Identification of unknown irradiated nuclear fuel through its fission product content

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A procedure is demonstrated, through a simulation study, for the determination of the origin of unknown spent nuclear fuel, an important and challenging task in nuclear forensics. The procedure is an isotopic fingerprinting method relying on the fission product content of the unknown. The 'unknown' nuclear material is represented by the spent nuclear fuel of known origin in order to demonstrate the method and verify its predictive capabilities. The method is based on the comparison of the fission product compositions of the 'unknown' material and simulated known spent fuels from a range of commercial nuclear power stations using the multivariate statistical technique of factor analysis. Then, the provenance of the 'unknown' spent fuel is the commercial fuel with which it exhibits the highest similarity with respect to the fission product content.

Introduction

Nuclear forensics, a scientific discipline developed over the last decade, is concerned with the illicit trafficking of nuclear material across the borders of different countries. Its primary objective is the timely detection of illicit handling of nuclear material and the determination of its origin and trafficking route from diversion to interception.[1–5] Then, appropriate measures and effective physical barriers towards combating future diversions of nuclear material away from its designated areas could be taken. Hence, particular attention to nuclear forensics has been given by the scientific and political communities, through technical and legislative measures, aiming to combat the illicit handling of nuclear material with radiotoxic potential.[6–8]

The determination of the origin of an unknown nuclear material, intended for illicit use, is an important and challenging task in nuclear forensics. Source attribution is generally done by comparing measurements of well-chosen parameters of the unknown material to appropriate data contained in databases. Then, the origin of the material can be identified as the one of the material with the most similar parameters in the data bank. Spent nuclear fuels or spent fuel reprocessing solutions, are possible nuclear materials of interest to nuclear forensics.[3]

The objective of this work is to demonstrate, through a simulation study, a procedure for the determination of the origin of an unknown spent nuclear fuel, through an isotopic fingerprinting method.[9] The term origin in this work refers to the fresh fuel type from which the unknown material originates, the reactor type in which the fuel was irradiated and its final burnup. The method is based on the fission product content of the unknown nuclear material as the characteristic signature in the isotopic fingerprinting method.

Experimental

Procedure

The procedure is based on the fact that the content of individual radionuclides in spent nuclear fuel is inherently consistent. This content reflects the fresh composition of the fuel and its irradiation history, hence, carrying information which can be uniquely related back to the origin of the fuel. The identification procedure is based on the use of some measurable parameters, and in particular the fission product content in the spent fuel.

The procedure would compare the fission product content of an unknown spent nuclear fuel to that of commercial spent fuels originating from a wide range of nuclear power stations. The fresh composition of the commercial fuels, the reactor type in which they were irradiated and the irradiation history are well known. The comparison seeks the highest similarity in fission product content between the commercial fuels and the unknown material. Then the origin of the unknown spent fuel can be identified as the one of its most similar commercial fuel with respect to the fission product content. The required comparison is performed using the multivariate statistical technique of factor analysis.[9,10]

The choice of the fission products, required as signatures in the fingerprinting procedure, would depend on the techniques used in the analysis of their content in the unknown fuel. Mass and high resolution gamma-spectrometry are quantitative techniques extensively used to determine the composition of spent nuclear fuel.[11–13] The former would be used on spent fuel in a dissolved form, while the latter on solid and dissolved spent fuel. In order to overcome isobaric interferences in the measurement of fission products by mass-spectrometry, an ion chromatography (IC) column would be coupled on-line to an inductively coupled plasma mass spectrometer (ICP-MS).[11]

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The fission products chosen as signatures should evolve during the irradiation of fuel in the reactor in a way that depends on, and hence reflects, the sought information, which is the type of fresh fuel and its irradiation history in the reactor. Mass spectrometry provides information on the whole range of fission products, among them several stable ones whose content is independent of the cooling of the spent fuel. On the other hand, gamma-spectrometry identifies few radionuclides relying on their natural decay, hence been dependent on the cooling of the material.

