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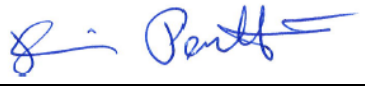
Prepared by:	H. Pentillä	15	10-06-2024	
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Summary

The purpose of this task was to study the applicability of PI-ICR technique to fission yield measurements. An important part of the test run in the task was a comparison of different techniques, to which extend their results agree.

The test run was originally intended to take place around the M24 of the project. Due to some incidents, including the break-ups of IGISOL gas purification system, the execution of the experiment was delayed until March 2023.

As hoped in advance, the PI-ICR technique turned out to be capable of resolving ions of adjoint isobars of the same mass number with a technique described more in detail in the report. The actual determination of the fission yields is based on comparison of number of detected ions of different isotopes. The methods for these ion count comparisons were beforehand baptised as “filtering” and “magnification”. Filtering means direct comparison of ion counts of different isotopes in the PI-ICR spectra taken in different mass numbers, while magnification means that PI-ICR technique is used to determine the relative intensity of overlapping mass peaks in each mass number separately, while the comparison between the mass numbers uses mass spectra achieved either with the sideband cooling technique in the first trap (purification trap) of the JYFLTRAP double Penning trap or with the Multi-Reflection-Time-Of-Flight (MR-TOF) device.

Because of all the delays, the development of the MR-TOF device, which was not mentioned at all in the SANDA proposal and only briefly in the original test run beam time proposal to JYU-ACCLAB Program Advisory Committee, had proceed so far that the device was available for on-line operation at the IGISOL. Some of the test run’s beam time was used to commission MR-TOF device for fission yield measurements. This commissioning was successful. A mass resolving power above 150000 for the fission products was reached. Such mass resolution is fully comparable with the one reached with the sideband cooling, and thanks to the much higher duty factor, the statistics was overwhelming.

The issue between of the possible precedence of the “filtering” and “magnification” techniques is still under investigation. A tentative impression is that the filtering technique is more sensitive to disturbances, as it was suspected. Prior to success of the MR-TOF in resolving fission products, the filtering was potentially providing a higher duty factor than magnification, because of the possibility of giving up the purification trap mass scan. Combining MR-TOF and PI-ICR techniques with “magnification” seems to be both the fastest and the most accurate method. However, the analysis of the experimental results is still in progress.

The results of this experiment will complete the Subtask 2.5.1 of the SANDA project.

The achieved data will be used in a master’s thesis, as well to complete the fission yield data of 25 MeV proton induced fission of ^{232}Th from earlier measurements (D. Gorelov's Doctoral thesis 2015) to bring the results publishable.

1. Background

1.1. Fission yield measurement via ion counting techniques

Determining the complete fission yields is a challenging task, where the production cross section for something like one thousand isotopes should be measured, at least in principle. Different methods have been applied, typically mass separation, chemical separation, and decay spectroscopy in variable combinations. Because of the limitations of these techniques, the most common way to present the fission yields even today is to show the yield fraction per mass number, which roughly corresponds to the cumulative yield of the last member of each decay chain.

The idea of measuring the fission yields via ion counting is not new as such. In [1] from 1972 the isotopic yields of Rb and Cs were measured using a selective ion source, 90 decree sector magnet and counting ions with an electron multiplier. This method allowed determining the independent isotopic fission yields for also stable isotopes such as ^{85,87}Rb and ^{133}Cs.

Applying the direct ion counting to determine the independent yields required an element selective ion source, since resolving the isobars using sector magnets is technically too challenging. The required mass resolution R typically exceeds 10^4, which can be reached only with specifically designed, very high-quality sets of sector magnets and basically energy spread free ion sources. The development of Penning-trap based mass separation techniques in the beginning of millennium changed the situation, by providing an order of magnitude higher mass resolution of $R \gtrsim 10^5$ with sideband cooling technique in the purification trap. During the recent decade, the multireflection time-of-flight mass spectrometers have achieved a comparable mass resolution. An even higher mass resolution $R \approx 10^6$ can be achieved with Penning trap using the Ramsey cleaning technique, although the identification of the mass peaks becomes more complicated. Finally, the PI-ICR technique can provide a resolution $R \gtrsim 10^6$.

Already the sideband cooling technique allows identifying majority of the fission products by their mass. In the following, the mass spectra produced using the sideband cooling technique are called “sideband spectra”.

1.2. Fission yield measurement using gas stoppers

When identification of fission products can be done unambiguously by mass measurement, the requirement of a selective ion source for mass spectroscopic measurement disappears. Instead, it is favorable to have a universal source of ions to be able to measure the yield of all fission products with the same device. The gas catcher (or ion guide) techniques provide this property. In these methods, the reaction products from fission (or from any other nuclear reaction) are stopped in noble gas, where they are kept as ions until transported to mass analysis.

1.3. Fission yield measurement at IGISOL using the sideband cooling technique

At the IGISOL, the yield measurements are performed as follows. The fission products are stopped in a helium gas filled ion guide, from where they are extracted as 1+ ions [2]. The ion beam formed from

the ions is mass analyzed with a dipole magnet. A selected isobar is stopped in a RFQ quadrupole trap [3], where the ions are bunched to be sent to JYFLTRAP Penning trap [4], operated as a high-resolution filter. Each trapped bunch is cooled and centered in the middle of cylindrical trap with gas cooling. Next, the radius of the magnetron trajectory of the ions is increased with an electric dipole RF-excitation. When the ions have been moved in the large (a couple of millimeters) radius, a mass selective electric quadrupole RF-excitation is applied to center the ions. The ions, whose resonance frequency is close to that of the quadrupole excitation, are centered, while the others remain in the larger radius. When the electric field in the direction of trap axis that keeps the ions centered in this "Z-direction" is removed from one side, the centered ions pass through a narrow channel and counted. By changing the quadrupole excitation frequency, a mass spectrum of the fission products is formed, where the center of each mass peak lies at the resonance frequency of the particular ion species. An example of a mass spectrum formed with this sideband cooling technique [5] is shown in figure 4, and in figures 1 and 1B.

For the determination of the independent fission yields from these sideband cooling spectra, the good news is that the intensity (more precisely the height) of the mass peaks in the sideband cooling spectra are directly proportional to the independent fission product yields. Since the ionization mechanism in the ion guide is the fission reaction itself, all the ions arriving to the mass analysis stage are primary fission products, not their beta decay daughters. During the mass analysis, any atomic ion that undergoes beta decay will be completely lost. The beta decay daughters never make it to the ion counting. The processes causing this are explained in detail in [6].

This proportionality is different for different elements. Therefore, the final result are **independent isotopic fission yields**, which are determined by comparing the decay-loss corrected number of counted ions for isotopes of the same element. These yields can be converted to absolute independent yields with additional information on the mass number and/or chain yields. In this respect the measurements are not entirely self-sustaining. Usually, the necessary supporting information is however available.

There are two main weaknesses in the sideband cooling technique: **low duty cycle** and **insufficient resolution** close to the stability and for isomeric states.

Insufficient resolution is a relative term for a technique that can routinely provide resolution between 10^5 and $2x10^5$. Close to the stability the masses however are so close to each other that the mass peaks are overlapping and difficult to resolve. Even if they are resolvable by fitting techniques, the fitting procedure produces high uncertainty in the mass peak intensity, in particular since the shape of the mass peaks is not any analytical function and varies from one Penning trap settings to another.

