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Developments for neutron-induced fission at IGISOL-4

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ABSTRACT

At the IGISOL-4 facility, neutron-rich, medium mass nuclei have usually been produced via charged particle-induced fission of natural uranium and thorium. Neutron-induced fission is expected to have a higher production cross section of the most neutron-rich species. Development of a neutron source along with a new ion guide continues to be one of the major goals since the commissioning of IGISOL-4. Neutron intensities at different angles from a beryllium neutron source have been measured in an on-line experiment with a 30 MeV proton beam. Recently, the new ion guide coupled to the neutron source has been tested as well. Details of the neutron source and ion guide design together with preliminary results from the first neutron-induced fission experiment at IGISOL-4 are presented in this report.

1. Introduction

During 2010–2012, the Ion Guide Isotope Separator On-Line (IGISOL) facility at the Accelerator Laboratory of the University of Jyväskylä was moved to a new experimental hall and upgraded. The main commissioning phase took place in 2012–2013, as reported in the previous EMIS conference [1]. Access to heavy-ion beams and high-energy light ions from the K-130 cyclotron and intense proton and deuteron beams from a new MCC30/15 cyclotron, extends possibilities for long term fundamental experiments and new developments. The present report includes technical details of the first experiments on radioactive isotope production using neutron-induced fission at the IGISOL-4 facility.

In addition to the start of the experimental program, various new developments have been going on at the IGISOL facility. Fig. 1 presents a schematic layout of the ground floor of IGISOL-4. There are several new features related to the post-trap spectroscopy setup (11). A new spectroscopy beam line (12) has been built for experiments that benefit more from fast and efficient transportation of reaction products rather than from the high mass resolving power of JYFLTRAP. The laser teams from the Universities of Manchester and Liverpool are building new instrumentation for collinear laser spectroscopy. There are improvements in the control system [2], extending the possibilities for ion manipulation with the RFQ cooler (7) and Penning trap facility (9). In addition, several new developments focus on the production of exotic nuclei via the construction of new ion guides (1).

Since the 1980s, neutron-rich medium mass nuclei have been produced at IGISOL via charged particle-induced fission of natural uranium and thorium [3,4]. In [5] it was shown that fission induced by neutrons with an average energy of about 20 MeV might be considered as an alternative method to produce the most neutron-rich species. The independent fission yields calculated with the FIPRODY code [6] for proton-, deuteron- and neutron-induced fission of ^{238}U at projectile energies of 25, 15 and 10 MeV, respectively, and total fission cross-sections from [7] allow one to conclude that in these cases neutron-induced fission is preferable for the production of neutron-rich nuclides. For example, the calculated independent yield of ^{136}Sn is 500 times higher in fission of ^{238}U induced by 10 MeV neutrons than when induced by 25 MeV protons. Due to the new location of the IGISOL facility the high intensity beams of protons (~100 μA) or deuterons (~50 μA) from the light-ion MCC30/15 cyclotron can be utilized for the neutron production. We note however that despite the higher primary beam intensity offered by the light-ion cyclotron and calculated higher yields, these factors must be balanced against the proton-to-neutron conversion efficiency of only a few percent. Therefore, the efforts to