The stable fission products ^{133}Cs, ^{140}Ce, ^{150}Sm, ^{152}Sm, ^{144}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd are chosen as signatures in the case of mass spectrometry. The fission products ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{134}Cs, ^{137}Cs, ^{144}Ce are considered as the measurable characteristic parameters in the case of gamma-spectrometry. The nuclide ^{95}Zr could only be used in the origin prediction of dissolved spent fuel from thermal reactors, with the cladding removed prior to reprocessing, or solid spent fuel from fast reactors. This is due to the presence of zirconium in the cladding of the fuel rods from thermal reactors, which would interfere in the gamma-spectrometry with the ^{95}Zr fission product.

Simulations

In the absence of sufficient experimental data on fission product compositions in spent nuclear fuels, these were simulated for the unknown and the necessary commercial spent nuclear fuels considered in the comparison procedure. The compositions were calculated from reactor physics equations using appropriate depletion computer codes such as the zero-dimensional code ORIGEN-2.[14] These calculations are means to predict the changes taking place in the fuel composition as a result of irradiation in a reactor neutron spectrum during power operation and cooling. The simulations performed were coupled with burnup-dependent cross section libraries resembling, as closely as possible, the fuel type and neutron spectrum of irradiation. The isotopic compositions considered in the study were the ones at the End-of-Irradiation of the fuels (EOI).

The stable fission products were chosen on the basis of the good agreement that has been reported in appropriate studies, between their simulated and measured content in spent nuclear fuel:[11,15,16] ^{133}Cs (agreement within 4%), ^{140}Ce (1.5%), ^{150}Sm (<1%),

^{152}Sm (2%), ^{144}Nd (<6.5%), ^{145}Nd (<1%), ^{146}Nd (<1%), ^{148}Nd (<1%), ^{150}Nd (<1%). In the case of gamma-spectrometry, all radioactive fission products that can be seen in the spectrum, depending on the cooling time, are considered as signatures. Some of them show an agreement better than 3%, between calculated and measured content:[11] ^{95}Zr (3%), ^{95}Nb(3%), ^{106}Ru (9%), ^{134}Cs (4%), ^{137}Cs (3%), ^{144}Ce (1%). The level of the agreement reflects both the accuracy of the fission product measurements and the uncertainties of the simulations due to inaccuracies in the neutron reaction cross sections incorporated in ORIGEN.

Simulations were carried out for a range of commercial UO_2 and MOX fresh fuels, irradiated in PWR, BWR, CANDU and LMFBR power stations. In each case, the simulations were performed for a range of burnup values that could be encountered in a fuel cycle (Table 1). In order to demonstrate the approach, a spent fuel with 38 GWd/tU burnup, from the simulated PWR commercial UO_2 spent fuels, was chosen to be the 'unknown' material, whose origin is sought. The approach is based on the fact that the content in individual radionuclides in spent nuclear fuel is inherently consistent, as a result of the fresh fuel composition, reactor neutron spectrum and history of irradiation of the fuel. Hence, the 'unknown', although would cluster with the PWR fuel from which it originates, it would be expected to differentiate itself from the other fuel-reactor types.

Statistical analysis

The multivariate statistical technique of factor analysis was employed for the required comparisons. This pattern recognition technique groups together objects in such a way that similarities and differences between them can be graphically revealed. In general, each object may be characterized by a number of different parameters, N, for example ^{137}Cs composition, mass, porosity of material, size, shape, mechanical properties. The objects are then grouped together on the basis of the similarity of these parameters. Factor analysis is a dimension reduction technique combining and reducing the N coordinates of each object in the N-dimensional space into 3 dimensionless coordinates termed 'components' in a three-dimensional (3D) space.[10] Consequently, a projection of the objects onto the easier to interpret 3D space is obtained.

