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Determination of the origin of unknown irradiated nuclear fuel

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Abstract

An isotopic fingerprinting method is presented to determine the origin of unknown nuclear material with forensic importance. Spent nuclear fuel of known origin has been considered as the 'unknown' nuclear material in order to demonstrate the method and verify its prediction capabilities. The method compares, using factor analysis, the measured U, Pu isotopic compositions of the 'unknown' material with U, Pu isotopic compositions simulating well known spent fuels from a range of commercial nuclear power stations. Then, the 'unknown' fuel has the same origin as the commercial fuel with which it exhibits the highest similarity in U, Pu compositions.

Keywords: Nuclear forensic; Isotopic fingerprinting; Multivariate statistics

1. Introduction

During the last decade, nuclear forensic has emerged from the concern over the illicit use of nuclear material and is primarily occupied in the determination of the nature of a seized material, its intended use and origin (Duftschmid, 2002). The concern stems from the human and environmental consequences that may rise because of the explosive and radiotoxicity potential

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of such material. As a result, particular attention to nuclear forensic was given by the scientific and political communities, both at national and international levels (International Atomic Energy Agency, 2002a,b,c), with the introduction of appropriate technical and legislative measures towards combating the illicit use of nuclear material.

Irradiated nuclear fuel is a material of potential illicit use. However, handling difficulties due to high radiation doses render it less attractive for criminal use than fresh fuel. Nevertheless, material containing U and Pu from reprocessed irradiated fuel can be easily handled and hence particularly attractive for illicit use. Irradiated fuel or material containing U and Pu could be found undeclared in non-designated locations in the fuel cycle as bulky samples in the case of illicit trafficking. Alternatively, they can be traced as particles at the microscopic level in environmental samples, due to operational or accidental releases from nuclear installations and possible clandestine production of fissionable nuclear material.

The origin determination of unknown seized nuclear material is an important and challenging task in nuclear forensic. An isotopic fingerprinting method aiming to tackle this task is presented in this work for the case of unknown irradiated nuclear fuel, either in bulk or particle form. The origin is determined in terms of the fuel type and its fresh composition, the reactor where it was irradiated and its final burnup. The method is based on the U, Pu isotopic compositions of the unknown irradiated nuclear fuel.

2. Materials and methods

The determination of the origin is based on some measurable characteristics, namely the U and Pu isotopic compositions of the irradiated nuclear fuel. The method is based on the fact that the composition of irradiated nuclear fuel is inherently consistent and depends on the fresh fuel composition and its irradiation history. This composition carries therefore information, which can be uniquely related back to the origin of a fuel.

The method compares the U, Pu isotopic compositions of the unknown material with commercial spent nuclear fuels from a wide range of nuclear power stations. The commercial fuels have known characteristics, namely fresh composition, reactor type where they were irradiated and irradiation history. The comparison seeks the commercial fuel with the highest similarity in U, Pu compositions to the unknown material. Then the origin of the unknown material is identified in terms of the characteristics of its most similar commercial fuel. The multivariate statistical technique of factor analysis (Everitt and Dunn, 1991) is used for the comparison purposes.

The isotopic compositions of U and Pu, namely ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu, are considered as the measurable characteristic parameter for the comparison purposes. The choice of these nuclides is twofold:

1. the evolution of these nuclides during irradiation depends on, and hence reflects, the sought information, which is the type of fuel and reactor where the fuel was irradiated;
2. they can be measured in the unknown material through highly accurate destructive chemical analysis, such as Isotope Dilution Thermal Ionisation Mass Spectrometry (IDMS) (Koch et al., 1989).

2.1. Experimental

Known spent nuclear fuels were assumed to be the ‘unknown’ nuclear material whose origin is sought. Parameters such as initial enrichment, irradiation history, final burnup and the U, Pu isotopic compositions of these fuels were well known (Guardini and Guzzi, 1982). However, for the purpose of this work, these

fuels are assumed to be the 'unknown' material whose origin is sought through the determination of these parameters. These fuels, from PWR nuclear power stations, were as follows: UO_2 [3% enriched in ^{235}U, declared burnup 37 GWd/tU ('UNKNOWN' 1); 3.2% enriched in ^{235}U, declared burnup 32 GWd/tU ('UNKNOWN' 2); 3.2% enriched in ^{235}U, declared burnup 38 GWd/tU ('UNKNOWN' 4)]; and, MOX [4% enriched in fissile Pu, declared burnup 44 GWd/tU ('UNKNOWN' 3)].

