



Short communication

Provenance of unknown plutonium material

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ABSTRACT

The determination of the provenance of 'unknown' plutonium material is demonstrated through a simulation study based on an isotopic fingerprinting approach. Plutonium of known provenance was considered as the 'unknown' nuclear material in order to evaluate the potential of the approach and verify its predictive capabilities. Factor analysis was used to compare the Pu isotopic composition of the 'unknown' material with Pu isotopic compositions simulating well known spent fuels from a range of commercial nuclear power stations. The provenance of the 'unknown material' is assigned to the commercial fuel with which exhibits the highest degree of similarity with respect to the Pu composition. The approach appears promising since it accurately predicted the provenance of the one 'unknown' sample considered; nevertheless, the approach is still at the development stage. Important challenging issues related to the simulation uncertainties and its testing on real laboratory samples have to be explored prior to evaluating the potential of the approach.

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1. Introduction

Over the last decade, illicit handling of nuclear material has become an issue of the highest concern (International Atomic Energy Agency, 2006). This stems from the potential of such material to cause human and environmental damage due to its explosive and radiotoxic nature. Hence, nuclear forensics has emerged with the primary objective of detecting illicit handling of nuclear material and the determination of its provenance and intended use (International Atomic Energy Agency, 2002a,b,c; Duftschmid, 2002; Morgenstern et al., 2002).

Plutonium (Pu) from reprocessed spent nuclear fuel is a material of potential illicit use due to its radiotoxic and fissile properties. In the context of nuclear forensics, the material could be found as trace amounts or in a bulky form. In the former, it appears as particles in environmental samples indicating possible clandestine production of fissile material (Tamborini et al., 2002; Ranebo et al., 2007). The latter case refers to material found undeclared in non-designated locations within the fuel cycle indicating possible illicit trafficking of the material (Ray et al., 2002).

The objective of this work is to demonstrate an approach, based on isotopic fingerprinting (Nicolaou, 2006), that can determine the provenance of unknown plutonium material. The term provenance in this work refers to the fuel type from which the plutonium was

retrieved during reprocessing, the reactor type in which the fuel had been irradiated and its final burnup characteristics.

2. Materials and methods

2.1. Simulations

The approach is based on the inherent consistency of the content of individual radionuclides in spent nuclear fuel, as a result of the fresh fuel composition, reactor neutron spectrum and history of irradiation of the fuel. The content carries information which can be uniquely related back to the provenance of the fuel. Hence, the measurement of selected radionuclides could allow the identification of fuel in terms of its provenance. The determination of the provenance of unknown plutonium, retrieved through reprocessing from the spent fuel, would be based on its isotopic composition with respect to ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{242}Pu .

The approach compares the Pu isotopic compositions of an unknown plutonium material and commercial spent nuclear fuels representing a wide range of nuclear power stations. The commercial fuels have a known provenance, namely fresh composition, reactor type in which they were irradiated and irradiation history. The comparison seeks to identify the commercial fuel with the highest similarity in Pu composition to the unknown material. Then, the provenance of the unknown material can be identified in terms of the provenance of its most similar commercial fuel.

A data bank required for the comparisons was created, comprising Pu isotopic compositions in a wide range of commercial spent nuclear fuels from different nuclear power reactors. The lack of sufficient experimental data on Pu isotopic compositions was compensated through their calculation using the zero-dimensional depletion computer code ORIGEN-2 (Croff, 1983). The simulations predict changes taking place in the fuel composition as a result of irradiation in a reactor neutron spectrum during power operation. In this respect, the generation of the plutonium isotopes is significantly affected by the neutron spectrum, the fresh fuel content of ^{235}U and ^{238}U , the burnup of the fuel at the end of irradiation (EOI) and cooling time of the spent fuel. The calculations performed for this study were coupled with burnup-dependent cross-section libraries which resembled, as closely as possible, the reactor – fuel combinations under consideration.

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Plutonium isotopic compositions were simulated for a range of commercial fresh fuels (Table 1). These included Uranium Oxide (UO₂) fuels in Pressurized Water Reactor (PWR), Boiling Water Reactor (BWR) and Canadian Deuterium Uranium (CANDU) power stations, and Mixed Oxide (MOX) fuels in PWR and the fast reactor Liquid Metal Fast Breeder Reactor (LMFBR). In each case, the simulations were carried out for a range of burnup values, while isotopic compositions at EOI were considered in the data bank. In order to demonstrate the approach, a spent fuel with 42 GWd/tU burnup, from the simulated PWR commercial UO₂ spent fuels, was chosen to be the 'unknown' material, whose origin is sought.