For isotopic yields the knowledge on the population of different isomeric states is an issue because of the decay corrections for the ion count. The problem arises when the beta decay corrections that need to be applied are very different for different isomeric states. If the lifetimes of both isomeric states are long, which means that the corrections are negligible, or the lifetimes are similar, so that the difference between the corrections is small, resolving of the isomers is not necessary. Whenever the situation is not so lucky, IYR need to be either determined or estimated to get the most accurate decay corrections. If the IYR is taken from literature or from theory, the uncertainty of the achieved results is increased.

The insufficient resolution of the sideband cooling technique can at least in principle be corrected by using PI-ICR technique. How the yields are determined in practice from the PI-ICR spectra is the main research question of the current work.

The second, even bigger issue is the low duty cycle. In the sideband cooling, the mass spectrum is formed by scanning the spectrum mass point by mass point, or technically, frequency by frequency. A typical mass peak width in sideband cooling spectra is 10 Hz FWHM, which is mass independent. Because the quadrupole excitation frequency in the trap is inversely proportional to the mass, the mass resolution factually decreases towards higher masses.

Close to mass A = 100 the 10 Hz FWHM corresponds to about 1 MeV. The measured mass range in a mass number from the stability line to the neutron richest fission products is of the order of 30 - 35 MeV in this region. To be able to follow the position of a mass peak, its possible shape evolution, to discriminate false counts, and be able to resolve neigbouring mass peaks by a fit, one should have at least 5 - 6 data points within the FWTM region of the peak. In the A=100 region this corresponds to a frequency step of about 2 Hz and consequently, 150 data points for scanning the entire mass number. In a typical scanning procedure, measuring of one data point takes ~400 ms and scanning through of all 150 frequency points just once approximately a minute. To avoid pile-up in the ion counting device (typically an MCP) the number of ions in a single pulse hitting MCP should preferably be limited below 20. An MCP detector can take a continuous rate of the order of 10^7 cps, but since the ion bunch arrives in the detector within microseconds, the signals start pile-up easily. 10^7 cps is a significant value, but converts to only 10 counts per µs. In a half an hour run where the pile-up limit has not been exceeded, the number of counts in the biggest peak's most intense data point is therefore less than 600 and the number of counts in the entire peak less than 2500.

In other words, the duty cycle in the sideband cooling spectrum measurement is less than 2%. In one hour scan any single isotope's yield is measured roughly one minute. This can be slightly improved by distributing the measured frequency points unevenly so that they were sparser between the mass peaks. The improvement would however be small compared to the efficiency with which the mass spectra can be produced utilizing the Multi-Reflection Time-Of-Flight (MR-TOF) technique.

1.4. Multi-Reflection Time-Of-Flight (MR-TOF) technique

A Multi-Reflection Time-Of-Flight (MR-TOF) device is a precision mass spectrometer, where the flight path is extended by trapping the studied ions between isochronous electrostatic mirrors [7]. The number of revolutions in the Jyväskylä MR-TOF in the measurements done in this work varied from 800 to 950, which provided mass resolution R of the order of $R \approx (1.5 – 2.0) \times 10^5$.

A mass spectrum is measured with the MR-TOF as follows. The mass separated ions of a certain mass number are trapped in the RFQ cooler, cooled, and released as a sharp pulse to the MR-TOF. The pulse is accelerated to high enough energy that the ions can pass the voltage barrier of the entrance side electrostatic mirror to the drift tube between the mirrors. While the pulse is moving in the drift tube, the drift tube voltage is lowered so that the ions become trapped between the electrostatic mirrors in both ends. In the acceleration with electric field each ion gets the same energy; since they have different masses, they have different velocities and become separated. After an appropriate number of revolutions, the drift tube voltage is increased, and the ions are detected with a multichannel plate (MCP) detector. The time-of-flight spectrum (see figure 1) is constructed from the time difference between the release of the beam bunch and the signals from ion implantation in the MCP detector. The appropriate number of revolutions depends on the required resolution and the possible contaminant ions released in the bunch – any ions on different lap than those of interest are not allowed hit in the detector at the same time when the time of flights of ions of interest are recorded. For this reason, the optimal number of revolutions can be and is different in each mass number.

The potential of the MR-TOF lies in its rapidity. It provides the same mass resolution as the sideband cooling much faster. The resolving time is the same as the time of flight through the device, a few tens of milliseconds. The repetition rate of the measurements in this work was 20 Hz. This reduces dramatically the decay losses of the shortest-lived nuclei as compared to sideband cooling, where the repetition rate in high resolution measurements is of the order of 2 – 3 Hz. In addition, in contrary to Penning trap, MF-TOF is not a scanning device. While sideband cooling requires 100 – 150 bunches to be analyzed to get an isobar scanned through once, MR-TOF determines the time of flight and thus the mass of every ion in every bunch, as long as the MCP signals do not start pile up. The rate limit of the MR-TOF is essentially the MCP capability to resolve the signals of the ion implantations.

1.5. PI-ICR technique

The main idea of the PI-ICR technique is to imprint ions' motional phase onto a position-sensitive ion detector [8,9]. The ions are collected and cooled in the purification trap, from where they are released to the second, precision trap. There the ions are first excited to a large cyclotron orbit with a fast, 1 ms long, dipolar excitation pulse at ions cyclotron frequency (ν_+). The short duration ensures that all ions are excited even if their cyclotron frequency (or mass) is far away. The ions are then let to rotate for a certain time so that different ion species develop large enough phase difference to be distinguishable from each other. Finally, a quadrupolar pulse is applied to convert the cyclotron motion to magnetron motion to preserve the motional phase in the extraction. The extracted ions are let to hit the position sensitive ion detector with their phase recorded.

The insert of figure 1 shows an example of the separation of isotopes using PI-ICR technique. The PI-ICR spectrum in figure 1 is taken in the current measurements. The two-dimensional spectrum can further be projected on the rotation angle, allowing normal one-dimensional fitting techniques to be applied to determine the ion count also in the still overlapping "blobs" in the two-dimensional spectrum [10].

Two possible approaches to utilize the PI-ICR technique in the fission yield measurements was envisaged beforehand. In the "magnifying" approach, the sideband cooling or MR-TOF spectra are used to deduce the total isotope yield ratios, while the PI-ICR technique is applied to resolve more precisely the detailed composition of the mass peaks in sideband cooling spectra. The measurements are technically quite like the IYR measurements [11]. Beforehand, this seemed the more reliable method to improve the measurement accuracy.

Another approach, "filtering", is however in principle more straightforward. It is based on comparing the count rates in *different* PI-ICR images – for example, the counts in the phase images of ^{119}Cd and ^{119m}Cd in figure 1 are to be compared to the counts of ^{120}Cd in a similar image taken in mass number A = 120, to get yield ratio between ^{119}Cd and ^{120}Cd. This approach may however be vulnerable to a small, unintentional, detune of each of these devices, which may result in erroneous yield ratio between the isotopes. Next, the reproducibility of the measured yields, and their tolerance to variation of adjustment will be experimentally tested.

