of various measurement techniques based on the measured results and the ability to simulate these measurements. Monte Carlo simulation results are used to arrive at anticipated results through the use of the MCNPX-PoliMi [2] code.

2. Monte Carlo analysis

Monte Carlo simulations were performed to study methods to characterize MOX fuel and to support the INL measurement campaign described below in detail. These simulations examined MOX fuels and detector response via passive interrogation of materials of interest. This approach allows development of accurate nuclear material characterization schemes, providing detailed insights into the sensitivity of nuclear materials and measurement approaches. The simulations include basic tallied neutron energy distributions, pulse-height distributions (PHDs), and time-correlated particle detections.

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2.1. Description of MCNPX-PoliMi

Monte Carlo simulations of nuclear processes, like the MCNP codes, utilize interaction physics in conjunction with stochastic particle transport. However, standard MCNP (MCNP5/MCNPX) [3,4] does not incorporate accurate time-correlated particle transport required in some SNM-characterization applications. MCNP-PoliMi, originally released from the Radiation Safety Shielding Center at Oak Ridge National Laboratory in 2003, was developed to fill this void. The more recent MCNPX-PoliMi is an enhanced version of the MCNPX code which incorporates methods into the MCNPX code to obtain time-correlated quantities, specifically, the time correlation between neutron interactions and their consequent gamma-ray production. MCNPX-PoliMi utilizes an event-by-event modeling technique that uses analog physics to correctly simulate physical reality within the accuracy of the nuclear data.

Most recent versions of MCNP/MCNPX-PoliMi (version 2.0 is the latest) have the ability of simulating all standard MCNP sources with additional custom sources. Novel sources, including ^{240}Pu and ^{242}Pu spontaneous fission, encompass energy distributions with specific neutron and gamma-ray multiplicity distributions. Additionally, alpha-n distributions are source options available for samples involving plutonium, uranium, and americium isotopes in oxides.

The data outputs from MCNPX-PoliMi simulations include details about each individual interaction that takes place in the detector volume. The data are subsequently post-processed and tailored to simulate a particular detector's response accurately. These simulations of well-defined INL MOX fuel pins provide the information necessary to obtain PHDs and cross-correlation functions.

2.2. Description of the Monte Carlo models

The MCNPX-PoliMi model of the measurement set-up, as shown in Fig. 1, includes four lead-shielded EJ-309 liquid scintillation detectors [5] placed around the axis of the MOX fuel pin set-up (Fig. 2), with each detector equidistant from the source. Each detector is 12.7 cm in diameter and depth and each lead shield is 5 cm thick. Parameters that are adjusted during the simulations include the composition of the fuel pins (two pin types; see Table 1), the distance between the detectors (20, 40, 60, and 80 cm) and the number of pins under investigation (45 and 90 pins). A light-output threshold of 75 keVee (keV electron equivalent) is used in post-processing.

MCNPX-PoliMi provides unique tools to model the spontaneous-fission (SF) and alpha-n (AN) sources that are common in MOX fuel.

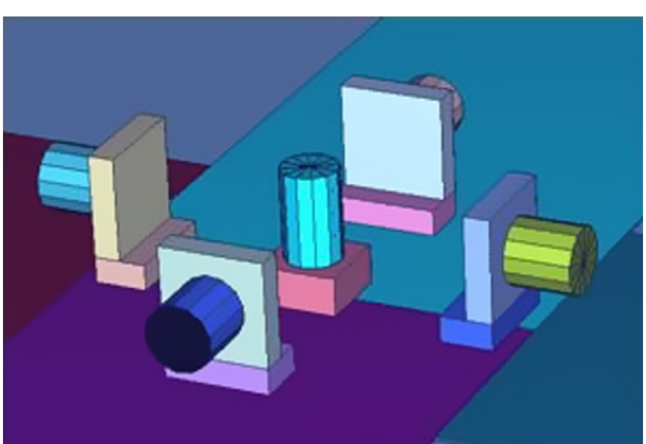

Fig. 1. MCNPX-PoliMi simulation of four cylindrical EJ-309 liquid scintillators surrounding a MOX fuel can. Each detector is shielded by 5 cm of lead. The MOX fuel can is supported by a 7.5-cm thick styrofoam stand.

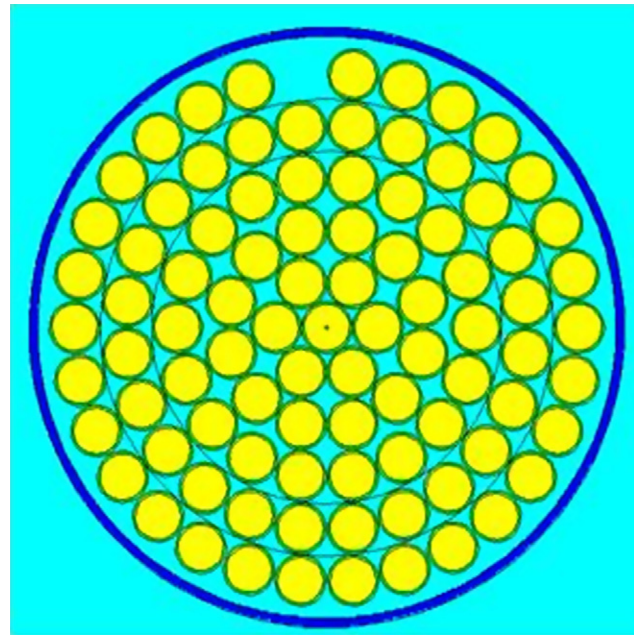

Fig. 2. Cross-sectional view of the 90-pin MOX fuel can where the MOX fuel is modeled within the cladding (stainless steel, 0.5 mm thick) and the pins are contained by an aluminum can.

Table 1
Isotopic composition of MOX fuel pins used for this work at INL (age corrected to June 2009) [6].

Isotope	Pin ♯1 (wt%)	Pin ♯2 (wt%)
^{238}Pu	0.01	0.01
^{239}Pu	11.42	10.98
^{240}Pu	1.53	4.10
^{241}Pu	0.17	0.58
^{242}Pu	0.02	0.02
^{241}Am	0.06	0.16
^{235}U	0.17	0.16
^{238}U	74.78	72.13
O	11.85	11.86

Two well-defined fuel-pin types were available for measurement at the INL. Both pin types are composed of various uranium, plutonium, and oxygen isotopes. The primary difference in the materials is the mass of the ^{240}Pu isotope. This detail is significant as ^{240}Pu is the primary neutron contributor for this type of advanced fuel. Table 1 shows the composition of the INL fuel pins. Fig. 3 shows what sources were simulated and their contributions to the total neutron production rate. The SF of ^{238}Pu ($\sim$75% of the neutron production rate of the SF of ^{242}Pu in the pins), as well as the AN sources ^{235}U, ^{238}U, and ^{241}Pu, was neglected. The total neutron multiplication values for the two fuel-pin assemblies were 1.1264 and 1.1350.

2.3. Initial simulation results

Fig. 4a shows the simulated energy distributions of the neutrons emitted from the two 90-pin MOX fuel assemblies. The valleys located in the lower energy region of both spectra, specifically near 0.05, 0.1, and 1.3 MeV, are due to resonances in the neutron elastic scattering cross-section of ^{16}O, which is also shown in Fig. 4(a) [7]. Fig. 4(b) shows the individual neutron source contributions (SF and AN) to the total neutron energy distribution of the Pin ♯1 MOX fuel assembly.

Currently, the measurement of neutron energy distributions of fissile materials is an area of much needed development. If the