2.2. Statistical approach

The multivariate statistical technique of factor analysis was used for the required comparisons of the 'unknown' with the fuels in the data bank. This pattern recognition technique groups together objects in such a way that similarities and differences between them can be revealed in a graphical presentation (Everitt and Dunn, 1991). In general, the objects may be characterised by N different parameters, such as shape, size, mass, porosity of material, mechanical properties, Pu isotopes. Factor analysis groups together objects on the basis of their similarity with respect to these parameters (Everitt and Dunn, 1991). Furthermore, the N coordinates of each object in the N -dimensional space are combined and reduced into three dimensionless coordinates termed 'components', resulting in a projection of the objects onto 3D space which is easy to interpret.

In this study, the objects considered were the different Pu materials retrieved during reprocessing from the spent fuels described in Table 1. The N different characteristic parameters of these objects were their simulated compositions of ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu and ²⁴²Pu in the spent fuels given in Table 1. The materials can be grouped together by factor analysis in such a way that patterns of similarities between the simulated and the unknown fuel can be uncovered. Then, the fuel-reactor combination, i.e. the provenance of the unknown Pu material, can be identified as the one which provides the closest grouping to the simulated fuel in the 3D space on the basis of their similar Pu compositions.

3. Results and discussion

The 3D plot from the factor analysis of the seven simulated spent fuels described in Table 1 is shown in Fig. 1. The plot shows them clustered in distinct groups resolved from each other on the basis of the different reactor-fuel combinations considered. The approach is sensitive enough to resolve groups of the same reactor on the basis of their fresh fuel type. Hence, PWR-U and PWR-MOX are well resolved, as well as the groups CANDU-N and CANDU-S (Fig. 1). Furthermore, the three thermal reactor U fuels, namely PWR-3.5%U, PWR-4.15%U and BWR-U are well separated from each other when a magnification of their region in Fig. 1 is plotted (Fig. 2). Fuels of similar enrichment, irradiated in neutron spectra of different hardness, i.e. PWR-3.5%U and BWR-3.4%U, are also well separated (Fig. 2). The ability to resolve the fuels stems from the evolution of their composition due to the transmutation mechanisms of nuclear reactions $[(n, \gamma)$ and $(n, 2n)]$ and decay $[\alpha$ and β]. The plot is influenced by the combined effect of reaction rate, i.e. neutron flux and target nuclei, the neutron spectrum and the irradiation time of the fuel in the reactor.

The provenance of the 'unknown' fuel, namely its fresh composition, the reactor type in which it was irradiated and its final burnup, can now be deduced from its clustering in relation to the simulated commercial fuels. The 'unknown' material is clustered correctly with the simulated PWR-3.5%U spent fuel (Fig. 2), indicating its provenance. Therefore, the approach is sensitive

Table 1
Simulated spent fuels for different reactors, fresh fuel compositions and targeted burnup

Reactor	Fuel characteristics	Burnup range (GWd/tU)
PWR-U	UO ₂ , 3.5% ²³⁵ U	40–55
PWR-U	UO ₂ , 4.15% ²³⁵ U	40–60
PWR-MOX	Thermal MOX [natural U, 3.4% fissile Pu]	40–50
BWR-U	UO ₂ , 3.4% ²³⁵ U	20–40
CANDU-N	Natural U	4–9
CANDU-U	Slightly enriched UO ₂ , 2% ²³⁵ U	14–30
LMFBR	MOX [80%U depleted, 12% fissile Pu]	60–100

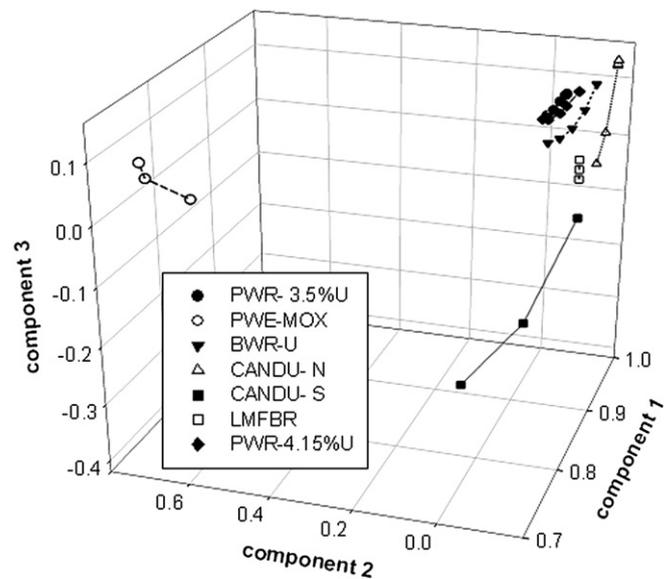


Fig. 1. 3D plots from the factor analysis of the simulated fuels showing their similarities.

enough to cluster the 'unknown' fuel only with the correct type of fuel, while resolving it well from the other ones.

