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Plutonium fingerprinting in nuclear forensics of spent nuclear fuel

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ABSTRACT

A fingerprinting procedure, based on the direct use of Pu isotopic ratios, is presented to discriminate spent fuels of different origin. Simulated and real spent fuels from PWR, BWR and LMFBR have been used, with different charge compositions in uranium, plutonium and minor actinides. The sensitivity of the procedure was sufficient to resolve the spent fuels from the same reactor type but of different enrichment and of the same enrichment from different reactor types. A dependence on the ^{234}U and ^{236}U charge composition is observed in the simulated fuels. The fingerprinting methodology is not influenced by the cooling time of the spent fuel and the Pu age. The discrimination of the actual spent fuel pins, although of the same enrichment and from the same assembly, indicates a dependence of the procedure on the pin location within the assembly.

1. Introduction

Nuclear forensics have emerged and developed over the last two decades following the smuggling of nuclear material over the borders of different countries. Prompt response to intercept such material, preventing its misuse, and establishing its route are the main tasks in nuclear forensics (Smith et al., 2008; Mayer et al., 2013; Kristo and Tumey, 2013; Fedchenko, 2014). If the smuggled material is spent fuel, its origin depends on its charge composition, the type of the reactor where it was charged and its final burnup. Fingerprinting methods have been demonstrated to resolve spent fuels of different origin in a 3D space (Nicolaou, 2006 & 2008; Robel and Kristo, 2008; Jones et al., 2014). They have combined the U, Pu and fission product isotopes present in simulated or actual spent nuclear fuels and multivariate statistical/pattern recognition techniques. Resolving spent nuclear fuels of known origins is essential in order to identify the origin of a seized one since it would cluster with the known fuel of the same origin.

This paper explores a fingerprinting approach, based on the direct use of 3 types of Pu isotopes ratios, without employing statistical techniques. The ratios reflect the isotopes present in the fresh fuel and the neutron flux and spectrum in the reactor where the fuel was charged. Then, the ratios are directly plotted as coordinates on a 3D space to resolve spent fuels originating from different reactors. The scope of the paper is to assess: (1) the potential of the methodology towards discriminating spent fuels of different origin based on the three Pu ratios; simulated commercial and emerging spent nuclear fuels and actual commercial ones have been considered; (2) any dependences of the methodology on the inclusion of ^{234}U and ^{236}U in the charge composition for the simulated cases; (3) the influence of the spent fuel cooling time and the Pu age on the resolving capabilities of the methodology; and (4) any dependence of the procedure on the location of a pin within a core, using the actual commercial fuel pins.

2. Materials and methods

The composition of a discharged irradiated nuclear fuel depends on the isotopes present in the fresh fuel and its irradiation history in a reactor, hence, uniquely reflecting the origin of the fuel. Plutonium isotopes are built up and depleted through successive neutron capture reactions on ^{235}U and ^{238}U and radioactive decay of the reaction products. The higher ^{235}U enrichment enhances the buildup of ^{238}Pu, while hardening of the spectrum of the neutrons in the core and higher neutron fluxes would lead to lower concentration of the higher Pu isotopes and higher amount of ^{240}Pu relative to ^{239}Pu respectively. Hence, the ratios ($^{242}Pu/^{240}Pu$), ($^{238}Pu/Pu_{Total}$), ($^{239}Pu/^{240}Pu$) are used as fingerprints to discriminate spent fuel pins of different provenance. The procedure was assessed using simulated conceptual spent nuclear fuels from Pressurised Water Reactor (PWR), Boiling Water Reactor (BWR) and Liquid Metal Fast Breeder Reactor (LMFBR) and samples from real

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commercial PWR spent fuels.

In the case of the conceptual spent fuels, these were simulated for the following cases of reactors and commercial and emerging fresh fuel compositions (Koch and Nicolaou, 1993; Salvatores et al., 2015):

- *Commercial fuels:* PWR-U (enriched in ^{235}U to 2.75–4%); BWR-U (enriched in ^{235}U to 3–4%); PWR-MOX (UPu); LMFBR-MOX (UPu).
- *Emerging fuels:*
  - *Transmutation fuels:* fresh fuels containing either 2% ^{237}Np or ^{241}Am have been considered, resembling their homogeneous transmutation in the same reactor and under the same operating conditions as in the commercial LMFBR-MOX.
  - *Fuel with plutonium and minor actinides* from self-generated recycling in an LMFBR.
  - *Fuels with plutonium or plutonium and minor actinides* from self-generated recycling in a PWR.

Fresh fuels were considered to contain 300 and 600 mg kg^{-1} ^{234}U and ^{236}U respectively, which are at the upper end of their concentrations in commercial fuels. In all cases, the Pu isotopes, required for the isotopic ratios, have been simulated for spent fuels with burnup values in the range of 25–45 GWd/tU (PWR & BWR cases) and 60–100 GWd/tU (LMFBR cases). First generation plutonium from the PWR-U (3.5% ^{235}U), with an isotopic vector for 238/239/240/241/242 of 3.1/49.1/23.66/15.5/8.64, was used in the commercial PWR-MOX and commercial and transmutation LMFBR-MOX fuels. In the self-generated recycling schemes, the Pu and minor actinide (MA) composition of the fresh fuels are considered after the 5th recycling step (PWR cases) or the 16th recycling step (LMFBR case). These steps correspond to the periods necessary in order that an 'equilibrium' content of the recycled nuclides is achieved (Koch and Nicolaou, 1993). At this stage, the amounts of nuclides being formed and transmuted during a fuel cycle are the same.