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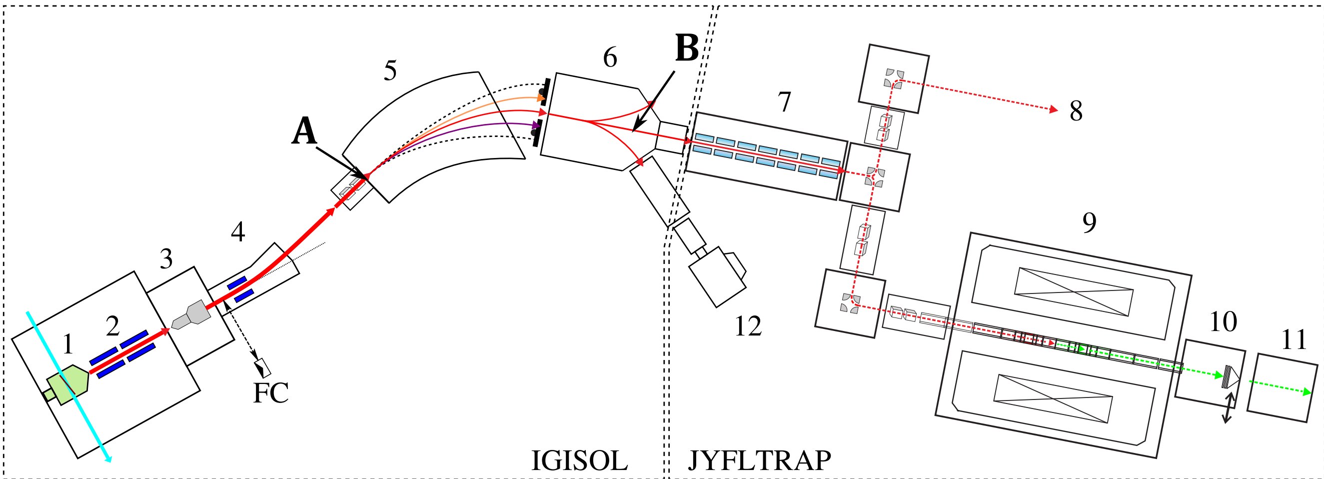

Fig. 1. Schematic layout of IGISOL-4, ground floor. (1) ion guide, (2) sextupole ion guide (SPIG), (3) extraction electrodes, (4) bending electrodes, (5) 55° dipole magnet, (6) switchyard, (7) radiofrequency cooler-buncher, (8) beam line towards a collinear laser spectroscopy setup, (9) Penning trap (JYFLTRAP), (10) microchannel plate (MCP) detector, (11) post-trap spectroscopy setup, (12) β-γ spectroscopy setup. FC is the Faraday cup used to measure the extracted ion guide current in this work. Positions A and B are explained in the text.

realize neutron-induced fission at IGISOL will be motivated by the production of the most exotic neutron-rich species.

At IGISOL we are using either natural uranium (consisting primarily of ^{238}U) or ^{232}Th targets for charged particle-induced fission. The cross sections for neutron-induced fission of these isotopes have a typical plateau structure with thresholds of about 1 MeV and their values vary from hundreds of millibarns to a barn for neutron energies below 10 MeV. Increasing the neutron beam energy increases the total cross section but also moves the distribution of the produced isotopes closer to stability.

In this work, a neutron source based on a thick beryllium target has been designed and tested. Different characteristics of the neutron- and the light-ion beams also have an impact on the design of the ion guide housing the fissioning target material. A new ion guide designed specifically for neutron-induced fission was developed and its performance characterized.

2. Neutron source

Historically two different methods have been used to produce energetic neutron beams at IGISOL; from the break-up of an accelerated projectile such as deuterium or generated in a nuclear reaction [5,8].

The new MCC30/15 cyclotron is intended to be the driver of the neutron production. Because the highest energy of the deuterium beams is limited to 15 MeV, production of neutrons in proton-induced nuclear reactions has been adopted. Light target materials are favorable for neutron production with the 30 MeV protons available from the cyclotron [9]. In addition to the relatively high cross-section and low energy thresholds for the ^{9}Be(p,n) reactions, beryllium was chosen as a target material because of its suitable physical and chemical properties. For example, beryllium has a high mechanical stiffness and low thermal expansion. It is an excellent heat conductor, has a high melting point and specific heat, and a low vapor pressure. Unlike lithium, beryllium does not react with water, which allows direct water cooling of the target.

The construction of the neutron source is based on the thick target concept. This gives the maximum neutron intensity with a broad neutron energy spectrum. In addition, the thick target stops the primary beam, allowing placement of the ion guide with fissile targets directly behind the neutron converter. The experimental setup of the neutron converter (I) and the ion guide (II) is shown in Fig. 2. Both were installed inside the IGISOL vacuum chamber. The neutron converter was connected directly to the cyclotron beam line without additional beam windows.