Table 1. Simulated reactor – fuel combinations and targeted burnup values

Reactor	Fuel	Burnup range, GWd/tU
PWR-U	UO_2, 4% ^{235}U	35–65
PWR-MOX	Thermal MOX [natural U, 4% fissile Pu]	30–40
BWR-UE	UO_2, 3.4% ^{235}U	25–40
CANDU-NU	Natural U	3–7.5
LMFBR-MOX	MOX [80% U depleted, 12% fissile Pu]	60–100

In this study, the objects considered were the simulated spent fuels described in Table 1 and the 'unknown' spent fuel. The N different characteristic parameters of these objects were their content in the fission products of interest. Then, providing that the objects are distinctly grouped together in a space according to the fresh fuel composition and the reactor type in which the fuel was irradiated, an unknown sample could be classified according to its proximity to the grouped objects. The provenance of the 'unknown' material would then be identified in terms of the provenance of its most similar commercial fuel in terms of their fission product content.

Results and discussion

The factor analysis results are shown in Figs 1–2 and 3–4 for for the cases when the radioactive and stable fission products, respectively, are considered as signatures. Patterns of similarities and differences between the 'unknown' and the fuels PWR-U, CANDU-NU, PWR- and LMFBR-MOX (Figs 1 and 3), and BWR-UE (Figs 2 and 4) can be identified. The plots were exploited from the view points of clustering the different fuel – reactor combinations and origin determination of the 'unknown' fuel. Furthermore, the effect of cooling time on the origin determination of the 'unknown' was investigated.

The origin of the 'unknown', namely its fresh composition, the reactor type in which it was irradiated and its final burnup, can now be inferred from its clustering with the simulated commercial fresh fuels. The simulated fuel – reactor combinations of PWR-U, CANDU-NU, PWR- and LMFBR-MOX form distinct groups well resolved from each other (Figs 1 and 3).

The method is sensitive enough to resolve groups of the same reactor type, on the basis of their fresh fuel composition. The exception was the case of BWR-UE which was not sufficiently resolved from PWR-U requiring an individual comparison with the 'unknown' (Figs 2 and 4). The 'unknown' has been clustered only with the simulated PWR-U fuels from which it actually originates (Figs 1 and 3), while it was well resolved from the other ones. Clustering of the fuels in the 3D space is influenced by the fresh fuel type, the neutron spectrum within the reactor, and the fuel burnup at EOI, resembled in the 3D plots by components 2, 1 and 3, respectively.
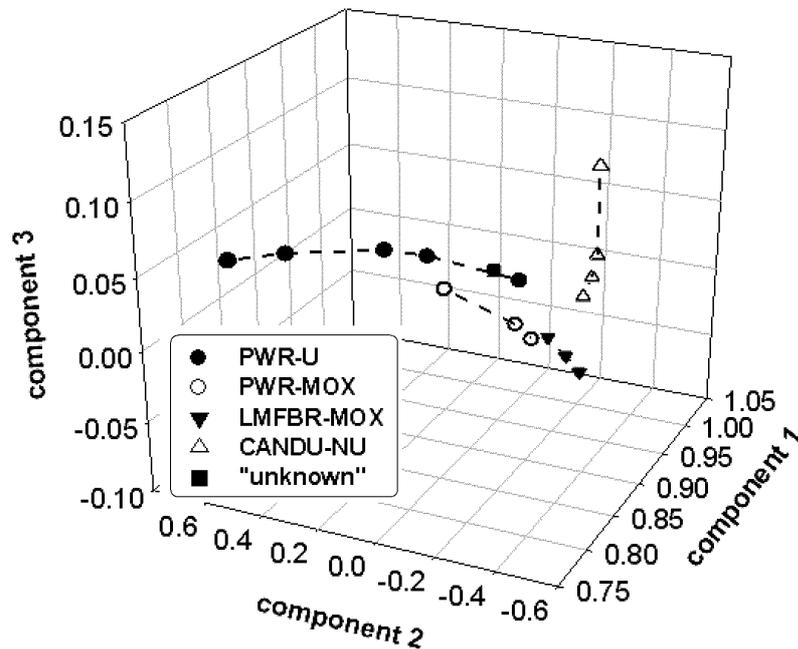

