STUDY OF THE β DECAY OF FISSION PRODUCTS WITH THE DTAS DETECTOR*

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Total Absorption Spectroscopy measurements of the β decay of ^{103}Mo and ^{103}Tc, important contributors to the decay heat summation calculation in reactors, are reported in this work. The analysis of the experiment, performed at IGISOL with the new DTAS detector, show new β intensity that was not detected in previous measurements with Ge detectors.

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1. Introduction

The β decay of fission products is responsible for most of the energy released in a nuclear reactor after shut-down. The so-called *decay heat* varies as a function of cooling time, and can be calculated from the nuclear data corresponding to all nuclei produced during the fission process by means of the summation method. This method consists of summing the activities of the nuclide involved weighted with the mean γ, β and α energies released per decay, as it is represented by the following equation:

$$f(t) = \sum_i \left(\bar{E}_{\beta,i} + \bar{E}_{\gamma,i} + \bar{E}_{\alpha,i}\right) \lambda_i N_i(t)\,, \tag{1}$$

where $f(t)$ is the power function, $\bar{E}_i$ is the mean decay energy of the i^{th} nuclide (β, γ and α components), λ_i is the decay constant, and $N_i(t)$ is the number of nuclide i at cooling time t. $\bar{E}_\beta$ and $\bar{E}_\gamma$ depend on the β-intensity distribution, I_β, for those nuclei that undergo β decay.

The main limitation of this method is related to the lack of information for some nuclei. In particular, I_β data available in the international databases that come from high resolution experiments with Ge detectors, may suffer systematic uncertainties due to the *Pandemonium* effect [1], which occurs when feeding to high-energy levels is underestimated due to the modest efficiency of such detectors to high-energy γ rays. Total Absorption Gamma-Ray Spectroscopy (TAGS) has been shown to avoid this systematic uncertainty, and some of the most important contributors to the decay heat have been already measured with this technique, giving rise to important improvements in decay heat calculations [2]. In TAGS measurements, the detection efficiency is maximized by using large scintillator crystals covering a solid angle of $\sim 4\pi$ to absorb the entire γ cascades de-exciting the daughter nucleus after the β decay, rather than detecting individual γ rays. The "inverse problem" $\boldsymbol{d} = \boldsymbol{R}(B)\boldsymbol{f}$ has to be solved in order to determine I_β with this technique [3], where $\boldsymbol{d}$ represents the experimental data, $\boldsymbol{f}$ is the feeding distribution we wish to determine, and $\boldsymbol{R}$ is the response function of the detector, that depends on the branching ratio matrix of the decay (B) and is calculated with Monte Carlo (MC) simulations [4].

2. Experiment

Measurements of several fission products of interest in reactor technology were carried out in 2014 at the upgraded IGISOL IV (Jyväskylä, Finland) [5] using the new segmented Decay Total Absorption Gamma-Ray Spectrometer (DTAS) for the first time [6]. This detector has been developed for the Decay SPECtroscopy (DESPEC) experiment at FAIR, and it is made of 18 rectangular NaI(Tl) crystals of 150 mm $\times$ 150 mm $\times$ 250 mm [7].

The experiment was carried out with protons from the K130 cyclotron impinging on a natural uranium target. The resulting fission fragments were separated and driven to the JYFLTRAP double Penning-trap system [8] where the beam was purified. The nuclei were extracted from the trap and implanted on a tape placed in vacuum in front of a plastic β detector at the centre of DTAS. The software sum of the 18-crystal was reconstructed off-line and β–γ coincidences were required in order to obtain a spectrum free from environmental background. The DTAS spectrometer was characterized with Geant 4 [9] MC simulations of calibration sources.

Among all the cases measured in this campaign, here we will focus on the decay of ^{103}Mo and ^{103}Tc, that were pointed as priority one decays for the decay heat by the Nuclear Energy Agency (NEA) [10].

3. Analysis

^{103}Mo: The β decay $^{103}\mathrm{Mo} \rightarrow ^{103}\mathrm{Tc}$, with $Q_\beta = 3.635$ MeV and $T_{1/2} = 67.5$ s, was measured with an implantation rate of 800 nuclei/s. The first step in the analysis consists of identifying all the contaminants. Since the decay of the daughter was also measured, this contamination was subtracted and normalized according to Bateman equation. The summing-pileup contamination was also taken into account. It was calculated with a MC procedure based on the random superposition of two stored events within the ADC gate, and it was normalized with a theoretical expression based on [11]. The detector response to the decay was calculated with the information of the decay scheme at low excitation energies [12] (a very incomplete information for this decay, in fact no evaluation for I_β was done before), and the nuclear statistical model at high excitation energies. Figure 1 (left) shows the quality of the reproduction of the measured spectrum with this response. From our preliminary analysis, 9% of the total I_β is obtained above 1621.1 keV, the last populated level known so far [12].

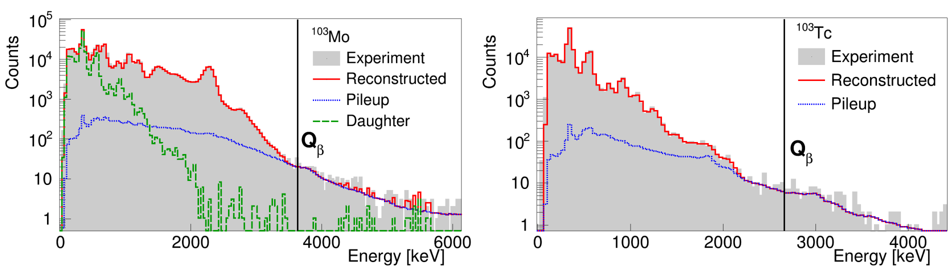

Fig. 1. (Colour on-line) Relevant histograms for the analysis of ^{103}Mo (left) and ^{103}Tc (right): parent decay (gray filled), daughter decay (dashed/green, only for ^{103}Mo), summing-pileup (dotted/blue) and reconstructed spectrum (solid/red).

^{103}**Tc:** The decay ^{103}Tc $\rightarrow^{103}$Ru with $Q_\beta = 2.662$ MeV and $T_{1/2} = 54.2$ s was measured with an implantation rate of 200 nuclei/s. The summing-pileup is the only contaminant, and the detector response was calculated as described above. The nice reproduction of the experimental spectrum is shown in figure 1 (right). From this preliminary analysis, 1.4% of the I_β is obtained above 1065.6 keV, the last populated level known [13]. Moreover, we can compare the absolute γ intensities measured with Ge detectors in [13], with the γ intensities determined from our I_β distribution. For example, at 346 keV, the main level, we obtain a 27% against the 28% from ENSDF.

In conclusion, we have obtained for the first time decay intensities for ^{103}Mo and our data for ^{103}Tc show a small amount of new intensity detected. Finally, in Table I, we present a preliminary evaluation of the mean energies obtained with the I_β distributions determined from our analysis. For ^{103}Tc, the new intensity does not increase $\bar{E}_\gamma$ and decrease $\bar{E}_\beta$ as expected, because we obtained a larger value for the ground state feeding.

TABLE I

Mean γ (left, in bold) and β (right) energies obtained from the preliminary TAGS analysis compared with values from ENSDF.

Nucleus	**ENSDF [keV]**	**DTAS [keV]**	ENSDF [keV]	DTAS [keV]
^{103}Mo	—	**343(150)**	—	1400(70)
^{103}Tc	**265**	**254(14)**	976	981(10)

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