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The identification of spent PWR nuclear fuel in terms of its initial enrichment and final burnup is demonstrated. Spent UO_2 fuel from a PWR power station was used as the nuclear material of supposed unknown irradiation history. The identification procedure was based on determining the U and Pu isotopic composition of the fuel by chemical analyses, simulation calculations of fuel evolution and statistical analysis. The procedures followed and associated limitations are discussed.

1   INTRODUCTION

Nuclear material, during residence in different installations within the fuel cycle or in transport between them, is accompanied by declarations of its origin, initial composition, burnup and cooling time. Frequent accountancy of nuclear material, in order to verify its declared characteristics, is a fundamental procedure to ensure that material has not been diverted. It is based on the destructive assay through isotopic composition measurements and the non-destructive assay through detection and quantification of characteristic radiation emissions by the material. Considering that the evolution in nuclide concentration of the nuclear material is dependent of its irradiation and cooling histories, one would expect to extract information of the material and its history through the assay of the material. The objective of this work is to demonstrate the identification of spent fuel in terms of its initial enrichment and final burnup achieved during irradiation.

2   METHODOLOGY

The content of individual nuclides in a spent fuel is inherently consistent. Hence, the measurement through chemical analyses of selected nuclides, e.g. U and Pu isotopic composition, complemented by simulation of the evolution of nuclear material under irradiation, would allow the fuel identification.

A commercial UO_2 spent fuel of known initial enrichment and final burnup was considered for the purpose of the study as a nuclear material of unknown irradiation history. The commercial spent fuels considered for the purpose of the study as the unknown material had the following characteristics: UO_2 type of fuel from a PWR power station,

enrichment in ^{235}U 3.5 at%, declared final burnup 47 and 29 GWd/t ('UNKNOWN').

The isotopic composition of uranium and plutonium was determined through chemical analyses [1]. A data bank comprising sets of U and Pu isotopics, for a range of enrichments and burnups of UO_2 and (U, Pu)O_2 spent fuels, was created using the simulation code ORIGEN [2]. The isotopic composition of the fuel analysed was then compared with each one of the theoretical composition sets in the data bank in search of similarities. The comparison was performed through conventional statistics [3]. The burnup and enrichment of the theoretical data set with the highest degree of similarity to the spent fuel characterises the fuel in terms of these two parameters.

A data bank comprising sets of U and Pu isotopics, for a range of enrichments and burnups of commercial UOX and (U, Pu)O_2 spent fuels, was created using the simulation code ORIGEN. These simulation calculations are means to predict changes taking place in the fuel composition as a result of irradiation, in a reactor neutron spectrum during power operation and cooling. They can be calculated from reactor physics equations, using appropriate computer codes such as ORIGEN. Alternatively, experimentally obtained data can also be used to set up these data banks. This procedure requires however, the accumulation of a sufficient number of reliable and accurate data, while the data banks thus obtained are only applicable to fuel types similar to those which provided the data.

The calculations performed for this study were coupled with burnup-dependent cross-section libraries resembling, as closely as possible, the type of reactors where the fuel was irradiated. They are thus fuel- and reactor-dependent. Consequently, they reflect each individual case, and are based on the initial enrichment and type of fuel data supplied by the operator of the reactor [4]. Calculations were performed for UOX and (U, Pu)O_2 fuels from power stations: PWR UO_2 [enrichments in ^{235}U 3.5%, burnup range 35, 40, 45, 50, 55 GWd/t (f1 - f5)]; PWR (U, Pu)O_2 MOX [U natural, 5% Pu, burnup range 40, 45, 50 GWd/t (f6 - f8)]; BWR UO_2 [enrichments in ^{235}U 2.7%, burnup range 20, 25, 30, 35, 40 GWd/t (f9 - f13)]; CANDU U [U natural, burnup range 3, 5, 6, 8, 10, 12 GWd/t (f14 - f19)]; CANDU UO_2 [enrichments in ^{235}U 1.7%, burnup range 5, 10, 15, 20 GWd/t (f20 - f23)]; LMFBR (U, Pu)O_2 MOX [U natural, 25% Pu, burnup range 60, 80, 100 GWd/t (f24 - f26)]; The sets of uranium and plutonium isotopics for each enrichment-burnup combination formed the data bank for the identification purposes.

Statistical treatment of the results was carried out through the inverse of the Euclidean distance coefficient between each of the data sets and the 'UNKNOWN' fuel acting as a measure of comparison. In search for the smallest difference, i.e. highest similarity. The comparison was based on the isotopic composition of U and Pu, namely ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu, considered as the measurable characteristic parameter for the comparison purposes. The choice of these parameters was based on the fact that they can be measured with the highest accuracy. ^{235}U and Pu reflect the initial enrichment of the fuel, whereas U/Pu indicate whether it is a UO_2 or (U, Pu)O_2 fuel. Clearly, the smallest distance coefficient indicates the two objects compared are similar. Hence, possible similarities between the fuels under identification and the isotopic composition sets in the data bank could be established

3   RESULTS

The degrees of similarity between the 'UNKNOWN' and data bank sets, as obtained from the conventional statistical approach, are given in Tab. 1. The results are presented in the form of the matrix ['UNKNOWN' vs. (f1 - f26)]. The elements of the matrix are the inverse of the Euclidean distance coefficient from the comparison of each 'UNKNOWN' with each of the data bank sets. An 'UNKNOWN' is identified as similar to that set from the data bank (f 1 to 26) with which it exhibits the smallest distance coefficient. Hence, the burnup and enrichment of that data set characterise the 'UNKNOWN' in terms of these two parameters. The parameters identified for the spent fuel, acting as the material of the 'UNKNOWN' burnup and initial enrichment in this study, are the ones of fuel f4 (highest coefficient). The enrichment and burn-up declared by the operator are included for comparison purposes. It is seen from the table that the 'unknown' fuels are predicted correctly.

A major source of errors in the calculation of the evolution of a fuel in a reactor is the burnup dependence of the cross-sections used. The cross-section libraries should accurately represent a given reactor-fuel system. The data bank of this study holds strictly for these PWR spent fuels since burnup-dependent cross-sections for such reactors were employed in ORIGEN. The three ratios of which the present study was based have predicted the burnup and cooling time of the 'unknown' fuels. However, a general data bank covering the whole range of reactor types and fuels should contain information such as complete actinide and fission product inventories and physical characteristics of the fuels. However, the feasibility of the procedure described have for a wide range of reactor-fuel combinations to be demonstrated along with the criteria with which to optimise the comparison.

Table 1
Matrix of the inverse of the Euclidian distance coefficient between the 'unknown' material and the members of the data bank.

	f1	f2	f3	f4	f5	f9	f10	f11	f12
'UNKNOWN'	0.950	0.962	0.989	0.994	0.964	0.763	0.856	0.939	0.988

	f13	f14	f15	f16	f17	f18	f19	f20	f21
'UNKNOWN'	0.985	0.766	0.776	0.781	0.876	0.867	0.810	0.746	0.939

	f22	f23	f6	f7	f8	f24	f25	f26
'UNKNOWN'	0.733	0.566	0.140	0.111	0.104	0.451	0.452	0.453

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