2.2. Theoretical

A data bank was created with the U, Pu isotopic compositions covering a wide range of commercially spent nuclear fuels from different nuclear power reactors. In the absence of sufficient experimental data on U, Pu isotopic compositions, these were calculated using the zero-dimensional depletion computer code ORIGEN-2 (Croff, 1983). The simulations were based on reactor physics equations and predict the changes that take place in the composition of the spent fuel during irradiation in a neutron spectrum and cooling after the end of irradiation [EOI]. The simulations were coupled with burnup-dependent cross-section libraries resembling the reactor–fuel combination, and hence the hardness of the neutron spectrum, where the fuel was irradiated.

Simulations were performed for a range of fuels with different fresh compositions, irradiated in different types of reactors for a wide range of burnup values. They included the cases of PWR (UO_2 and MOX fuels), BWR (UO_2 fuel), CANDU (U_N – natural uranium and U_S – slightly enriched uranium fuels) and the fast reactor LMFBR. In each case, the calculations were performed for a range of burnup values that could be encountered in a fuel cycle. Table 1 lists the fuels, reactors and the range of burnup values considered in the simulations.

The required comparisons were performed using the qualitative multivariate statistical technique of factor analysis. This is a pattern recognition technique, in that it presents graphically objects grouped together in a way that patterns of similarities and differences between them can be uncovered. Each object is characterised by a number of different parameters *N*, for example mass, size, shape, isotopic composition, porosity of material, mechanical properties. Then, the objects are grouped together on the basis of the similarity of these parameters. Essentially, factor analysis is a dimension reduction technique in that, the *N*-dimensional space onto which objects are projected is reduced to a 3-dimensional (3D) space (Everitt and Dunn, 1991). The *N* coordinates of each object in the *N*-dimensional space are combined and reduced into three dimensionless coordinates termed 'components'. Hence, a projection of the objects onto the easier to interpret 3D space can be achieved.

In the present study, objects are the simulated and the four 'unknown' spent fuels (Tables 1 and 2). The characteristic parameters of these objects are their isotopic compositions of U and Pu as stated in Chapter 2. Then the fuels are grouped together by factor analysis in a way that patterns of similarities between the simulated and the 'unknown' fuels can be uncovered. The origin of an 'unknown' fuel would then be the one of the simulated fuel with which it is grouped together in the 3D space on the basis of their similar U and Pu compositions.

Table 1
Simulated reactors, fuels and targeted burnup values

Reactor	Fuel	Burnup range (GWd/tU)
PWR	UO_2, 3% ^{235}U	25–45
PWR	UO_2, 3.2% ^{235}U	25–40
PWR	Thermal MOX [natural U, 4% fissile Pu]	35–50
BWR	UO_2, 3.4% ^{235}U	15–25
CANDU	Natural U	0.3–8
CANDU	Slightly enriched UO_2, 1.2% ^{235}U	5–20
LMFBR	MOX [80% U depleted, 12% fissile Pu]	50–100

Table 2
Predicted and declared fuel characteristics

'UNKNOWN' fuels	Predicted parameters	Declared parameters
'UNKNOWN' 1	PWR-UO_2, 3% ^{235}U 35 GWd/tU	PWR-UO_2, 3% ^{235}U 37 GWd/tU
'UNKNOWN' 2	PWR-UO_2, 3.2% ^{235}U 35 GWd/tU	PWR-UO_2, 3.2% ^{235}U 32 GWd/tU
'UNKNOWN' 3	PWR-MOX 44 GWd/tU	PWR-MOX 45 GWd/tU
'UNKNOWN' 4	PWR-UO_2, 3.2% ^{235}U 38 GWd/tU	PWR-UO_2, 3.2% ^{235}U 40 GWd/tU

3. Results and discussion

The factor analysis results are shown in Fig. 1. The 3D plot shows patterns of similarities between the simulated spent fuels, which are exploited from the following view points: clustering of the different fuels and the effects of the fresh fuel composition, origin determination of the 'unknown' fuels. Furthermore, the effects of cooling time and accuracy of chemical analysis of the 'unknown' material on its origin determination are studied.

