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Measurements of isomeric yield ratios of fission products from proton-induced fission on ^{nat}U and ^{232}Th via direct ion counting

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Abstract. Independent isomeric yield ratios (IYR) of ^{81}Ge, ^{96}Y, ^{97}Y, ^{97}Nb, ^{128}Sn and ^{130}Sn have been determined in the 25 MeV proton-induced fission of ^{nat}U and ^{232}Th. The measurements were performed at the Ion Guide Isotope Separator On-Line (IGISOL) facility at the University of Jyväskylä. A direct ion counting measurement of the isomeric fission yield ratios was accomplished for the first time, registering the fission products in less than a second after their production. In addition, the IYRs of ^{nat}U were measured by means of γ-spectroscopy in order to verify the consistency of the recently upgraded experimental setup. From the obtained results, indications of a dependence of the production rate on the fissioning system can be noticed. These data were compared with data available in the literature, whenever possible. Using the TALYS code and the experimentally obtained IYRs, we also deduced the average angular momentum of the fission fragments after scission.

1. Introduction

The angular momentum of fission fragments is regarded as an important quantity in order to understand the fission mechanism because it can provide information on the scission configuration. One of the means to deduce the angular momentum of highly excited nuclei is by determining the yield ratio of low lying isomeric states.

As the fission theories do not provide directly information on the Isomeric Yield Ratio (IYR), their experimental determination can be particularly significant for simulations of processes such as the r-process, which is believed to be terminated by the fission of very neutron-rich heavy nuclei. Moreover, calculations of the effect of delayed neutrons in nuclear reactors require knowledge of the direct yield of metastable states in fission, since the β-delayed neutron emission probability from the isomeric state can be notably different from that of the ground state. Likewise, calculations of the reactor decay heat also require awareness of the population of the isomeric states. In addition, IYRs data can be of importance to experiments related to antineutrino spectra generated by nuclear reactors [1].

In the past, most of the studies on IYRs were performed by means of γ-spectroscopy, either by applying radiochemical separation or by using an ordinary isotope separator. These techniques have their perspective constraints. In the former case long lifetimes of the fission products are required, while in the latter one the method cannot be applied to refractory elements. Furthermore, precise knowledge of the branching ratio of the emitted γ-rays is required.

2. Experimental facility & measurements

In the present work, the isomeric fission yield ratios were measured at the Ion Guide Isotope Separator On-Line (IGISOL) facility at the University of Jyväskylä, in various experimental campaigns, from April 2010 until May 2014. Thus, a direct ion counting measurement of the relative intensities was accomplished for the first time, registering fission products even with very short lifetime. With the IGISOL technique [2], fission products can be accumulated, and by employing the high resolving power of the Penning trap JYFLTRAP, isomeric states separated down to a few hundred keV from the ground state can be measured. The first measurement of the Th(p,f) system, was performed at IGISOL-3, right before the facility had a shutdown for a major upgrade in order to be re-commissioned as IGISOL-4 in a new experimental hall [3,4].

The fission products were extracted using the IGISOL technique in all cases. Subsequently, two different techniques were employed to obtain the IYRs: the Penning trap was employed in all measurements, while in June 2013 the relative yields were also determined by means of γ-spectroscopy. The reason for employing the second technique was to check the consistency of the Penning trap method. In Table 1 we list information about the isomeric pairs that are presented in this work.

3. Analysis

3.1. Penning trap data

A detailed presentation of the data treatment can be found in [6]. In these measurements only the first trap (purification trap) was used. The selection of the desired

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Table 1. Information for the nuclides presented in this work. All data are retrieved from [5].

	Ground State		Isomeric State			
Nuclide	Spin	$\tau_{1/2}$ (s)	E_{exc} (keV)	Spin	$\tau_{1/2}$ (s)	Decay Mode
^{81}Ge	$9/2^+$	7.6	679	$1/2^+$	7.6	β^-=100%
^{96}Y	0^-	5.34	1140	8^+	9.6	$\beta^- = 100\%$
^{97}Y	$1/2^-$	3.75	667	$9/2^+$	1.17	$\beta^- > 99.3\%$, IT$< 0.7\%$
^{97}Nb	$9/2^+$	4326	743	$1/2^-$	58.7	IT = 100%
^{128}Sn	0^+	3544	2091	7^-	6.5	IT = 100%
^{130}Sn	0^+	223.2	1946	7^-	102	$\beta^- = 100\%$