2. The test run

The ways to improve the fission yield measurement techniques were tested experimentally at the IGISOL facility with JYFLTRAP Penning trap. The proposal for the JYU-ACCLAB program advisory committee is attached to this report (Appendix 1). To maximize the scientific output of the experiment, the tests were combined with finalizing the data of 25 MeV proton induced fission of ^{232}Th, studied earlier [12] thoroughly but not sufficiently for publishing the results, with sideband cooling technique.

The proposal was approved in March 2021 PAC meeting (M19 of SANDA). After that, the execution of the experiment was first delayed because the scheduled JYU-ACCLAB maintenance shutdown had to be extend by a few months, and later due to a major failure of IGISOL on-line helium gas purification system that led to eight months long revision of the safety procedures and to some mechanical changes of the gas feeding system.

The test run at IGISOL took eventually place in March 2023 (M43 of SANDA).

By March 2023 the Jyväskylä MR-TOF device had been commissioned with off-line ion sources as well as on-line experiments. It was thus natural revise the research plan and include fission yield measurements with MR-TOF device in the experimental run, which indeed provided very successful results.

2.1. MR-TOF measurements

The operation principle of the MR-TOF is described above. In figure 1 a mass spectrum of mass number A = 104 taken with Jyväskylä MR-TOF is shown. For comparison, a sideband spectrum of the same mass number that was measured with the same fission reaction in 2014 is shown. In the displayed case, the resolution of MR-TOF determined from the FWHM of mass peaks is better than that of sideband cooling, however, the MR-TOF peak shape includes the tail on the long time of flight side. The tails produce additional uncertainty to the determination of the mass peak intensity, since the number of counts in the part of the tail under the adjoint heavier peak need to be estimated. Even the total intensity of the tail is however small, and the uncertainty due to the tail typically less than per mille. Figure 2 shows the sideband and MR-TOF spectra in the linear scale.

The fission yields are determined from the MR-TOF spectra in a similar way as from the Penning trap mass spectra since the transmission efficiency of ions from the ion guide to the counting depends on the element of ions in the MR-TOF measurements as well. The MR-TOF has an additional correction to consider, the scattering losses of ions in the MR-TOF, which depend on the length of the flightpath. This length cannot be equalized since it has impact on the MR-TOF resolution and overlapping of the spectrum with impurity ions. An estimated half-range in which the number of counts drops to 50 % is ~600 revolutions. A more precise correction factor for the scattering losses may turn out to be necessary, which investigation should be possible to perform off-line.

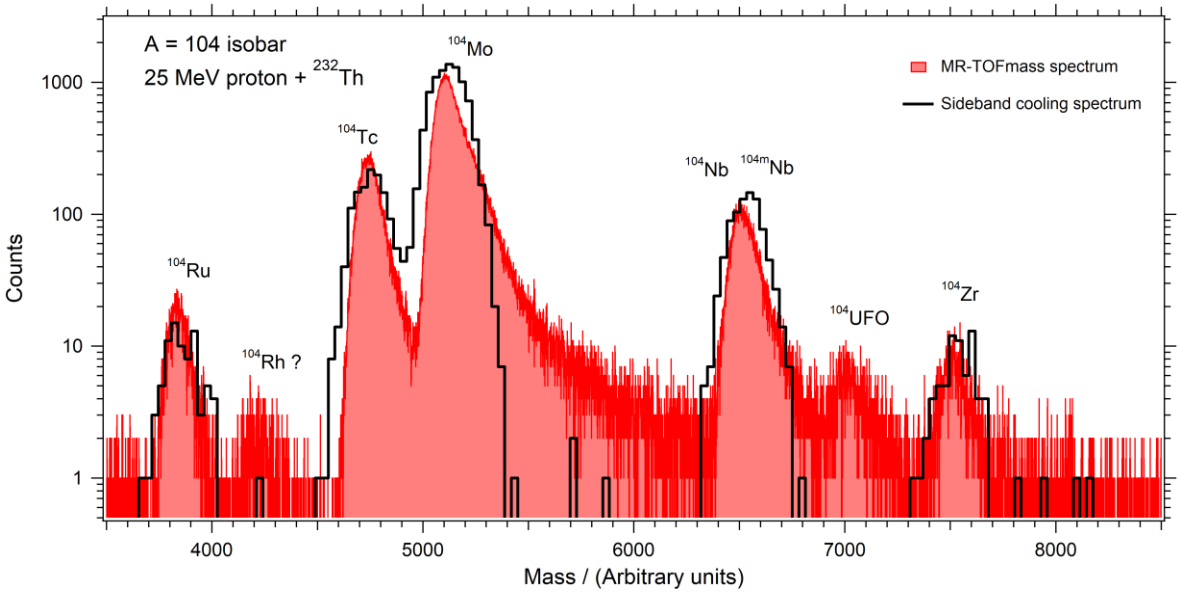

Figure 1. The MR-TOF mass spectrum (time-of-flight, red) measured in 30 minutes in mass number A = 104. For comparison, the sideband spectrum (frequency, black histogram) of the same mass (from 2014) is overlaid. The sideband cooling spectrum combines four runs with total measurement time of about 2 hours at A = 104. The spectra are shown without converting the original units to mass value, so the time of flight increases from left to right and frequency from right to left. The MR-TOF spectrum has slightly better resolution, but the peaks have a tail of scattered ions on the longer ToF/higher mass side of the peaks, while sideband cooling spectrum has regions of zero counts between the peaks. The MR-TOF spectrum consists of 5000 ToF points, the sideband spectrum of 150 data points. The ^{104}Tc mass peak has ~1200 counts in the sideband spectrum, ~32000 in the MR-TOF spectrum. In this particular case, MR-TOF appears about hundred times more efficient.

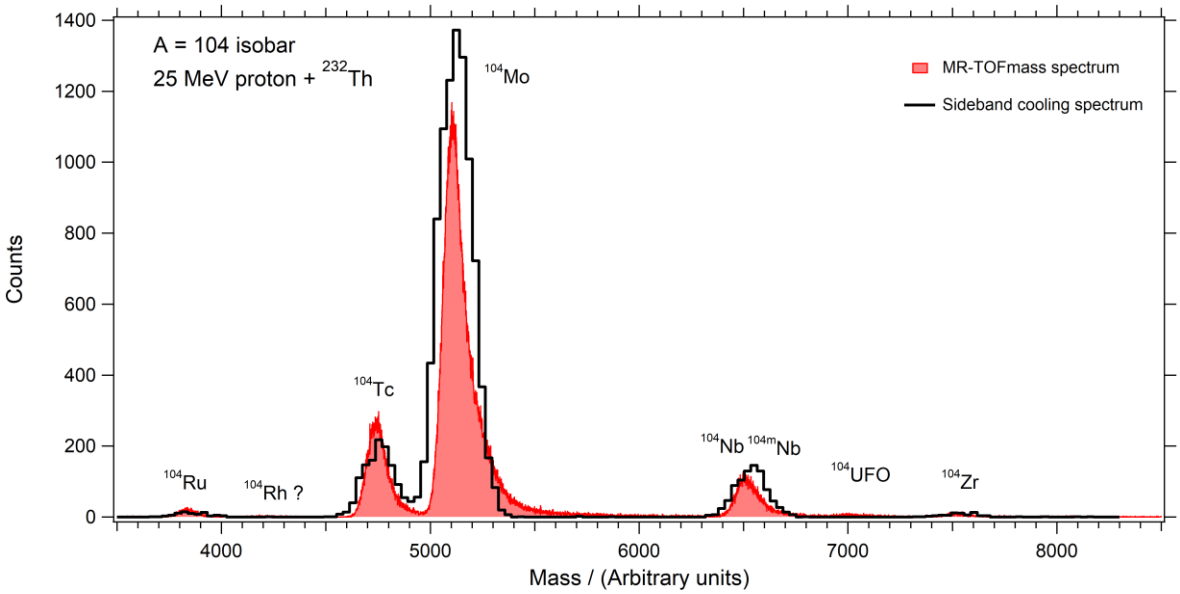

Figure 2. Same as figure 1 on linear scale.