The full (∼100 μA) intensity proton beam at the maximum energy deposits 3 kW into the neutron source target. The cooling water, pressurised to 7 bar, comes in direct contact with the target, providing an efficient method of heat transportation. The thick beryllium target is sufficiently rigid to isolate the cooling water volume from the cyclotron beam line vacuum.

The primary proton beam, indicated in Fig. 2, is stopped in the water volume. This concept is motivated by the experience of the low-energy neutron source (LENS) group at Indiana University [10,11]. A high-intensity proton beam fully stopped in the target causes degradation of material ultimately destroying the beryllium target. This can be avoided if the neutron production target is made slightly thinner than the stopping range of the projectiles and the beam is dumped in the cooling water.

Protons with energies below the thresholds of ^{9}Be(p,n) reactions (∼2 MeV) do not contribute to the total neutron intensity. About 2 MeV would thus be the optimal exit energy of the protons. Due to the proton range straggling in beryllium the target needs to be at least 0.15 mm thinner than the average range of 30 MeV protons. This corresponds to two standard deviations of range straggling. The thickness of such an optimised target would be 5.7 mm and the exit energy of protons 3.8 ± 1.2 MeV. Such proton energy is already sufficient to activate the cooling water. Therefore, the neutron converter is equipped with a closed cooling circuit. For the prototyping, a thinner 5.0 mm beryllium target was used. The exit energy of protons for this thickness is 11 ± 1 MeV.

The neutron source is installed at high voltage in the IGISOL target chamber and the main body of the closed cooling water system in the basement at ground potential. Cooling water is circulated through plastic hoses with the water conductivity low enough so as not to drain current from the high voltage power supply. The protons stopped in the cooling water have sufficient energy to ionise the water. Therefore, a de-ionizing filter has been installed in the closed loop to keep the water conductivity at the level of several μS/m.

3. Fission ion guide

A new ion guide was designed for preliminary tests of neutron-induced fission. The principle of the ion guide is as follows: the reaction products (neutron-induced fission products) recoil out of

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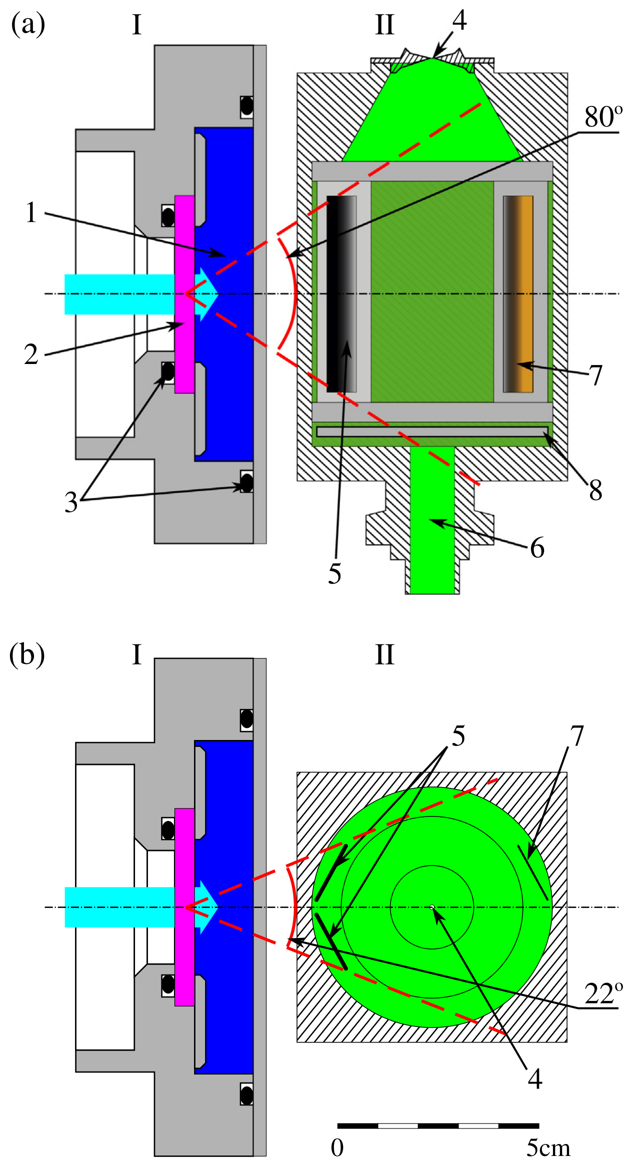

