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Gamma-ray spectrometric measurements of fission rate ratios between fresh and burnt fuel following irradiation in a zero-power reactor

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ABSTRACT

The gamma-ray activity from short-lived fission products has been measured in fresh and burnt UO_2 fuel samples after irradiation in a zero-power reactor. For the first time, short-lived gamma-ray activity from fresh and burnt fuel has been compared and fresh-to-burnt fuel fission rate ratios have been derived.

For the measurements, well characterized fresh and burnt fuel samples, with burn-ups up to 46 GWd/t, were irradiated in the zero-power research reactor PROTEUS. Fission rate ratios were derived based on the counting of high-energy gamma-rays above 2200 keV, in order to discriminate against the high intrinsic activity of the burnt fuel.

This paper presents the measured fresh-to-burnt fuel fission rate ratios based on the ^{142}La (2542 keV), ^{89}Rb (2570 keV), ^{138}Cs (2640 keV) and ^{95}Y (3576 keV) high-energy gamma-ray lines. Comparisons are made with the results of Monte Carlo modeling of the experimental configuration, carried out using the MCNPX code. The measured fission rate ratios have 1σ uncertainties of 1.7–3.4%. The comparisons with calculated predictions show an agreement within $1–3\sigma$, although there appears to be a slight bias ($\sim$3%).

1. Introduction

The ongoing trend of increasing the initial fuel enrichment in order to obtain higher discharge burn-ups has led to more and more heterogeneous core configurations in modern light water reactors. As a consequence, a general need has been recognized for experimental data in the high burn-up range to validate the predictions of reactor physics code packages [1]. To provide such data, the Paul Scherrer Institute (PSI) and the Swiss association of nuclear utilities (swiss*nuclear*) jointly launched the experimental program LIFE@PROTEUS [2] in 2006, the main goal being the investigation of mixed lattices of fresh and highly burnt fuel (burn-ups of up to 60 GWd/t) in the test zone of the zero-power research reactor PROTEUS at PSI. A key type of measurement to be made thereby is that of fission rate distributions across fresh-to-burnt fuel interfaces.

One standard technique to determine fission rates is based on the measurement of the gamma-ray activity of fission products after irradiation [3]. So far, however, the measurements on burnt fuel have been limited to commercial power reactors, where power distributions in fuel assemblies during the last weeks of operation were determined by measuring the ^{140}La gamma-ray line at 1596 keV [4]. The measurement of fission rates in highly burnt fuel following an irradiation in a zero-power reactor requires, because of the low power, the development of a new experimental method that is able to discriminate against the high intrinsic gamma-ray and neutron background of burnt fuel. Two approaches are being investigated currently at PSI. One is based on the measurement of delayed neutrons from the irradiated fuel and has been presented elsewhere [5]. The other, which is the topic of this paper, is based on the detection of gamma-rays from short-lived fission products the energies of which are above the intrinsic background.

During preliminary measurements, fresh fuel was irradiated in PROTEUS and several fission products with gamma-ray lines above 2200 keV were identified as potential candidates for the proposed new measurement technique [6]. Among them, the ^{142}La (2542 keV), ^{89}Rb (2570 keV), ^{138}Cs (2640 keV) and ^{95}Y (3576 keV) gamma-ray lines were later measured in a fresh and a 36 GWd/t burnt UO_2 sample. Each of these samples was irradiated at different lattice positions in the PROTEUS test zone, and ratios of fission rates between the different positions were derived [7]. These inter-position fission rate ratios had uncertainties

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of about 1–3% and 3–6% for the fresh and burnt fuel sample, respectively, the main contributions being the counting uncertainties for the different gamma-ray peaks.

In the current paper, first results for inter-sample fission rate ratios between a fresh and two different burnt fuel samples (with burn-ups of 36 and 46 GWd/t, respectively) are presented. In comparison to the previously published inter-position fission rate ratios, the derivation of the present results has been considerably more challenging. This follows from the fact that various factors related to the counting efficiency of the detection system no longer cancel out and need to be accounted for explicitly for the two different fuel sample types.

This paper is organized in the following way. In Section 2, the conducted experiments are described. Section 3 provides a description of the Monte Carlo (MCNPX) models that were used (a) to predict the inter-sample fission rate ratios, and (b) to estimate the solid angle and attenuation correction factors. The methodology to derive inter-sample fission rate ratios from the measured gamma-ray lines is presented in Section 4. Section 5 investigates the sensitivity of the results to certain specific experimental features. The calculation-to-experiment comparison is presented and discussed in Section 6, while Section 7 gives the final conclusions and recommendations for future experiments.