As pointed above, there is a significant difference between the duty factors of the sideband cooling and MR-TOF. If the MCP count rate limitations are not exceeded, there is no principal reason that all

the ions in every released ion bunch that belong to isobaric chain of interest could not be detected. In the spectra shown in figure 1, the ^{104}Tc mass peak has ~1200 counts in the sideband spectrum that has collected in two hours, ~32000 in the MR-TOF spectrum collected in a half an hour. This difference has nothing to do with the production rate of the fission products, because that had to be limited to what the ion counting can tolerate. In this particular case, the bunch rate in sideband cooling was 2 Hz, in the MR-TOF measurement 20 Hz. Sideband cooling was letting ^{104}Tc ions through the trap to be counted at 10 frequencies out of 150 measured ones. This matches well to the factor of 100 between the rate of detected ^{104}Tc ions.

2.2. Effect of RFQ cooler and buncher

To study the possible impact of the RFQ on the isobaric charge distribution, the work included measurements in some selected mass numbers (A=80, 86, 100, 104, 110, 124 and 136) utilizing beta gated gamma spectroscopy in the so-called spectroscopy beam line of the IGISOL. These measurements took place after dipole magnet mass separation, and the isobaric charge distribution of fission products in each mass number represents the situation after the ion guide, before possible charge exchange of the ions in the RFQ. Examples of these distributions are shown in figure 3.

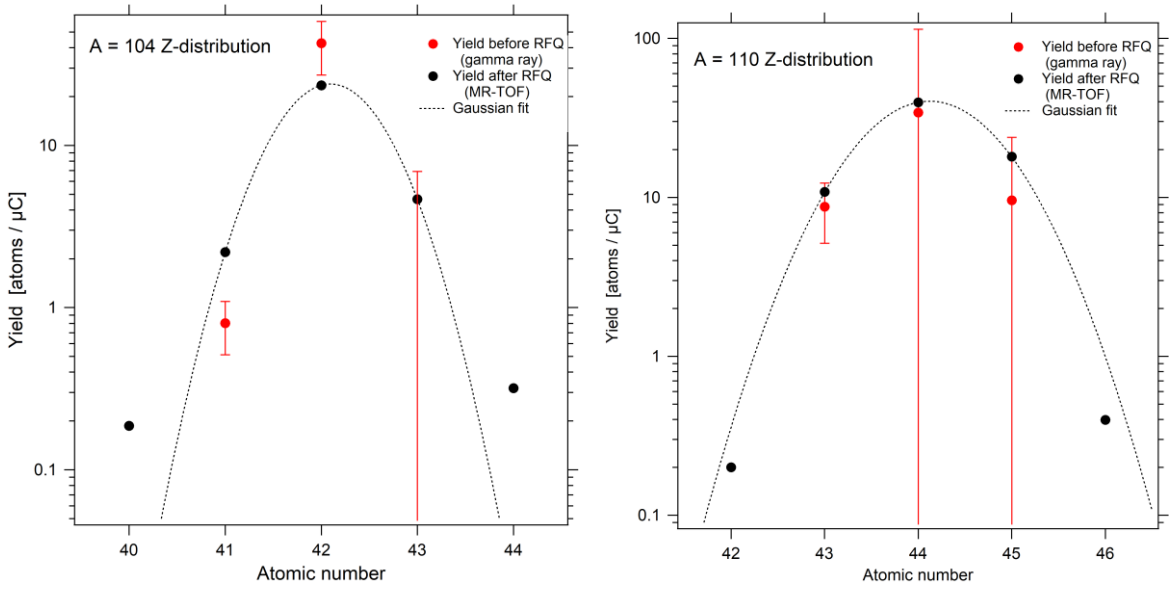

Figure 3. Tentative nuclear Z-distributions before (red points) and after (black) the RFQ cooler. The independent yield of ^{104}Tc determined from the gamma ray data is negative but the upper 1σ limit is 7 atoms/µC, as indicated by the error bar. The large uncertainties are due to uncertainty of the nuclear data, as well as the fact that the independent yields of isotopes need to be corrected for the beta decay feeding of their precursors. The statistical uncertainty of the MR-TOF yields is less than the size of data symbol. The possible systematic trends are sought for.

In the original plan the purpose was to measure yields with gamma spectroscopy both before and after the RFQ cooler. This way the comparison of yields would have been more straightforward since no knowledge of the gamma branching would have been necessary. The gamma setup could not unfortunately be installed after the RFQ, and the post-RFQ isobaric distributions are determined with MR-TOF, which makes the interpretation of the results more challenging. The analysis and interpretation of the results is in progress.

2.3. Testing of "filtering" and "magnification" techniques

The mass spectra to test the PI-ICR aided methods tentatively baptized as "filtering" the direct determination of the fission yields from comparison of 2-dimensional PI-ICR ion image position spectra, and "magnifying", where the PI-ICR technique is applied to determine the detailed composition of the mass peaks in sideband or MR-TOF spectra, were produced as follows. The ion bunch was collected in the purification trap and released to the precision trap for PI-ICR analysis after applying sideband cooling; the ions were thus passing similar mass filtering than in the sideband cooling technique. The pass window need not to be as narrow as in the sideband cooling, because the main mass resolving was provided by the PI-ICR technique. The window was then repeatedly scanned over the frequency region of interest. The ions were not transported from purification trap to the precision trap using a single, wide pass window, since the pass efficiency is different at the edges of the window than in the center. Scanning the pass window equalizes the ion transmission to the precision trap in the region of interest.