Fig. 2. Schematic views (top view (a) and side view (b)) of the experimental setup in the test of neutron-induced fission at IGISOL facility. These views are based on technical drawings. The common scale is given at the bottom of view (b). The neutron converter (I) is placed in the closest position to the ion guide (II). (1) cooling water, (2) beryllium (Be) target of 5.0 mm thickness, (3) viton o-rings, (4) extraction nozzle, (5) ^{nat}U targets of 14 mg/cm^2 thick, (6) helium gas inlet, (7) titanium foil, (8) gas diffuser. A blue arrow marks the primary proton beam. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the thin production target and are stopped and thermalised in the helium buffer gas. Because of the high ionisation potential of helium, a large fraction of the stopped recoils remain as ions long enough to be flushed with the buffer gas flow through a nozzle to a differential pumping section, where the ions are guided to the acceleration stage of the mass separator with electric fields [3]. Since the neutrons interact only in nuclear collisions, neither entrance nor exit window for the neutron beam is needed. The thickness of the aluminium wall of the guide towards the neutron source was reduced to ~3 mm.

A constraint for the design included the requirement of similar outer dimensions to the proton-induced fission ion guide in order to use the current mounting platform and buffer gas connectors. The stopping gas volume for the fission products is also similar, resulting in a comparable total evacuation time of the gas cell volume and thus a meaningful comparison between the performance of the different ion guides. Cross-sectional views of the ion guide are presented in Fig. 2(II). A gas volume of about 150 cm^3 corresponds to an evacuation time of 250 ms in room temperature helium buffer gas through an exit hole 1.2 mm in diameter.

A target holder supporting the fissile targets was designed to carry 6 foils of 20 × 50 mm^2 each, thus covering almost the entire inner surface of the ion guide. The first tests were performed with two 10 × 50 mm^2, 14 mg/cm^2 natural uranium targets which were placed in the closest position to the neutron production target (Fig. 2). The distance between the back surface of the beryllium and the center of the uranium targets is 35 mm. The fissile material covers an 80° angle in the horizontal plane (Fig. 2(a)) and a 22° angle in the vertical one (Fig. 2(b)), which corresponds to a solid angle coverage of 0.5 sr. A 2 mg/cm^2 thick titanium foil was placed in one of the spare target slots to obtain an estimate of the fraction of the fission products implanted into the ion guide walls.

The diagnostics for the IGISOL separator beam were the following (numbers in parentheses refer to Fig. 1): faraday cups before the bending electrodes (4) and in position (B) in the switchyard (6); 300 mm^2 silicon detector β-counters in position (A) before the dipole magnet (5) and in position (B) in the switchyard; a microchannel plate (MCP) detector in the switchyard just behind the silicon counter.

4. Results

4.1. Neutron measurements

In the tests of the neutron source the neutron activation analysis and time-of-flight technique have been employed to characterize neutron energies and intensities at different angles. The time-of-flight measurement was performed with thin-film breakdown counter (TFBC) detectors [12]. The detector consists of a very thin fissile target and a TFBC. The TFBC gives a signal from a neutron-induced fission event in the thin target and is insensitive to any other radiation. The time-of-flight was measured between the TFBC signal and the radiofrequency of the cyclotron.

Samples of naturally abundant aluminum, nickel, cobalt, indium and bismuth were used in the neutron activation analysis. The samples with dimensions 25.0 mm × 25.0 mm × 1.0 mm were placed at a distance of 14.5 cm from the target at angles of 0°, 45°, 90° and 135° with respect to the primary beam direction. The neutron irradiation of the samples was performed over 2.0 h with a primary proton beam intensity of 1.0 µA. Following irradiation γ-spectra of the samples were recorded with a high-purity germanium detector inside a low background shielding. Deduced activities have been used to unfold the neutron spectra with the respective neutron-induced reaction cross-sections. The obtained neutron spectra and those calculated with the MCNPX code [13] are presented in Fig. 3.