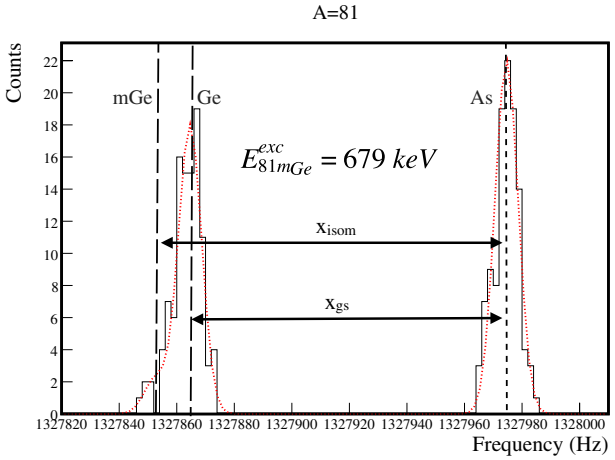

Figure 1. Frequency distribution spectrum of mass A = 81. The peak positions of ^{81m}Ge and ^{81}Ge are calculated relative to ^{81}As, which is taken as reference. The peak identification is based on a frequency to mass calibration. The red dashed line shows the Gaussian fit to the registered counts.

events and a background subtraction is achieved by Time Of Flight (TOF) gating, since the ions leave the purification trap with the same energy. For the identification of the peaks appearing in the spectrum a cyclotron frequency calibration based on the evaluated atomic masses was used [7].

The height of the mass peak can be considered as a direct measurement of the beam intensity since the maximum transmission efficiency through the trap is achieved at the cyclotron frequency [8]. By fitting a gaussian function to these peaks, their respective intensities can be calculated. The width of all peaks in the spectrum is assumed to be the same, since this is determined by the settings of the Penning Trap. Moreover, the relative positions of the two peaks are fixed to an isolated and well defined peak, based on their respective mass difference. In this way even close lying metastable states can be separated from the ground states. An example is given in the frequency distribution spectrum of mass A=81 in Fig. 1, where the position of the ground state and metastable state relative to a reference peak is shown. The red dashed line represents the fit to the data using Gaussian distributions.

The determined intensities were corrected due to the radioactive β-decays of the nuclides. The ions spend most of their time, from their creation to their detection, in the radio-frequency cooler and buncher and in the Penning Trap (∼400–600 ms in each apparatus). Any β-decay product captured in the trap will be doubly charged and hence, effectively removed in the purification cycle.

3.2. γ-spectroscopy

The ions were implanted in an aluminium foil which was placed in front of the HPGe detector, located at a dedicated stationary station. The data acquisition was performed over several hours. The intensities of the peaks were estimated from Gaussian fits. Before every measurement, the background activity was recorded and afterwards subtracted from the actual spectrum in order to reduce the contribution of the long-lived isotopes along the beam line. If more than one γ-ray could be determined quantitatively, the yield was calculated from the weighted average of all considered γ-rays. A full-energy peak efficiency calibration for the HPGe detector was performed with a ^{152}Eu source. The registered number of counts were corrected for the detector's efficiency and the γ-ray intensity, taken from the evaluated library ENDF [10].

Corrections to the determined activities due to contributions of precursors were applied. Nevertheless, due to the nature of fission there is a significant difference in the production rate distribution for the same mass number A, when moving from a nuclide with (A, Z-1) to (A,Z). Therefore contributions from just one precursor, if any, was taken into account. Since the irradiation time was much longer than the half-lives of the measured isotopes, except for the cases of the ground state of ^{128}Sn and ^{97}Nb, the obtained activities were converted to corresponding cross sections by correcting for saturation conditions.

4. Results

In Table 2 the results are presented in the form of ratios of the isomeric yield over the ground state yield. The only existing comparable data set in the literature is also included in the table [9]. The uncertainties include only the statistical ones, and thus they are underestimated. Specifically, they are between 3% and 25% for the Penning trap data, while they are between 6% and 28% for the γ-spectroscopy.

The comparison of IYRs from different fissioning systems is a complex situation. It is claimed, in general, that the population of the isomeric states is much more related to the nuclear properties of the individual products, than to the dynamics of the fission process. Nevertheless, the probability of populating a certain isomeric level depends, even weakly, on the nature and the excitation energy of the fissioning system.

