Independent Fission Yield Studies at the JYFL Accelerator Laboratory

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ABSTRACT

Low-energy-particle induced fission is a cost-effective way to produce neutron-rich nuclei for spectroscopic studies. Fission has been used at the IGISOL facility of the Accelerator laboratory of the University of Jyväskylä to produce neutron-rich isotopes for nuclear structure studies, collinear laser spectroscopy and precision mass measurements. The IGISOL facility utilises the ion guide technique that, in addition to isotope production, is also very suitable for independent fission yield measurements. The most throughout yield studies so far have been performed on proton-induced fission of natural uranium and thorium. Studies of the independent proton-, neutron- and deuterium-induced fission yields of natural uranium has taken place as well. In the near future, the yield measurements can be speed up by utilising a MR-TOF spectrometer, while the close-lying isomeric states and nearby isobars close to stability can be resolved by applying the new techniques developed at the JYFLTRAP. This development is expected to allow comprehensive independent yield distribution studies for intermediate energy neutron induced fission.

1 INTRODUCTION

Accurate knowledge of the fission yields is of fundamental importance for better understanding of the fission process, which is required for the theoretical fission model development. For nuclear energy applications, the fission yield data are essential for development of nuclear technologies and assessment in safety issues. The Generation IV reactor concepts include fast reactors, where the fission yields are not the same as those of thermal ones. The accuracy of the fission yields as a function of the neutron energy is a challenge for the criticality and reactivity calculations, as well as for prediction of the fission product inventory of the spent fuel.

The Ion Guide Isotope Mass Separator On-Line (IGISOL) facility in the Accelerator laboratory of the University of Jyväskylä (JYFL-ACCLAB) utilises ion guide technique that was developed in the University of Jyväskylä in early 1980’s [1,2]. The facility produces high quality, mass separated ion beams of short-lived nuclear reaction products isotopes for nuclear structure studies, collinear laser spectroscopy and precision mass measurements. Particle induced nuclear fission provides means to supply also neutron-rich nuclei for these studies. In addition, it has been turned out to be extremely well suited instrument to determine the independent fission yields [3-5].

2 THE ION GUIDE TECHNIQUE

The ion guide method is a concept for an online ion source for an ISOL-type mass separator. In addition to the ion guide, IGISOL facility utilises a laser ion source as well as an advanced hot cavity ion source developed specifically for silver. The initial mass selection is provided by a 55° self-focussing dipole magnet with resolution \( \frac{M}{\Delta M} \approx 500 \). The mass separated beam is thereafter manipulated in a series of ion traps to improve the purity, time structure and optical quality of the radioactive beam before impinging it in the spectroscopic setups.

The ion beam is formed in the ion guide as follows. The reaction products recoiling out of a thin production target are stopped in flowing, extremely pure helium gas (figure 1). These reaction products are already highly ionised in the nuclear reaction. When these primary ions are thermalized in helium, their charge stage is reduced in charge-exchange reactions with helium, until they cannot draw any more electrons from helium atoms, whose first ionisation potential is the highest among all atomic species. As consequense, the final charge state of the stopped ions is concentrated to +1. The ions are flushed out of the stopping cell with the helium flow in milliseconds. Outside of the exit nozzle the neutral helium is pumped away with differential pumping.
while the ions are directed with electric fields to the acceleration stage of a magnetic mass separator.

It is worth of repeating that the ionisation is due to the nuclear reaction, which means that an ion beam of any element can be produced. The ion formation is independent of properties such as melting and boiling temperatures or vapour pressure of the element.

![Image of ion guide principle](image)

Figure 1: The ion guide principle depicted with the particle-induced fission reaction. Note in particular the thin foil between the production target and the stopping volume of the ion guide.

The main handicap of the method is the effective (or useful) target thickness being low, since the reaction products need to recoil out of the target with the momentum from the primary beam. This however applies only to the light-ion induced fusion evaporation, where the range of the low-energy recoils limits the effective thickness of the target to a few tens of nanometers. When particle induced fission is employed to produce neutron-rich nuclei, the efficiency of the ion guide is enhanced by the fission kinematics. The kinetic energy of the fission products does not come from the momentum of the projectile, but the energy release and kinematics in the fission reaction. The range of the fission products known to have kinetic energy of $\sim 1 A \cdot MeV$ is $3 - 4 \mu m$. In addition, the angular distribution of the fission products is isotropic, which allows locating the stopping chamber of the ion guide on the side of the target, instead of behind, and tilting the target to almost parallel to the primary beam. In the current fission ion guide the angle between the primary beam direction and the target is $7^\circ$. The thickness of the uranium target for the primary beam is thus $60 \mu m$ while its thickness for the fission products recoiling to the stopping volume is still $4 \mu m$ or less.

3 FISSION YIELDS

The fission ion guide was not developed to determine fission yields in the first place. The goal throughout years has been maximising the output of neutron-rich isotopes for spectroscopy experiments. However, it has been turned out to be well suited to determine the independent fission yields as well.

Firstly, all the elements produced in fission are accessible with the ion guide. There are no gaps in the systematic studies.

Secondly, the ion guide is fast. The fission products are transported from the target to the mass separated sample in tens of milliseconds, which is much less than the half-life of the majority of the fission products.

Thirdly, all the isotopes in the mass separated sample are primary fission products. This is because the ionisation of the reaction products that travel through the mass separator takes place in the fission itself. Moreover, no daughter isotopes of the primary products are released from the ion guide. There are secondary ionisation mechanisms of the species stopped in the target such as heating by the primary beam. The target region is however separated from the stopping region of the ion guide by a volume by a thin (1.2 $\mu m$) nickel foil. While the energetic primary fission recoils easily penetrate the foil, the secondary ions moving at thermal velocities do not.

