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Technical note

Discrimination of spent nuclear fuels in nuclear forensics through isotopic fingerprinting

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ABSTRACT

Isotopic fingerprinting in nuclear forensics, developed through a simulation study to resolve spent nuclear fuels from different reactors, has been applied on real samples from fuels irradiated in nuclear reactors. The U and Pu isotopics of the real samples, used as fingerprints, have been retrieved from the OECD/NEA SFCOMPO databank comprising compositions of spent nuclear fuels from their Post-Irradiation Examination at the End of their Irradiation (EOI). The method has grouped together nuclear spent fuels from the same reactor, resolving distinctly those with similar ^{235}U enrichment. Furthermore, spent fuel pins of the same enrichment, from different positions within a reactor core, are resolved. Differentiating of the spent fuels has been achieved, whether (U, Pu) or Pu are used as fingerprints.

1. Introduction

Illicit trafficking of nuclear materials, and the consequences on humans in case of their misuse, has led to the development of nuclear forensics in the area of nuclear security. Nuclear forensics focuses on the timely interception of illicit nuclear material to prevent their misuse and determine the trafficking route from its diversion to interception (Smith et al., 2008; Kristo and Tumey, 2013; Fedchenko, 2014). Identification of such a material, with respect to its provenance, is a key step toward combating their illicit trafficking. Hence, pure simulation fingerprinting studies were developed and reported, classifying conceptual spent nuclear fuels according to their charge composition, the reactor from which they originated and the final burnup attained (Nicolaou, 2006; Nicolaou, 2008; Robel and Kristo, 2008). The U and Pu isotopics of the spent fuels, simulated through the depletion 0-D code ORIGEN (Croff, 1983), were used as the appropriate fingerprints.

In this work, the potentiality of the fingerprinting method is tested on real spent nuclear fuel samples from existing power reactors. The required fingerprints have been obtained from the OECD/NEA SFCOMPO databank comprising isotopic compositions from the Post-Irradiation Examination (PIE) of UO_2 commercial reactor spent fuels (http://www.oecdnea.org/sfcompo/Ver.2/Eng/index.html). Furthermore, due to the availability of pins from different positions within an assembly or different assemblies, any dependence of the method on the pin location within the core is sought. This issue could not be tackled previously, since U and Pu compositions had been obtained through simulations with a 0-D code.

2. Materials and methods

The methodology relies on the dependence of the composition of spent nuclear fuel on the charge composition and the irradiation of the fuel in the reactor. Therefore, the composition can uniquely reflect the origin of the fuel. The isotopic compositions, used in the form of isotopic ratios, were in the case of (U, Pu) or Pu: $^{242}Pu/^{240}Pu$, $^{238}Pu/(\text{total Pu})$, $^{235}U/^{238}U$, $^{240}Pu/^{239}Pu$, $^{241}Pu/^{240}Pu$, $^{239}Pu/^{235}U$, $^{242}Pu/^{238}U$. The most similar ratios of the different spent fuels are sought, through their comparison using factor analysis (Everitt and Dunn, 1991; Nicolaou, 2006), allowing clustering of the spent nuclear fuels with the same origin. The spent nuclear fuels used in the study are shown in Tables 1 and 2 for the PWR and BWR considered. The reactors had been charged with fresh fuels based on UO_2 and UO_2 with Gd_2O_3 as a burnable absorber. Post-Irradiation Examinations on the spent fuels from the reactors have provided the required compositions compiled in the SFCOMPO data bank by JAERI and disseminated by OECD/NEA (Naito et al., 1993). The U and Pu isotopic compositions of the spent fuels within the SFCOMPO data bank are quoted at the End of their Irradiation (EOI).