The approach has been demonstrated on the basis of the Pu composition at EOI for both the simulated unknown and commercial spent nuclear fuels. However, in an actual forensic situation, the EOI and time of Pu retrieval from the reprocessing of the spent fuel would be unknown. The following transitions affect the Pu composition in spent nuclear fuel during decay after EOI: depletion of ²³⁸Pu due to its α -decay (half-life $\tau_{1/2} = 87.75$ y) to ²³⁴U; depletion of ²⁴¹Pu due to its β^- -decay ($\tau_{1/2} = 14.9$ y) to ²⁴¹Am; buildup of ²³⁸Pu from α -decay of ²⁴²Cm ($\tau_{1/2} = 163$ d) and buildup of ²⁴⁰Pu from α -decay of ²⁴⁴Cm ($\tau_{1/2} = 18.1$ y). Following reprocessing, the Pu isotopic composition is influenced only by the decay of ²³⁸Pu and ²⁴¹Pu.

The effect of cooling time of the 'unknown' material on the prediction of its provenance has been investigated. Plutonium isotopic compositions of the 'unknown' material corresponding to

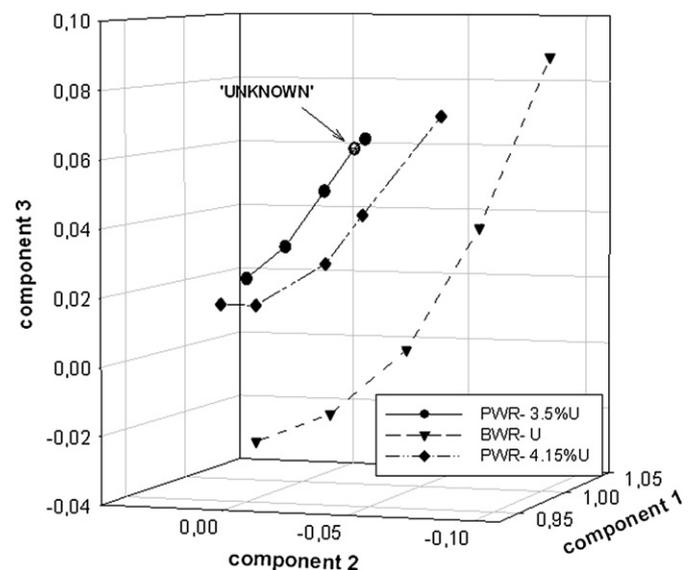


Fig. 2. 3D plots from the factor analysis showing the similarities between the simulated PWR-U, BWR-U, LMFBR and the 'unknown'.

cooling times up to 5 y were compared with the data bank comprising the compositions of the commercial spent fuels at EOI. It was found that, for a composition of the 'unknown' material corresponding to a cooling time beyond 0.5 y, its provenance was wrongly predicted as that of the PWR-4.15%U. Hence, the data bank has to be extended in order to include Pu compositions at different cooling times. Alternatively, the 'age' of the plutonium has to be determined through parent/decay products, such as $^{238}\text{Pu}/^{234}\text{U}$, $^{241}\text{Pu}/^{241}\text{Am}$, allowing the composition of the 'unknown' to be corrected to the one at EOI (Wallenius et al., 2001).

4. Conclusions

An isotopic fingerprinting approach is presented as a possible tool to determine the provenance of an 'unknown' seized nuclear material in the context of nuclear forensics. Plutonium of known provenance was considered as the 'unknown' nuclear material in a simulation study in order to perform a preliminary evaluation of the potential of the approach. The sensitivity of the approach was sufficient to resolve spent fuels of similar enrichment but different neutron spectrum hardness, as well as spent fuels from the same reactor but of different initial enrichments. The approach accurately predicted the provenance of the only one 'unknown' plutonium material considered.

Although the approach appears promising, it is only at the initial stages of its development. Therefore, prior to fully assessing its potential and limitations, the approach should be exploited from the viewpoint of different issues that may well affect its predictive capabilities. Such issues include the uncertainty of the composition simulations carried out by ORIGEN; the dependence of the composition on the axial or radial position within a fuel rod, as well as the position of the rod within the core, the position of the rod in the core relevant to control rods which would affect the neutron spectrum; hence, the necessity of using 3D codes instead of ORIGEN should be explored; the accuracy of the nuclear data used in the simulations; the effect of the different designs of the same type of commercial reactors; and, the accuracy of the chemical analyses

available to determine the composition of seized unknown materials. Finally, the approach should be tested with well known plutonium samples which might be available in hot-cells laboratories, in order to assess its potential under conditions as close to a real situation as possible.

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