Fig. 1. Patterns of similarities between the simulated fuels and the 'unknown' based on the signatures from gamma-spectrometry
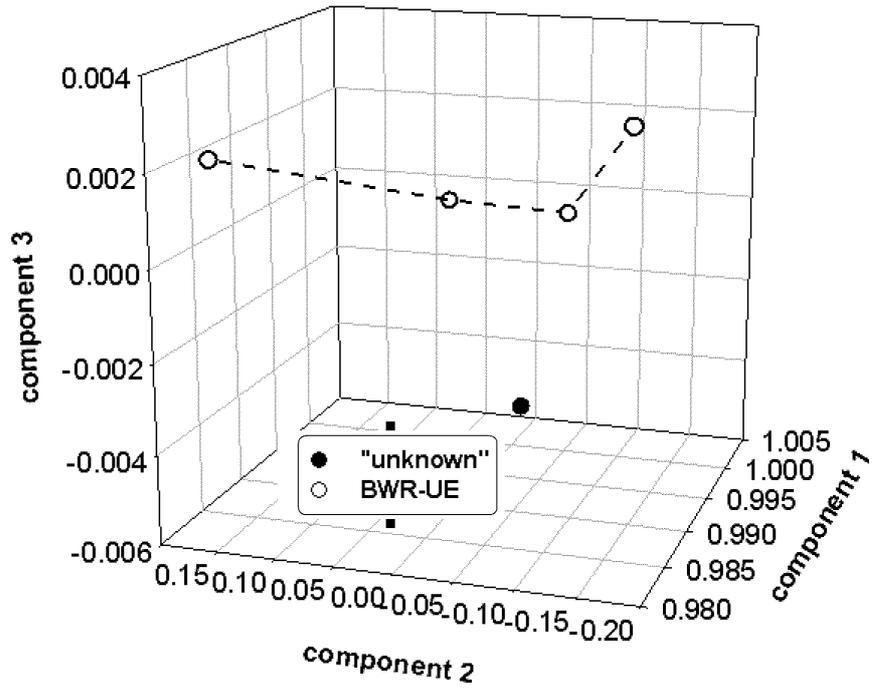
Fig. 2. Patterns of similarities between the simulated fuels and the 'unknown'
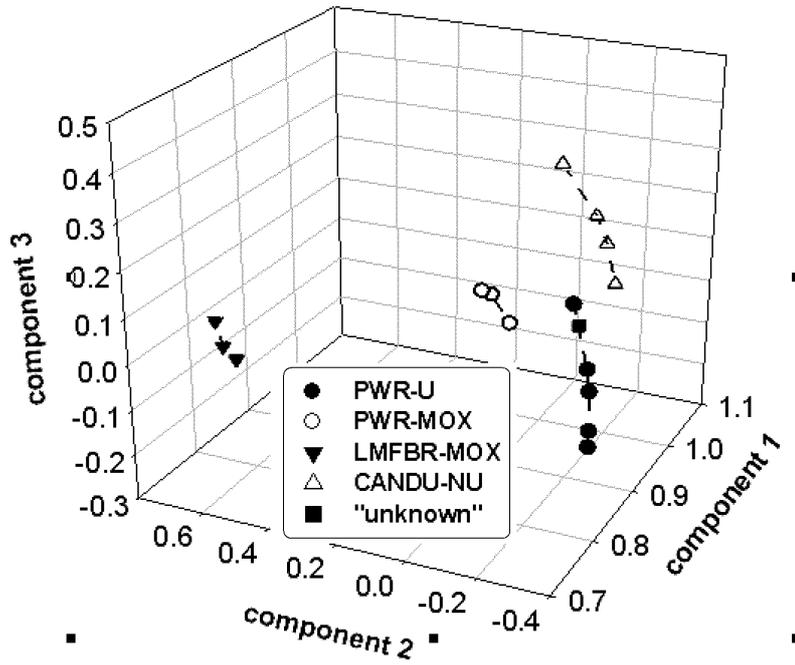based on the signatures from gamma-spectrometry