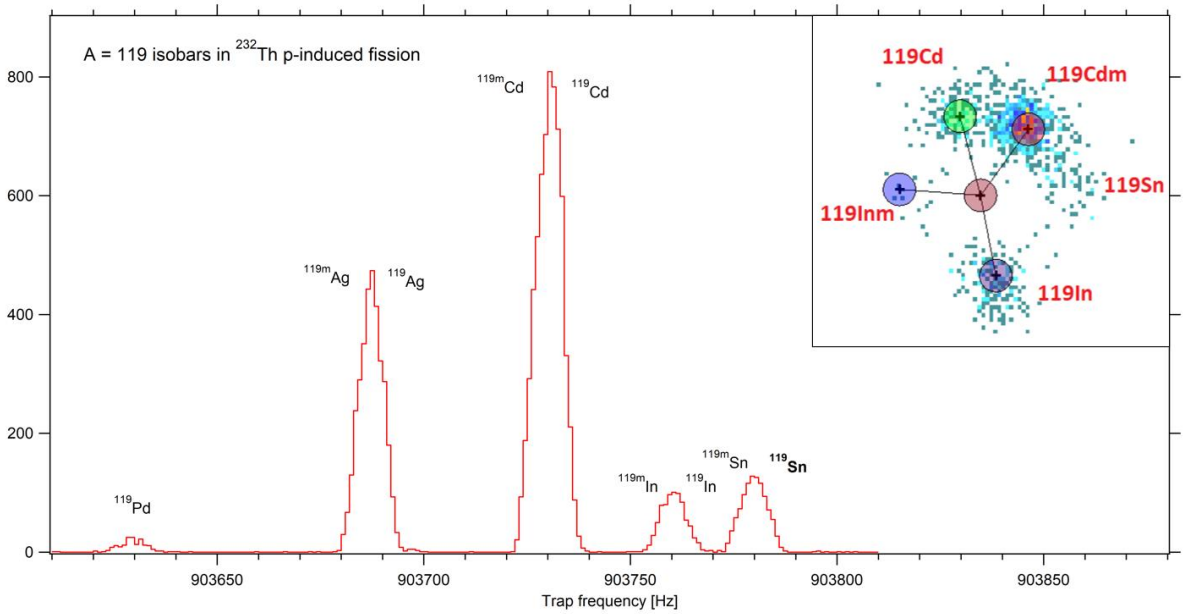

Figure 4. The mass spectrum of A = 119 fission products in 25 MeV proton induced fission of ^{232}Th. In the main figure the separation is made with sideband cooling technique from stable ^{119}Sn up to ^{119}Pd. The ion count is given as a function of the purification trap quadrupole frequency, so the mass increases from right to left. Ag, Cd, In and Sn have isomers at 33, 146, 311 and 90 keV excitation energy, respectively. None on the isomers is resolvable with sideband cooling. The insert shows PI-ICR spectrum of the three lightest mass peaks (Sn, In, Cd) collected when the sideband cooling mass filter window was continuously scanned over the frequency region of interest. Please note that the sideband spectrum shown in this figure is collected in another experiment in 2014 with much narrower transmission band and demonstrates the typical best resolution achievable.

Figure 4 displays the sideband spectrum of A = 119 fission products (data from 2014). Four out of the five isobars are known to have a long-lived isomer so close to the ground state that the masses of these two states cannot be resolved with sideband cooling (or with MR-TOF). The inset of figure 1 shows the PI-ICR phase image spectrum, where ^{119}Cd and ^{119}In isomers are clearly separated. ^{119}Sn isomer is not as clear, but it may be a question of counting statistics. The analysis of these spectra is still in progress.

The phase image spectra such as in the inset of figure 4 allow the use of both “filtering” and “magnifying” methods. Technically it was important to verify that it is possible measure simultaneously several isotopes from a relatively wide mass range. The PI-ICR measurements utilize the precision trap, for which the ion bunch is prepared with the preparation trap. Passing ions with a wide frequency band from the preparation trap to the precision trap with equal efficiency is not an obvious task. It was realized by scanning the preparation trap frequency to generate a wider equally efficient transmission band.

Tentatively it seems that “filtering” is technically possible, but not necessarily feasible. The transmission of ions is sensitive and may not be sufficiently similar in different mass numbers, but the analysis is going on.

Nevertheless, what actually makes the “magnifying” method more appealing are the very promising results from the MR-TOF measurements. As up to 100 times more effective than sideband cooling (see figure 1), not yet considering the effect of reduced radioactive decay losses of ions in faster separation, the MR-TOF technique allows faster and statistically more accurate yield measurements. Since the MR-TOF resolution is similar to that of sideband cooling, it will run into difficulties with precisely same irresolvable mass peaks. As seen from the inset in figure 4, these mass peaks however are resolved in PI-ICR phase images, suggesting that combination of MR-TOF and PI-ICR would be the best solution for the independent fission yield measurements. The analysis is in progress.

3. Conclusions

In a test experiment at the IGISOL facility in the Accelerator laboratory of University of Jyväskylä in March 2023 the feasibility of new methods to determine independent fission yields. The methods tested were the use of a Multi-Reflection-Time-Of-Flight (MR-TOF) device for the mass separation of fission products and determination of the fission yields by ion counting, and applying the Position Image Ion Cyclotron Resonance (PI-ICR) technique to resolve the yields of fission products whose masses are too close each other to be resolved with either by MR-TOF measurement or by Penning trap by the sideband cooling technique.

The accumulation of the counting statistics of the fission product yields with the Multi-Reflection-Time-Of-Flight device turned out to be two orders of magnitude faster than with the sideband cooling. The result was expected in the sense that MR-TOF is not a scanning device while the Penning trap in the mass filtering mode is. The mass resolution of the Jyväskylä MR-TOF appeared to be like sideband cooling, ~1.5×10^5 as measured from the FWHM of the mass peaks.

The image spectra collected with the Position Image Ion Cyclotron Resonance (PI-ICR) technique showed that it is possible to resolve simultaneously several isotopes with relatively different masses with equal efficiency. The bottleneck here is not the PI-ICR technique but passing the ions with different mass from the purification trap to the precision trap equally efficiently. This would allow using of the both techniques that are in principle possible: “filtering”, which means the direct comparison of the ion counts in the PI-ICR image position spectra taken in different mass numbers, and “magnifying”, meaning that the yields are still determined from the mass peaks in in sideband or MR-TOF spectra, but the PI-ICR position images are used to determine the detailed composition of those mass peaks.

The analysis of the data is in progress. Tentatively it seems that the “magnifying” technique is more appealing, which is because it can be combined with the MR-TOF measurements, while the benefits of “filtering” lie in that all yield measurements could be performed with PI-ICR technique alone.

In addition to the development of the fission yield determination techniques, the achieved data will be used to complete the fission yield data of 25 MeV proton induced fission of ^{232}Th from earlier measurements [12] to bring the results publishable. A master’s thesis using the data from this experiment is in preparation.

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Appendix:

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Proposal to JYFL-ACCLAB PAC

Independent fission yields in 25 MeV proton induced fission of ^{232}Th

A test for applying PI-ICR technique to fission yield measurements

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Abstact:

In this proposal we apply for 6 days of beam time to investigate the applicability of PI-ICR technique to the fission yield measurements. The technique has already successfully applied to IYR measurements, where the compared isomers are analysed simultaneously [8-11]; now the yields of isotopes in different mass numbers will be compared. The test case would be 25 MeV proton induced fission of ^{232}Th, whose yields have already been studied at the IGISOL [12] using sideband cooling method [14]. The data has not been published as a peer reviewed article, since the extracted yields of Sb, Sn, Te and Xe isotopes seem not to fully agree with each other and need to be remeasured. On the other hand, many yields of the isotopes in the low-mass peak were measured accurately, which allows testing the impact of PI-ICR with a well-known cases. About 2 days of the beam time has been dedicated to the finalization of the ^{232}Th fission yield measurements using the most appropriate method.

1. The fission yield measurements with the IGISOL and the JYFLTRAP

The IGISOL technique can be combined with the JYFLTRAP Penning trap to determine independent fission yields. The concept has been verified in [1-5]. It has so far been used to determine fission yields in the charged particle induced fission, however, there is interest of utilizing the technique on neutron induced fission as well. Improving the accuracy of the fission yield measurements is thus of importance for future studies.