The 3D plot shows the seven simulated fuel–reactor combinations forming distinct groups well resolved from each other. The method is sensitive enough to resolve groups of the same reactor type, on the basis of their fuel fresh composition. This is in accordance with the changes in the fuel composition during irradiation which are a function of the fresh fuel composition

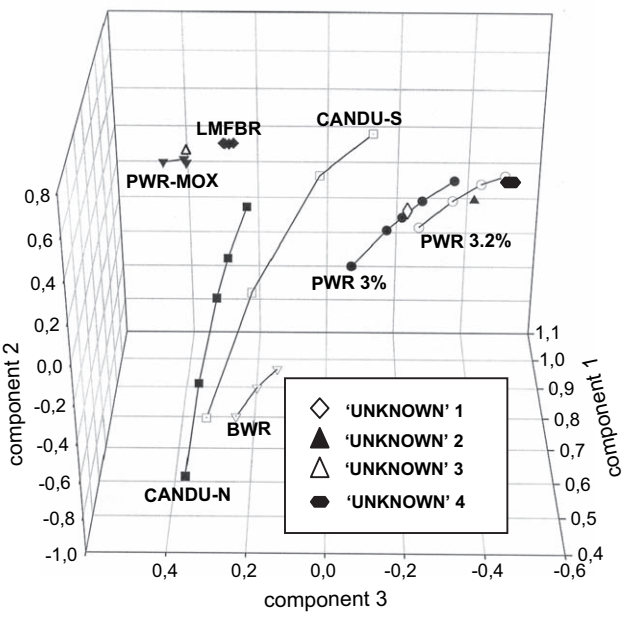

Fig. 1. A 3D plot of the factor analysis showing the similarities between the simulated and 'unknown' fuels.

and the neutron spectrum. During these changes, the U isotopes decrease forming isotopes of Pu, through (n, γ) and $(n, 2n)$ reactions and β^--decay. In this way, the two PWR-UO_2 cases are well separated on the basis of their different initial isotopic composition of U, i.e. the enrichments of 3% and 3.2% in ^{235}U. Similarly, the two CANDU cases of U_N – natural uranium fuel and U_S – slightly enriched uranium fuel are well separated.

The origin, namely the fresh composition, reactor type and burnup, of the four 'unknown' spent fuels can now be inferred from their clustering with the simulated fuels (Fig. 1). The four 'unknown' fuels are clustered with their corresponding fuel–reactor combination (Fig. 1), namely PWR-UO_2 and -MOX. The predicted origin of the 'unknown' spent fuels and their corresponding information declared by the reactor operators are shown in Table 2. The burnup is inferred to within 4% of the declared value. The method has, therefore, determined correctly the fuel type, the reactor where it was irradiated and the fuel burnup, parameters which characterize the origin of the 'unknown' spent fuels.

The method has been based on the U and Pu compositions at EOI of both the 'unknown' and the simulated spent nuclear fuels. Nevertheless, in an actual situation, the cooling time of a nuclear material with forensic importance would be unknown. The following transitions affect the composition of the spent nuclear fuel in ^{234}U, ^{238}Pu, ^{240}Pu and ^{241}Pu due to cooling: ^{238}Pu α-decay (half-live $\tau_{1/2} = 87.75$y) to ^{234}U, ^{241}Pu β^--decay ($\tau_{1/2} = 14.9$y) to ^{241}Am, buildup of ^{238}Pu from α-decay of ^{242}Cm ($\tau_{1/2} = 163$ d) and buildup of ^{240}Pu from α-decay of ^{244}Cm ($\tau_{1/2} = 18.1$ y).

The effect of using the U, Pu compositions at EOI, instead of the one due to the cooling of the 'unknown' material, on the origin determination was investigated. Factor analysis was carried out using the U, Pu compositions of the 'unknown' at 10 years cooling and of the simulated spent fuels at EOI. The origin determination, regarding the fuel and reactor types, of the 'unknown' fuels remained unaffected. However, the burnup prediction is now within 20%.

The effect of the accuracy of the measurement of the U and Pu compositions of the 'unknown' fuel on its origin determination was investigated. A sensitivity analysis carried out indicated that, for an error of less than $\pm 1\%$ on the measurement of the U and Pu compositions, the prediction of the origin of the 'unknown' fuel remained unaffected. This is a rather favorable outcome since analytical chemistry techniques of high accuracy ($< \pm 0.5\%$) are available in nuclear forensics.

4. Conclusions

An isotopic fingerprinting method was presented to tackle an important and challenging task in nuclear forensic, which is the origin determination of an unknown seized nuclear material. Spent fuel of known origin was assumed to be the 'unknown' nuclear material in order to demonstrate the method presented in this study and verify its prediction capabilities.

The method accurately predicted the fuel type, the reactor where it was irradiated and the fuel burnup, and hence the origin of the 'unknown' spent fuels. Furthermore, it was sensitive enough to resolve not only fuels from different reactor types but also fuels from the same reactor but with different fresh composition.

The use of the U, Pu compositions of the 'unknown' fuels at 10 years cooling and of the simulated spent fuels at EOI, affected only the prediction of burnup. A sensitivity analysis carried out indicated that the accuracy of present day analytical techniques available to measure U and Pu is adequate for the determination of the origin of unknown irradiated nuclear fuel.

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