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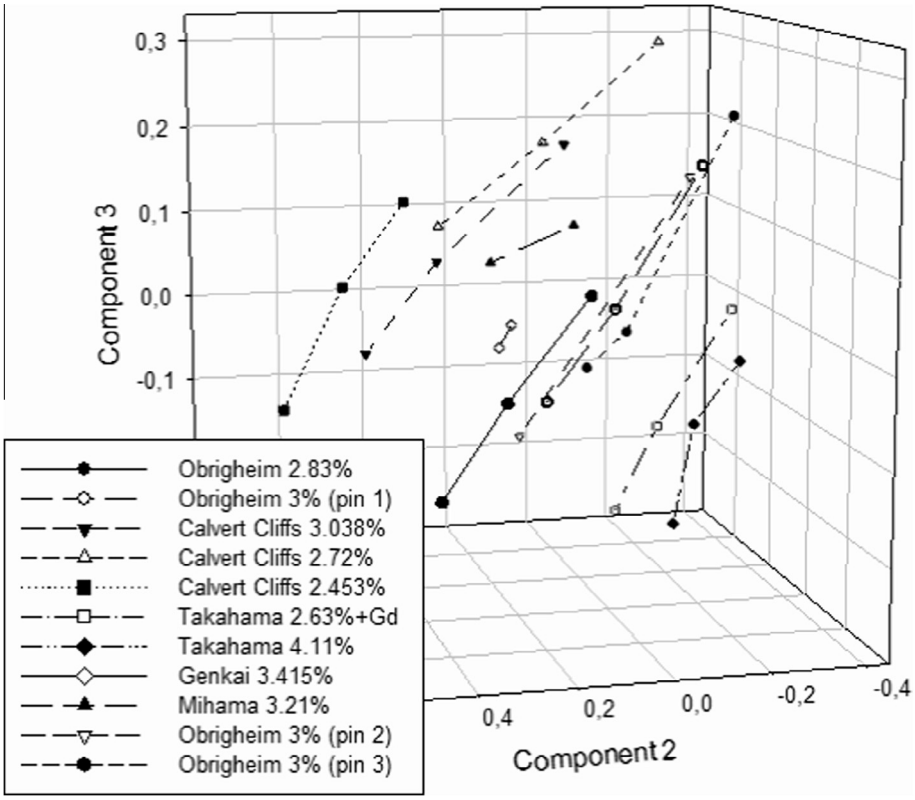

Fig. 1. Clustering of the PWR spent fuel cases considered, based on their (U, Pu) composition.

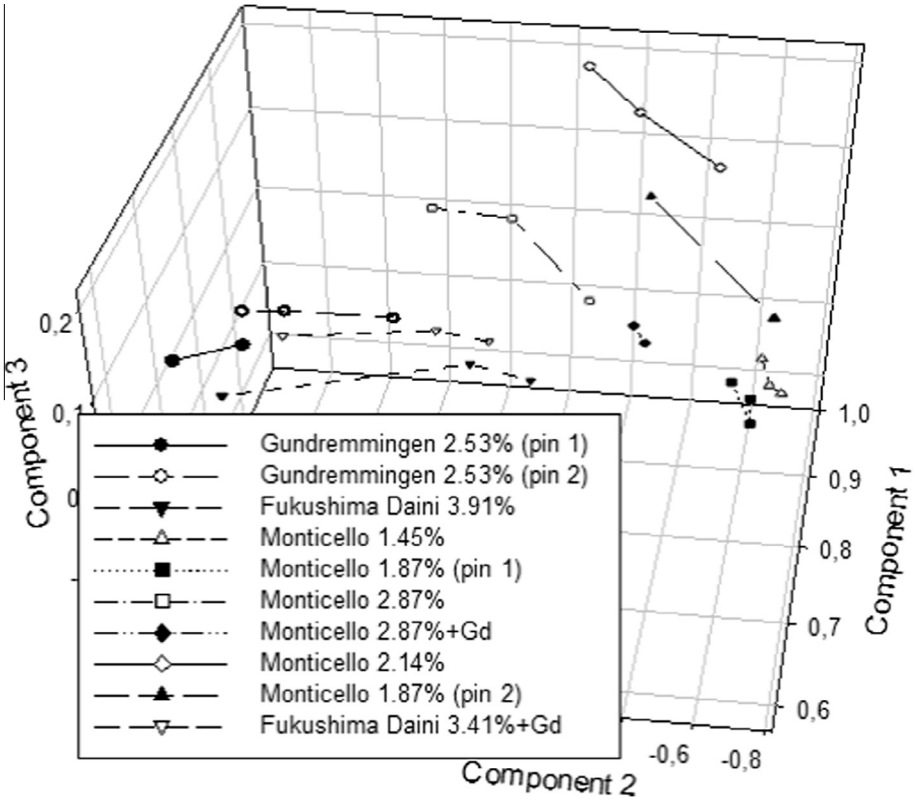

Fig. 2. Clustering of the BWR spent fuel cases considered, based on their (U, Pu) composition.