The IGISOL technique is fit to fission yield measurements, since

- all elements are ionized: the ionization is based on the primary ionization in the nuclear reaction
- since the nuclear reaction is the principal ionization mechanism, basically all the ions come directly from the fission; although some beta decay daughters of the fission products accumulated in the ion guide walls can be ionized in the beta decay, this fraction is well below the overall uncertainty of the measurements [2].
- as the first approximation, only 1+ ions are produced, so all the yield is concentrated to single charge state.

In addition,

- the fission products can be unambiguously identified by their mass with JYFLTRAP; this proposal is about improving the identification of the difficult cases that require higher mass resolving power than previously available
- the daughter nuclei of fission products, which decay during the transportation from production to detection are **not** transmitted to the ion counting : a simple decay loss correction is sufficient for the analysis

The whole analysis involves three gas-filled cells: the ion guide, the cooler/buncher quadrupole trap, and the purification Penning trap of JYFLTRAP. Due to charge exchange reactions, the total

transmission efficiency from the target to the final detection depends on the element. The efficiency is however same for all the isotopes of the same element, which allows measuring beta decay independent, isotopic fission yields. The absolute yields can be extracted from these, if the mass number fission yields of the studied fission reaction are known. This is described in detail in [5].

1.1. To be improved:

The largest shortcoming in the fission yield measurements is that the sideband cooling technique utilized to extract the mass spectra of the fission products does not provide sufficient mass resolution in all regions of nuclear chart. Typically, the masses of fission products are close to each other near the beta stability line, and for the isotopes further from stability near double-magic ^{132}Sn (Figure 1). It is worth noticing that stable isotopes are produced in fission as well, and the ion counting with JYFLTRAP allows detecting and identifying them. The yields of the isotopes close to stability are valuable for the analysis, since they provide information how the yield distribution behaves on the stability side of the maximum. Usually their mass peaks are however difficult to resolve in the analysis of the mass spectra.

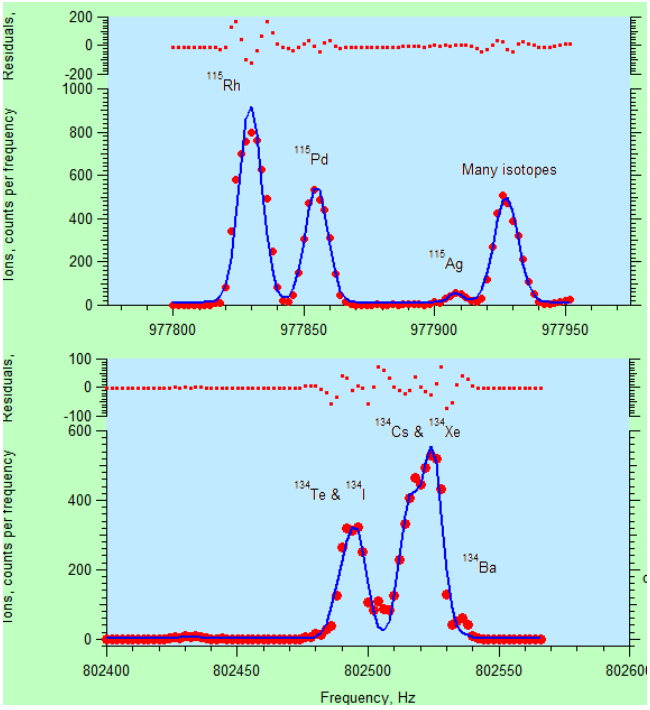

Figure 1. Mass spectra of fission products in A=115 and A=134. The displayed frequency range is the same 200 Hz in both panels. The mass spectra are shown as a function of Penning trap frequency (inverse mass), thus the ion mass increases to the left. In mass A=115 the fission products are so well separated that their yield could be determined even without a fit. The rightmost peak consists mostly of ^{115}In. In A=134 a seemingly similar peak as the single mass peaks of ^{115}Rh or ^{115}Pd consists of at least two mass peaks: ^{134}Te and ^{134}I. In addition, a considerable fraction of ^{134}I may be produced as ^{134m}I. The blue lines in the spectra represent fit of several Gaussian shape peaks in the spectrum. The resolution of the spectrum simply does not allow very accurate fits to the data points.

The isotopes in the low-mass peak of the fission yield distribution typically lie well off from stability, and they are easy to resolve. In the high-mass peak of fission yield, instead, the isotopes tend to be close to beta stability line, or close to ^{132}Sn. Their mass difference tends be small, and, in addition, the

resolution of the sideband cooling technique, or any Penning trap separation technique decreases towards heavier masses [2]. The mass peaks start to overlap, and resolving their intensity even by fit becomes challenging (Figure 1).

Improved mass resolution would also be beneficial for the analysis of the fission yields of such isotopes, where fission populates close-lying isomers. In many cases, the sum yield of the isomers for a particular isotope can be deduced sufficiently accurately without resolving the individual isomers. However, if any of the isomers is short-lived, it is necessary to apply decay-loss corrections to deduce the actual yields. When the calculated decay-loss corrections for the unresolved isomers are very different, the uncertainty of the yield becomes large. Knowing the isotope yield ratio would significantly improve the accuracy of the results.

A way to improve the accuracy of the fission yield measurements in these critical regions is utilizing the PI-ICR (phase-imaging ion-cyclotron resonance) technique, capable to resolve mass differences up to of the order of tens of keV/c^2 [6,7]. PI-ICR technique has already been successfully utilized in isotopic yield ratio (IYR) measurements at IGISOL [8-11]. In the IYR measurements, the situation is simplified in the sense that the compared isomers are analysed simultaneously. In the proposed experiment, the yields of isotopes in different mass numbers will be compared. The purpose of the proposed experiment is to investigate how the PI-ICR technique could best applied to improve the fission yield measurements.

1.2. Test case: 25 MeV proton induced fission of ^{232}Th

An appropriate test case is the independent fission yields in 25 MeV proton induced fission of ^{232}Th, which have been studied using JYFLTRAP and the sideband cooling technique already a few years ago. The deduced yields formed a considerable fraction of the doctoral thesis of Dmitry Gorelov [12]. The study was originally motivated by

- studies of fission dynamics, in particular charge polarisation of the hyperdeformed states, resulting in super-asymmetric fission fragment mass division
- nuclear data needs for nuclear waste transmutation calculations by improving the reliability of fission models in the intermediate energy
- development of RNB facilities by comparing the yields from intermediate energy proton induced fission of thorium and uranium. The mass number yields seem to favor the use of thorium in the region A < 95, even taking into account that the total fission cross section of thorium is 20% lower than that of ^{238}U, the main constituent of natural uranium (Figure 1). The mass number yield however does not yet provide information of the independent yield of the individual isotopes.

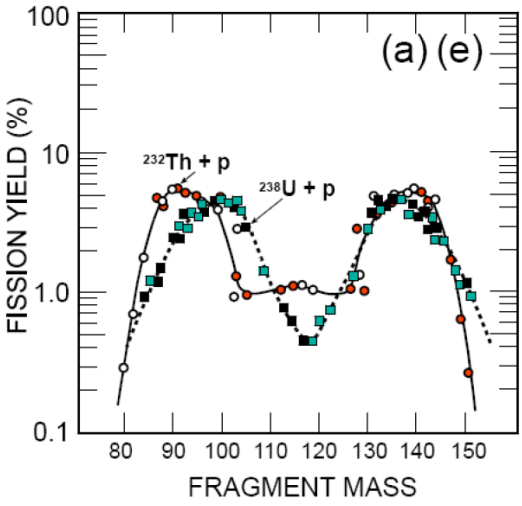

Figure 2. Experimental mass number fission yields of 25 MeV proton induced fission of ^{232}Th and ^{238}U. Data from [13].