Fig. 3. Patterns of similarities between the simulated fuels and the 'unknown'
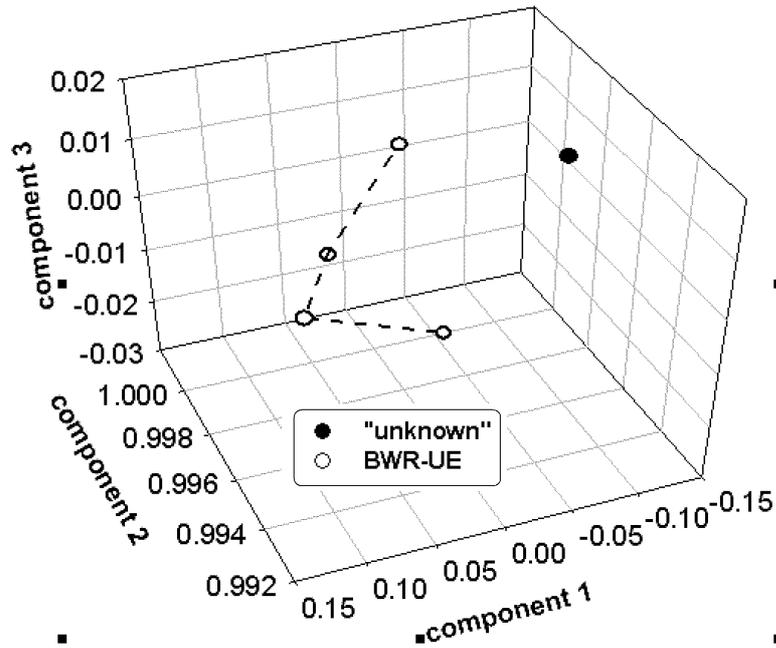based on the signatures from mass-spectrometry

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Fig. 4. Patterns of similarities between the simulated fuels and the 'unknown'
based on the signatures from mass-spectrometry

The method was based on the fission product composition at EOI for both the 'unknown' and the simulated spent nuclear fuels. The effect of cooling time of the 'unknown' on the prediction of its origin was investigated in the case the procedure was based on the radioactive fission products. The fission product compositions of the 'unknown' spent fuel at 1, 5, and 10 years cooling time were compared to the compositions of the commercial spent fuels at EOI. It was found that the origin prediction of the 'unknown' was erroneous, for all three cooling times considered. Hence, compositions of radioactive fission products at different cooling times should be included in the procedure, in order to allow for their evolution due decaying after the EOI.

The agreement, between simulated and measured content in spent nuclear fuel, for most of the fission products considered as signatures, has been reported to be within 3%.[11,15,16] The effect of this level of agreement on the origin determination of the 'unknown' PWR fuel considered in this study was investigated. Hence, in a sensitivity analysis carried out, the 'unknown', with its composition increased up to 5%, was compared to the EOI compositions of the commercial fuels. This increase effectively resembles the percent level of agreement, assuming that the 'unknown' of this study is an actual seized material whose composition is measured and compared to the simulated commercial fuels. The analysis indicated that, for a change less than 3% in the composition of the

'unknown', its provenance was predicted correctly. Therefore, the procedure shows a promising potential for the issue of provenance determination, at the current level of the agreement.

Conclusions

A sensitivity study was carried out to demonstrate an isotopic fingerprinting procedure suitable for the determination of the origin of an unknown spent nuclear fuel. Fission products were used as the characteristic signatures required in the procedure. Spent fuel of known origin was assumed to be the 'unknown' nuclear material in order to demonstrate the procedure and verify its prediction capabilities.

The method accurately predicted the origin of the 'unknown' spent fuel, which is the fresh fuel type, the reactor type in which it was irradiated and the fuel burnup. 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.

A sensitivity analysis carried out has shown that for an uncertainty beyond 3% in the simulation, the 'unknown' considered in the study does not cluster with the PWR from which it originates.

In order to fully assess the procedure, it would be necessary to explore its dependence on different issues that may affect its potential and hence identify its limitations. Such issues, associated with isotopic

compositions, include: the uncertainty of the ORIGEN simulation; the dependence of the composition on the sample 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; the accuracy of the nuclear data used in the simulations; the accuracy of the chemical analysis carried out to determine the composition of the unknown material.

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