The Pu and MA used in the fresh fuel for their recycling and transmutation schemes have been used 6 months after being retrieved from reprocessing carried out on spent fuel 1 y after the Enf-of-Irradiation (EOI). The simulations were carried out using the 0D isotope generation and depletion code ORIGEN-ARP. This is a code within SCALE 6.1 (ORNL/TM-2005/39, 2011), and ORIGEN-2.2 (Croff, 1983) employing nuclear data from the ENDF/B-VI (Rose, 1991) and ENDF/B- V (ENDF/B-V, 1979) libraries respectively.

Furthermore, samples of different burnup, from real spent fuel pins from PWR and BWR nuclear power stations, have been considered. Composition and burnup information of these samples have been drawn from the OECD/NEA- SFCOMPO databank. The cases considered were: samples from four pins (pin1-4), occupying different positions within a 14 × 14 square assembly from the Obrigheim PWR, having 3% ^{235}U enrichment and burnup in the range of 23–37 GWd/tU; samples from a pin from the Calvert Cliffs-1 PWR, with 3.038% ^{235}U enrichment and burnup values of 27–45 GWd/tU; and, samples from two pins from different assemblies in the Gundremmingen BWR, having 2.53% ^{235}U enrichment and burnup 15–20 GWd/tU (pin 1) and 20–25 GWd/tU (pin 2).

3. Results

3.1. Simulated conceptual spent fuel cases

The methodology, based on the three Pu ratios, is sensitive enough to discriminate the simulated spent fuels on the basis of their reactor type and charge composition (Fig. 1). Each curve corresponds to a spent fuel, having a particular charge composition

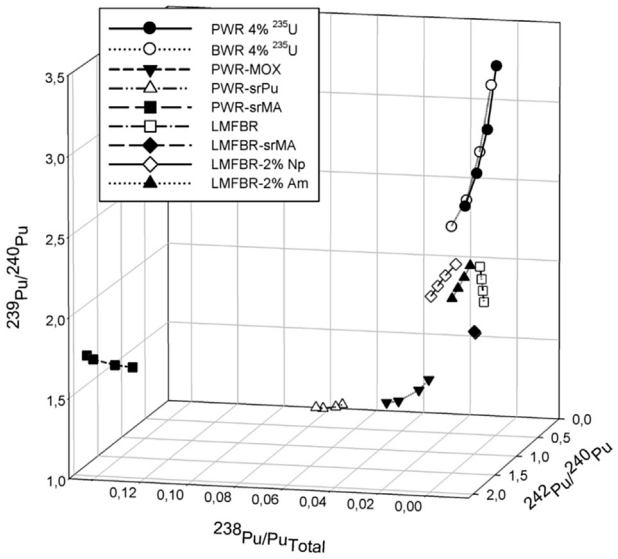

Fig. 1. Discrimination of conceptual simulated commercial, transmutation and recycling spent fuels.

and originating from a particular reactor. Along a curve, each point corresponds to the particular spent fuel having a different burnup. Three distinct groups of spent fuels are identified: the PWR- and BWR-U, the LMFBR cases and the three PWR recycling schemes. Within the first group, the PWR- and BWR-U 4% ^{235}U cases, although with the same charge composition, are discriminated between them on the basis of the different reactor type. In the second group, the commercial FR case is resolved from the transmutation and self-recycling cases on the basis of the plutonium and other actinides present in the fresh fuel composition. Similarly, in the third group the PWR-MOX and self-recycling schemes are resolved between them on the basis of their charge composition in Pu and other actinides.

The sensitivity of the methodology is further investigated for different enrichments of the PWR- and BWR-U cases. Hence, it is aimed to assess the potential of the procedure to differentiate spent fuels of: (1) different enrichments from the same reactor type PWR or BWR; and, (2) the same enrichment from the PWR and BWR. On the basis of the three Pu ratios considered, the simulated

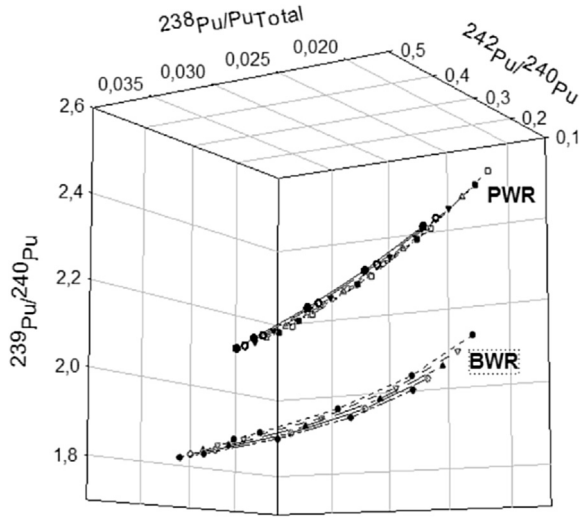

Fig. 2. Discriminating spent fuels of different enrichments from PWR-U and BWR-U.