The data in Gorelov's thesis have not been published as a peer-reviewed article so far, mainly because of some critical inconsistencies in the ^{132}Sn region. Despite a very careful analysis, the deduced independent fission yields of Sb, Sn, Te and Xe are not consistent. This is mostly due to insufficient resolving of the mass peaks, which becomes even more difficult, because these isotopes have close-lying isomeric states, increasing the number of overlapping mass peaks. On top of this becomes the contamination of stable isotopes. In the ^{132}Sn region and in the high-mass peak in general, significant amount of stable isotopes are produced directly in fission. In addition, ions of the stable isotopes are present in the IGISOL beam as impurities. The experience from the proton induced fission of natural uranium [5] shows that assuming the natural abundances for the stable impurity beam, results in plausible fission yields. To be able to make the correction for the stable impurities, the mass peaks of the stable isotopes should be first resolved from the overlapping mass peaks of other fission products. The ^{nat}U yield study would have benefited from such a better resolving of the mass peaks as well. The both measurements were in this region in the limit of the side band cooling technique resolution. The ^{nat}U data was considered publishable, since it was internally consistent, while the ^{232}Th data was not.

In addition, there are some isotopes in the low-mass peak, whose yield in ^{232}Th fission should be remeasured. These cases include some yield values that are inconsistent with systematics, A=99 that was missing, and some overlapping mass peaks that could be resolved. Excluding all these data from the final conclusions does not seem to alter the overall results. Nevertheless, time allowing, also these yields could be improved. On the other hand, the most of the yields of the isotopes in the low-mass fission peak were measured accurately, which allows testing the impact of PI-ICR with a well-known cases.

All this makes 25 MeV proton induced fission of ^{232}Th an excellent test case. The yields in the low-mass fission peak are known with a good accuracy from the work so far, providing a comparison to the results achieved with PI-ICR technique. In addition, if the yields for the isotopes in the ^{132}Sn region, and more generally in the high-mass fission peak are successfully extracted, the measurements will complete this yield data set.

2. Proposed experiment

2.1. Production and dipole magnet separation

The fission will be induced by 25 MeV protons in a at least 10 mg/cm^2 thick natural Th target, installed in a standard fission ion guide. About 1% of the fission products are stopped in helium, where their charge state rapidly reduces. Because of the high ionization potential of helium, a large fraction (5-10%) of fission products remain as ions until they are swept out of the ion guide. The majority of ions is concentrated on the 1+ charge state. The helium gas is removed with differential pumping; an ion beam is formed from the ions with electric fields. The survival probability of ions depends on the element, but is the same for all isotopes of the same element.

The first step of the counting of fission products is mass separation with a dipole magnet. The resolution of the IGISOL magnet is of the order of 500, which is sufficient to separate isotopes with the same mass number from the neighbouring mass. The spatial resolution of the beam is sufficient to focus entire beam into the RFQ cooler.

The yield of fission products from the ion guide is continuous, while the Penning trap operates with short ion bunches. The cooler/buncher quadrupole trap is used to bunch the fission product ions. The ions are stopped in the cooler filled with low pressure helium gas, which is used to reduce the kinetic energy spread of the ions. Since the cooler is a gas filled trap, gas chemistry, in particular the charge exchange reactions, can be expected to modify the composition of the ion beam of the fission products. The transmission of the cooler thus depends on the element, but again, the transmission can be expected to be the same for all the isotopes of the same element.

2.2. The impact of the cooler transmission to fission product ensemble distribution

However, the elemental dependency of the cooler transmission has not been systematically studied. The basic assumption that there is no significant difference between the transmission of the isotopes of the same element is consistent with the fission yield measurements so far. RF quadrupoles, however, have a small dependency on the mass for transmission. It is thus proposed to use the fission products to study, to which extend the cooler modifies the composition of the IGISOL beam. A possible way to analyse the IGISOL beam before and after the RFQ cooler is gamma spectroscopy. Such a measurement is quite straightforward, since the gamma decay branching plays no role. It is important to measure the gamma activity before and after of the cooler with well enough known geometries, preferably having the same efficiency. An important point is to use the same means to analyse the beam before and after the cooler.

2.3. MR-TOF filtering

The next step is mass filtering with a multi-reflection time of flight (MR-TOF) separator, installed in the beam line before JYFLTRAP in 2020 and still in commissioning phase. The MR-TOF allows the pre-selection of the mass of the ion bunch that is injected in the Penning trap. It reduces the space charge due to isobaric background in the purification Penning trap. The time-of-flight spectrum of the MR-TOF separator also has a similar mass resolution as the mass spectra that are measured with the purification Penning trap using the sideband cooling technique. This allows measuring the fission yields with the MR-TOF alone.

At the time of the execution of these experiments, if available, MR-TOF can provide comparable mass resolution to the purification Penning trap. In such case, the MR-TOF will be used to generate fission product mass spectra up to $R{\sim}10^5$, instead of the sideband cooling.

2.4. Sideband cooling in the purification trap

Finally, the ion beam is injected to the JYFLTRAP's purification trap and mass analysed with higher resolving power ($R \sim 10^5$). The purification trap is essentially a mass filter. The bunch of ions is trapped in the cylindrical trap. The variation of the energy of the ions is reduced and the ions are centered in the trap with gas cooling. Next, the radius of the magnetron trajectory of the ions is increased with an electric dipole RF-excitation. When the ions are moved in the large (a couple of millimeter) radius, a mass selective electric quadrupole RF-excitation is used to center the ions. The ions, whose resonance frequency is close to that of the quadrupole excitation, are centered, while the others remain in the larger radius. When the electric field in the direction of trap axis that keeps the ions centered in this "Z-direction" is removed from one side, the centered ions pass through a narrow channel into the precision trap, where the ions can be further manipulated. Alternatively, the ions can be passed through the precision trap and counted. By changing the quadrupole excitation frequency, a mass spectrum of the fission products is formed, where the center of each mass peak lies at the resonance frequency of the particular ion species.

2.5. PI-ICR technique principle

The new step here is the PI-ICR technique, that utilizes the precision trap. The main idea is to imprint ions' motional phase onto a position-sensitive ion detector [6,7]. The ions are first excited to a large cyclotron orbit with a fast, 1 ms long, dipolar excitation pulse at ions cyclotron frequency (ν_+). The short duration ensures that all ions are excited even if their cyclotron frequency (or mass) is far away. The ions are then let to rotate for a certain time duration so that different ion species develop large enough phase difference to be distinguishable from each other. Finally, a quadrupolar pulse is applied to convert the cyclotron motion to magnetron motion to preserve the motional phase in the extraction. The extracted ions are let to hit the positionally sensitive ion detector with their phase recorded. Figure 3 shows an example with 3 states in ^{130}In.