The case of ^{81}Ge is the most difficult one for both methods. Concerning the Penning Trap data the isomeric state is only separated from the ground state by 679 keV, as can be noticed in Fig. 1, approaching the maximum resolving power of the purification trap. This difficulty is reflected in the large uncertainties. For the γ-spectroscopy

Table 2. Isomeric yield ratios of proton-induced fission on ^{nat}U and ^{232}Th.

Nuclide	Y_{isom}/Y_{gs}						
	^{nat}Uranium				^{232}Thorium		^{238}Uranium
	June 2013		August 2013	May 2014	April 2010[a]	April 2014	Tanikawa[b] [9]
^{81}Ge	0.04±0.01	0.59±0.13[b]	0.08±0.02	(0.15±0.03[c])	0.07±0.01		
^{96}Y	0.97±0.03		1.13±0.07		1.66±0.13	1.40±0.04	
^{97}Y		2.79±0.37[b]	2.36±0.13		2.59±0.14	4.51±0.43	2.39±0.36
^{97}Nb		0.30±0.03[b]	0.27±0.01				
^{128}Sn		0.86±0.13[b]				0.98±0.03	1.09±0.07
^{130}Sn	(0.89±0.31[c])	1.01±0.06[b]		1.01±0.08		0.64±0.08	0.52±0.02

[a]Performed at IGISOL-3, all the other measurements were performed at IGISOL-4.

[b]Performed by means of γ-spectroscopy.

[c]Omitted due to contamination in the spectrum.

data, the relative uncertainties are at similar levels, mainly due to the large uncertainties (up to 18%) of the tabulated γ-ray intensities of the isomer. Whenever the results do not agree with each other within the uncertainties, it might be due to the absence of systematic uncertainties or due to insufficient knowledge of the level scheme. The ratio deduced by means of γ-spectroscopy is rather different than any other result for unknown reasons.

The yield ratio of ^{96}Y was only measured using the Penning Trap, since the ground state does not emit any significantly intense γ-ray. It is worth mentioning that the isomeric state is produced, and consequently defined quantitatively, only in fission. The results from the same target agree within ∼15%, although not within the uncertainties, possibly for the same reasons as mentioned above.

The yield ratio of ^{97}Y was determined with both methods. The results from the Th target disagree with each other, while for the U target the results of the two techniques agree within uncertainties. For still unknown reasons, the result from the Th(p,f) system from 2014 is much higher than all others. Compared to data in the literature the results agree within errors, except for the aforementioned value.

IYR for ^{97}Nb was measured with the U target, but using both techniques. The results for these two measurements are in good agreement.

For extracting the yield ratio of ^{128}Sn, the Penning Trap was used with the Th target, while γ-spectroscopy was used with the U target and the results agree with each other within uncertainties. Nevertheless, the comparison between IYRs from different fissioning system should be done cautiously, as explained earlier, due to the different decay paths in the de-excitation of the fission fragments. The ratios of the present work from both targets agree reasonably with the measurement in [9].

The yield ratio of ^{130}Sn was measured in all experiments performed at the upgraded facility. For the γ-spectroscopy data no corrections due to contributions from the precursors were applied since these are considered insignificant, taking into account the yield of the fission products as reported in [11]. The result from June 2013 has the largest uncertainty, but it is discarded since the spectrum was distorted by impurities of non identified elements. The agreement of the ratios for the experiments performed with the U target, with both methods, is excellent, but it differs by a factor of two from the results of Tanikawa et al.

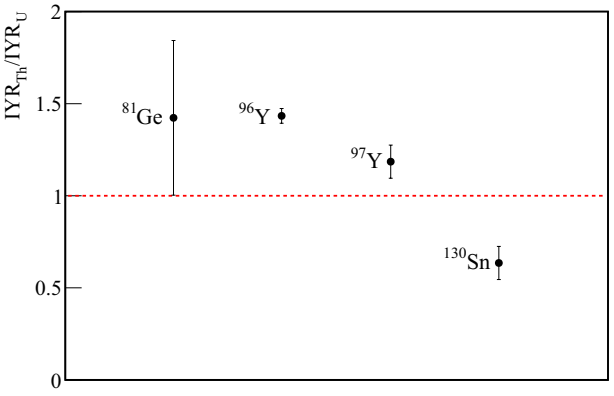

Figure 2. Ratios of IYR for the system of ^{232}Th(p,f) over the system of ^{nat}U(p,f) as studied in this work.