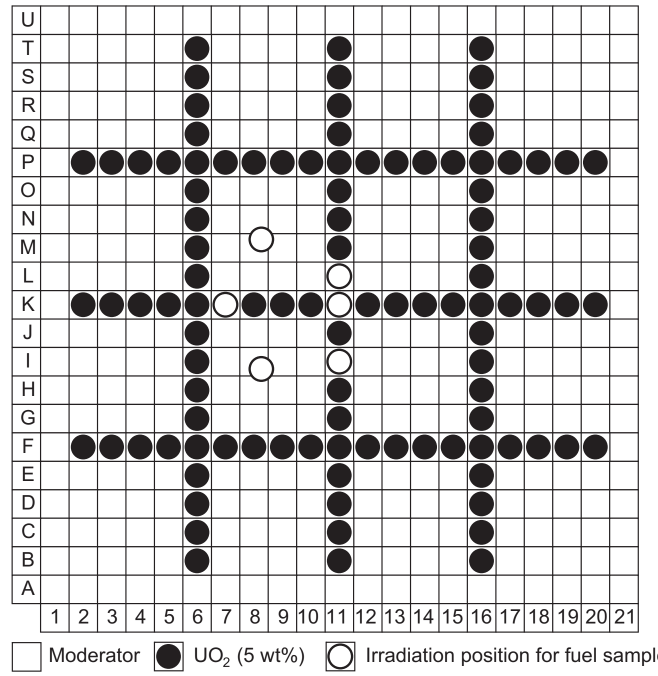

Fig. 1. Layout of the PROTEUS test lattice.

2. Experimental set-up and procedures

This section provides a brief description of the conducted experiments, a detailed description given in [8].

2.1. Experimental set-up

The experiments were carried out at the PROTEUS zero-power research reactor. This critical facility is a multi-zone driven system featuring a central test zone ($\sim 45 \times 45$ cm^2), which can be loaded with the fuel configuration to be studied. The test zone itself is subcritical and is fed with neutrons from the surrounding annular driver zones. Fig. 1 shows the test zone configuration that was currently used [7]. The test lattice consisted of 5% enriched UO_2 pins loaded in a pattern resembling that of a specific supercritical-water reactor (SCWR) assembly design. The test zone was filled with a H_2O/D_2O mixture to simulate water with a reduced density.

As indicated earlier, three different fuel samples were used in the present experiments: a 3.5% enriched fresh UO_2 sample, a 36 GWd/t burnt UO_2 sample (initial enrichment 4.1%) and a 46 GWd/t burnt UO_2 sample (initial enrichment 3.5%). All samples had been part of fuel pins irradiated in a Swiss PWR. The cooling time of the burnt samples was about 12.5 years. Each sample had a total pellet length of approximately 40 cm and a pellet diameter of about 0.91 cm.

For the experiments, the fuel samples were placed into a specially designed transport flask. The latter was made of steel and had an outer diameter of about 70 cm. The samples were loaded into a rotary revolver at the center of the flask. Placed above the test zone of the reactor, within the reactor shielding, the flask also served as a sample changer for lowering the samples into the PROTEUS test lattice. For this purpose, the sample changer was equipped with a control unit to rotate the revolver hosting the samples in the flask, and to move the samples up and down.

One at a time, the samples were inserted into different lattice positions for irradiation, and their gamma-ray activities were measured several minutes after the end of irradiation. The gamma-ray measurements took place directly within the steel body of the sample changer, using an HPGe detector (ORTEC model GEM-15180-P) that was located in a cavity drilled into the steel body. The signals were processed by a DSPEC Plus multi-channel analyzer from ORTEC. A live-time spectrum and a so-called zero-dead-time (ZDT) spectrum were recorded in parallel. The latter spectrum is continuously corrected for dead-time losses on a channel-by-channel basis during data acquisition. Considering the rapidly changing count rates during the data acquisition, the ZDT option was particularly suitable for the current experiments. Demonstration of the reliability of the ZDT methodology for high dead times and strongly varying count rates has been reported elsewhere [9]. The acquired data were saved with the ORTEC software Gamma-Vision [10], while the software HyperLab [11] was used for the analysis of the ZDT spectra.

Two different measurement positions were used for the fresh and the burnt fuel samples. As illustrated in Fig. 2, the fresh fuel sample was measured as close as possible to the detector, the distance between the sample and detector centers being about 20 cm. Due to their high intrinsic gamma-ray activity, the burnt fuel samples had to be moved to a position further away from the detector in order to keep the system dead time reasonably low. Consequently, the burnt samples were measured in a position below the detector, rather than in line with it, the distance between the top of the sample and the detector being about 26 cm. During the measurement of a given sample, the other burnt samples were moved to positions below the sample changer so as to avoid any interference from their intrinsic gamma-ray activity.

2.2. Irradiation and measurement conditions

The irradiation and gamma-ray measurement conditions for the three samples are summarized in Table 1. The cooling time between irradiation and data acquisition corresponds to the time needed to withdraw the sample from the core and bring it into the measurement position. All times were measured with a precision of 1 s. The maximum dead times were the system dead times at the beginning of data acquisition (5 min after irradiation), while the minimum dead times refer to the values at the end of