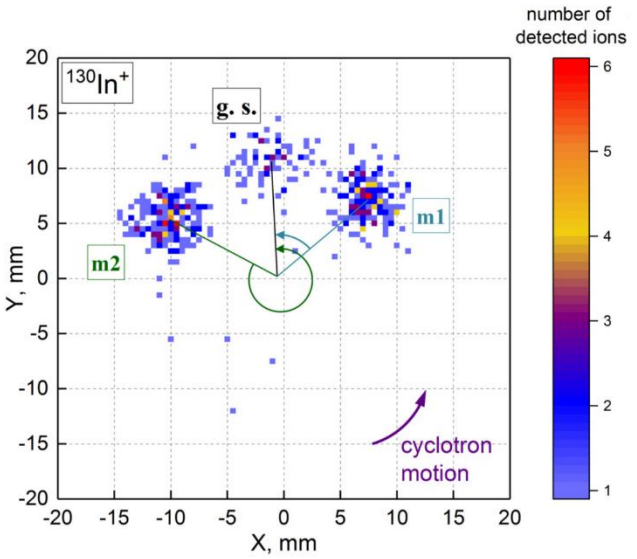

Figure 3: Separation of three isomeric states in ^{130}In. Ions were let to accumulate the phase difference for 320 ms. Figure from [15].

3. Applying the PI-ICR technique

In the independent fission yield measurements the direct yield of each isotope in fission is deduced. The first step is extracting the yield ratio between two isotopes. The ratio measurements can be arranged different ways. Either measuring the ratio always with respect to the same isotope and thus having a common reference, or measuring always the ratio between isotopes in neighboring masses. Because there are several gas filled cells involved in the measurement - the ion guide, the cooler/buncher, and the purification trap - which all can be expected to be elementally selective, the yield ratio measurement is made for different isotopes of the same element. The absolute independent yields can be calculated, if the mass yields are known from other sources, see reference [5].

Two possible scenarios, how the fission yield measurements could be improved with the PI-ICR technique, will be investigated.

3.1. Direct PI-ICR filtering

The most straightforward way of deducing the yield ratio of two isotopes is setting all mass selective devices - the dipole magnet, the MR-TOF (if utilised), and the purification trap - to the optimal transmission for each studied isotope. This ensures that no matter the ion species, the intensity (transmission) losses are the same for all ions. The phase imaging in the precision trap is used to have optimal separation of the isotope of interest from the other species passing to the precision trap. This approach may be vulnerable to a small, unintentional, detune of each of these devices, which impacts differently to different isotopes. The element-dependent effects clearly seem to cancel out, when the yield distribution is determined to the isotopes of the same element. The effect of the slightly different tuning for each isotope can result in erroneous yield ratio between the isotopes. It is worth of noticing that since the isotopes have different masses, precisely same settings cannot be used.

This approach will be tested with different combinations of isotope pairs. The simplest one is yield ratio between two well-separated, single mass peaks. In this case, PI-ICR can be seen as an unnecessary complication. However, the deduced yield ratio should be the same as yielded from plain sideband cooling (or MR-TOF) spectra. The most complicated case is the yield ratio between isotopes that both need to be resolved from a nearby isotope. The result of this case may be difficult to verify, except maybe with gamma spectroscopy. The most important issues are, however, the reproducibility of the results, and their tolerance to variation of adjustment.

3.2. PI-ICR magnifying

Another approach is to use the intensities from the mass spectra produced with the sideband cooling technique (or with MR-TOF) to deduce the total isotope yield ratios, and use the PI-ICR technique only to resolve the individual (isomer) peaks. The PI-ICR is thus used as a magnifying glass or a microscope to get more detailed view of the composition of the ion sample. The measurements are technically quite similar to the IYR measurements [8-11]. The transmission band of the purification trap has be adjusted so wide that the transmission is equal to all the isotopes that form the unresolved mass peak. Beforehand, this seems the more reliable method to improve the measurement accuracy.

3.3. Yields of the 25 MeV p-induced fission of ^{232}Th

In total 6 days beam time is requested. The need of the beam time is itemized below in table 2. The primary goal is investigating, how the fission yield measurements could best benefit from the PI-ICR technique, which has a superior mass resolution as compared to anything. In these tests, the fission products of 25 MeV proton induced fission of ^{232}Th will be used.

These tests are expected to provide improved fission yield values for the studied isotopes. There are however about 40 isotopes, whose yield value should be checked. The fission yield of 13 isotopes definitely have to be re-evaluated. The thoroughly investigation of the method does not require measurement of all these values. About 40 hours of the requested beam time are dedicated for systematic yield measurements with the technique that has turned out in the test to be the most appropriate one. The purpose of these is the finalization of the 25 MeV p-induced ^{232}Th fission yield measurements.

4. Justification of the beam time request

The experiment consists of three tasks:

- systematic investigation of the RFQ cooler transmission using the fission products and gamma spectroscopy
- testing the applicability of the PI-ICR technique for fission yield measurements and comparing two approaches that are called above “PI-ICR filtering” and “PI-ICR magnifying”.
- systematic yield measurements to complete the 25 MeV p-induced ^{232}Th fission yield measurements; the cases that should be remeasured are listed in table 1.

In addition, time that is needed for setting and tuning the cyclotron, the IGISOL and the JYFLTRAP. The usage of the beam time is itemized in table 2.

The fission cross sections are sufficiently high to allow measuring the yields of all the mentioned isotopes. To avoid signal pileup losses, the size of the ion bunch should not exceed a few tens of ions in the sideband cooling mass measurement, and five ions in the 2-dimensional detection in the PI-ICR technique. The estimated measurement time is based on these values.

In total 144 hours or 6 days is estimated to be needed to complete the aforementioned tasks.

Table 1. Isotopes to be remeasured to finalise the yield measurements, see also reference [12].

Mass number	Isotope	Priority	Mass number	Isotope	Priority
82	Ge, As, Br	moderate	118	In	
85	Se		121	Sn	high
96	Rb		122	Sn	high
99	all	moderate	123	Sn	high
100	Mo		126	Sn,Sb	high
101	Mo		127	Sb,Te	high
102	Mo		129	I,Xe	high
103	Mo, Ru		130	I	high
104	Ru		132	I	high
105	Ru	moderate	134	Cs	
106	Tc,Ru	moderate	135	Cs	moderate
107	Ru	moderate	136	Xe,Cs	high
108	Ru	moderate	147	La	
109	Ru				

Table 2. Itemised beam time usage during the experiment.

Beam time needed for	hours	Comments
Cyclotron setting and beam tuning to IGISOL	8	
IGISOL settings and tuning	8	
Cooler/buncher transmission measurements using gamma ray spectroscopy	16	
JYFLTRAP tuning and adjustments	8	
Traditional sideband cooling mass scans for 10 high priority mass numbers and corresponding reference mass á 2 hour	20	These measurements can also be made with the MR-TOF. Depends on, how fast the commissioning of MR-TOF proceeds
Traditional sideband cooling mass scans for 20 low or moderate priority mass numbers and corresponding reference mass á 1 hour	20	Same as above
Tests of direct PI-ICR measurements	24	
PI-ICR resolving of yields of isotopes whose mass peaks are overlapping in purification mass spectra, 20 cases á 2 hours	40	
Total	144	

Summary

6 days of beam time is requested to investigate the best way to utilize the PI-ICR technique in the independent fission yield measurements. The tests will also allow finalizing and verifying the independent fission yields in the 25 MeV proton induced fission of ^{232}Th, which have been studied at the IGISOL earlier.

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