![Image of fit results for Pd](image)

Figure 2: The relative independent isotopic fission product yields for palladium in the 25 MeV proton-induced fission of natural uranium. The yields are measured with the respect to the yield of the reference isotope $^{110}$Pd (solid mark). The fitted curve is a simple gaussian.

In addition, the simulations of the fission ion guide [6] show that separation foil also counteracts the mass dependency of ion stopping. The range of a heavy isotope is shorter than that of a light isotope of the same element. Because of the foil, the probability of stopping an ion in the stopping volume is however the same for the light and the heavy isotopes. This requires precisely the foil thickness used in the
fission ion guide. A thicker foil would favour heavier isotopes, thinner foil the lighter ones.

The main complication of the fission yield measurements with the ion guide is its chemical sensitivity. Although the ionisation is independent of the chemical properties of the ionised atom, the survival of the ions in helium is not, introducing the chemical dependency in the transport efficiency. As the result, only the yields of the isotopes of the same element can be directly compared, thus the measurements alone give the relative independent isotopic fission product yields (figure 2). When the mass yield data of the fission process in question is available, it can be combined with the measured yields to reveal the absolute independent yields. For detailed discussion, see [5].

3.1 Fission yields with γ-spectroscopy

The earliest fission yield measurements at the IGISOL facility date back in the early 1990’s. These measurements were based on producing a mass separated sample of fission products and deducing the cumulative yield of each isotope in the decay chain from the observed gamma ray intensities [7, 8]. Proton, deuterium and neutron induced fission yields were extracted from the gamma ray measurements. The measurements continued throughout the 1990’s [9,10], leading to the discovery of pronounced super-asymmetric fission mode in the intermediate excitation energy [10]. Neutron induced fission yields measurements with gamma spectroscopy continued still in 2000’s [9].

3.2 Fission yields with a Penning trap

Extracting fission yields with gamma ray spectroscopy is however tedious and vulnerable to error; usually the knowledge of the gamma ray branchings is limited, in particular far from stability. About 15 years ago, we started developing a method where the yield of a particular isotope is determined by counting ions instead of counting decays.

In most cases, the fission products can be unambiguously identified by their mass. The required resolution \( \frac{M}{\Delta M} \approx 10^5 \) is provided by the JYFLTRAP Penning trap [11]. JYFLTRAP is a double trap, housing two linear Penning traps inside one 7 T superconducting magnet. Usually the first, so called purification trap operated as a high resolution mass filter is sufficient. Figure 3 shows how clearly the isobars are resolved in the mass spectra of the fission products in the 25 MeV proton-induced fission of natural uranium in the mass numbers \( A = 110 – 118 \). Details of the measurements can be found in [3,5,11].

The yield of each isotope is determined by comparing the number of the counts in the corresponding mass peak to that of the reference isotope – \(^{117}\text{Pd}\) in the case of palladium. The recording of each mass spectrum and the reference spectrum are altered in such a manner that the measurement conditions are same for both. For details, see [3-5].

![Figure 3: Mass spectra of the fission products in the 25 MeV proton-induced fission of \(^{238}\text{U}\). Isotopes of palladium are aligned. The spectra are drawn as a function of reduced cyclotron frequency that is inversely proportional to the mass of the ion, thus the isotope mass decreases to the right.](image)

The mass resolution can be refined using more advanced techniques involving also the second trap, such as Ramsey cleaning [11], where \( \frac{M}{\Delta M} \approx 10^6 \) is approached, and PI-ICR (position imaging ion-cyclotron resonance) technique [12, 13] with mass resolving power of several millions. The higher resolution is needed for the isomeric yield ratio (IYR) measurements [13] and to resolve the masses in the \(^{132}\text{Sn}\) region, where the most probable yields come close to stable nuclei and the mass surface is remarkably flat.

The most throughout yield studies so far have been performed on proton-induced fission of natural uranium and thorium [5,14]. Studies of the independent neutron- and deuterium-induced fission yields of natural uranium has taken place as well.
For the development of the theory of the nuclear fission, charged particle induced fission is not different for neutron induced. The neutron induced fission yields are however more directly answering the needs of applications. Two projects are currently going on at the IGISOL facility to meet the request of neutron induced fission yields. They are development of proton-neutron converter target [14,16], for production of neutrons, and design of a faster, more effective ion guide.

3.3 Fission yields with MR-TOF

As fast and accurate the fission yield measurements with the Penning trap are, the mass spectra are collected one frequency point at a time by filtering the ions whose mass does not match the cyclotron frequency. Most of the time, everything is blocked. A novel mean is to use a multi-reflection time-of-flight (MR-TOF) spectrometer, recently installed at the IGISOL, primarily to serve as a pre-purification device for JYFLTRAP. Its mass resolution $\Delta M \approx 2 \times 10^5$ is the same order of magnitude as that of purification trap. With the MR-TOF, the time of flight is measured for all ions. It is therefore expected to be much faster and more sensitive than the Penning trap. This increased sensitivity of ion counting, together with improved ion guide efficiency, is expected to allow to measure independent fission yields with variable energy neutron fields also for more rare actinides in the future.

4 CONCLUSIONS

The IGISOL mass separator and the connected ion manipulation devices can be used to effectively determine independent fission yields. To fully utilise this feature, developments for studies of non-thermal neutron induced fission yields are in progress.

REFERENCES