In Fig. 2 we show the ratio between the IYR for the two fissioning systems studied in this work. In cases where more than one measurement for one nuclide from the same target existed, the weighted mean was calculated. The yield ratios deduced from the Th target are higher compared to the ones from the U target, especially for fragments in the light mass region. Although some indications on a dependence of the fissioning system can be observed, more systematic studies on the same isomeric pairs from various fissioning systems are required before any safe conclusions can be drawn.

5. Extraction of ⟨J⟩ of the initial fragments

The spin distribution of the initial fragments is not possible to be determined from the observed IYR of the secondary fragments. However, the average angular momentum (⟨J⟩) of the primary fragment can be determined if a functional form of the primary spin distribution is assumed [12].

In the current work, the TALYS code [13] was used in order to extract the <J> of the primary fragment using an iterative procedure. Specifically, the code was used to calculate the IYR of a specific product, as a result of the de-excitation of a certain primary fission fragment. The FF was defined from the fission product considering the emission of a certain number of neutrons in order for its de-excitation to result in the desired fission product. Moreover, the excitation energy and an initially assumed average spin of the initial FF were given to the code as input parameters. For the calculations, a gaussian

Table 3. Average angular momentum of the primary fission fragment as calculated with TALYS. The observed product is assumed to be formed after the emission of one, two or three neutrons from the fission fragment.

	<J>					
	n=1		n = 2		n = 3	
Nuclide	U	Th	U	Th	U	Th
^{81}Ge	4.4 (3)	4.0 (1)	4.8 (3)	4.1 (3)	5.9 (4)	5.0 (4)
^{96}Y	6.5 (1)	6.8 (1)	7.1 (1)	7.7 (1)	6.6 (1)	7.5 (1)
^{97}Y	4.6 (1)	4.7 (1)	4.0 (1)	4.4 (1)	4.1 (2)	4.8 (1)
^{97}Nb	3.8 (1)		3.3 (1)		3.5 (1)	
^{128}Sn	6.3(3)	6.4 (1)	6.9 (3)	7.2 (1)	6.2 (5)	6.7 (1)
^{130}Sn	6.3 (2)	5.8 (2)	7.1 (3)	6.2 (3)	6.7 (2)	5.4 (5)

probability distribution for the spin was presumed with mean value the deduced <J>. The mean value was varied until a good agreement between the calculated and the experimental IYR was reached. For the experimentally observed ratios the weighted mean of each fissioning system was used.

The total excitation energy of the system was assumed to be shared between the initial fragments proportional to the number of emitted neutrons over the average number of total neutrons. The total average number and the TKE of the fissioning system were taken from [14] and [15] respectively. One to three neutrons emitted from each fragment were investigated for each case, whereas the extreme cases of none or all neutrons emitted from the same fragment were not considered due to their low probability. The deduced <J> of the initial fragment are presented in Table 3. Since the contribution of each primary fragment to the measured yield is not well known, the extracted values of <J> are presented separately for each case. The uncertainties in the table arise from the experimentally determined IYR.

6. Conclusions & remarks

In the current study six isomeric yield ratios were determined for 25 MeV proton-induced fission in ^{nat}U and ^{232}Th. They span over a wide mass range (A = 81–130). For the ^{nat}U(p,f) system, three isomeric pairs were measured for the first time (^{81}Ge, ^{96}Y and ^{97}Nb), while there are no available data in the literature for any of the pairs measured for the Th(p,f) system. An agreement within uncertainties of the obtained isomeric yields ratio could be observed in most cases, especially for the products of the same fissioning system. Nevertheless, some differences were noticed in cases where a different actinide target was used, indicating a dependence of the produced isomeric yields on the fissioning system. Moreover, the observed IYR was used in order to deduce the <J> of the initial fission fragments using the TALYS code, considering different number of emitted neutrons from the initial fragment. The present methodology is still under development. See also [16].

One of the advantages of the IGISOL-JYFLTRAP technique is that it does not rely on information on the level scheme which is often poorly known in the case of isomeric states of nuclides far from stability. This is particularly evident in the case of ^{97}Y, as the results from the Penning Trap show significantly lower uncertainties, compared to the ones from the γ-spectroscopy. The new method applied here can contribute to lower relative uncertainties, since it is performed by means of direct ion counting. Another advantage can be noticed in the ^{96}Y case, where the IYR could not be quantified due to the weak intensity of the emitted γ-ray from the ground state. On the other hand, the limitations of the method mainly arise from the half-lives of the states or from the mass difference between the ground state and the isomer. In any case, more measurements of various isomeric pairs are required, either for proton- or neutron-induced fission which is planned to be implemented at IGISOL [17].

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