The measured and simulated spectra are in agreement above 5 MeV. The discrepancy below 3 MeV can be due to the unfolding process, where narrower energy bins might have been needed to properly account for resonance structure in the ^{115}In(n, γ)^{116m}In in cross section. Further details about neutron spectrum measurements can be found in [14].

Taking into account that both uranium targets are placed in the forward direction relative to the projectile protons and cover a solid angle of 0.5 sr, it is possible to estimate that $1.5 \cdot 10^{10}$ neutrons/s per 1 µA of the proton beam pass through those targets. Such a neutron flux induces $5 \cdot 10^{6}$ fissions/s per 1 µA of the proton beam. In comparison, the proton beam of 1 µA induces $3 \cdot 10^{9}$ fissions/s in the proton-induced fission ion guide, a factor of 600 times larger.

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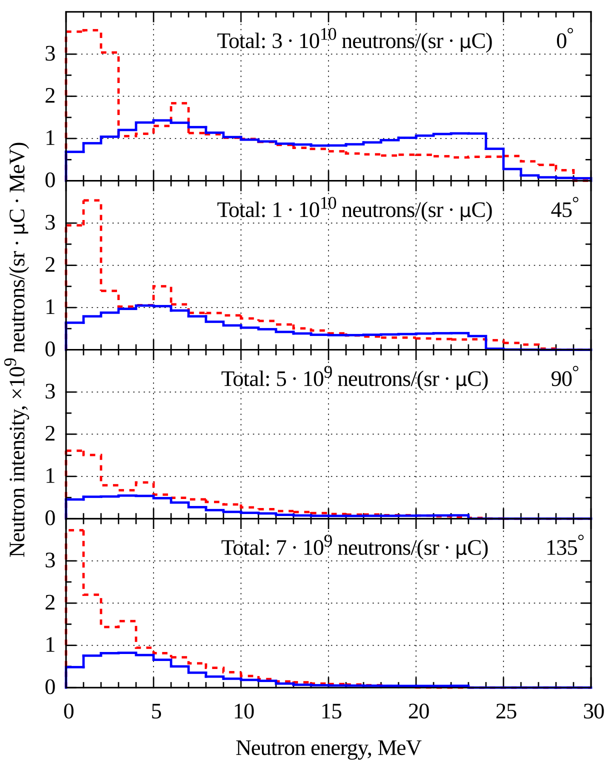

Fig. 3. Neutron intensities from the Be converter measured at different angles. Solid lines are measured with the activation method and dashed ones are calculated with the MCNPX code [13]. "Total" refers to the experimental neutron intensity integrated over the whole energy range.

4.2. Fission ion guide tests

With a primary beam current of 1 μA of 30 MeV protons on the neutron source a total current of 500 pA was measured with the FC faraday cup, see Fig. 1, for an ion guide pressure of 250 mbar. This is about three orders of magnitude less than a typical current of several hundreds of nanoamperes extracted from the proton-induced fission ion guide using the same primary beam intensity. We note that the extracted current is dominated by the ionized helium gas. In previous work we have estimated the ionization-rate density due to the energy deposited by the fission fragments as they stop (or pass through) the buffer gas [15]. This is approximately 4 orders of magnitude greater than the effect due to the scattering of protons in the same volume which therefore can be assumed to be negligible. The extracted current in this work agrees reasonably well with the expected loss factor compared to proton-induced fission, a combination of the proton-to-neutron conversion efficiency and the reduced solid angle.

The beam extracted from the ion guide was implanted in an aluminium foil in front of the silicon detector at position (A) in Fig. 1. The obtained β-count rate was 17 counts/s, which corresponds to about 50 fission products per second extracted from the ion guide. This number is to be considered as a lower limit due to the effects of cumulation and measurement time.

Fig. 4 shows the total current extracted from the ion guide and the count rate of the silicon detector in position (A) as a function of the ion guide helium pressure obtained with a 1 μA proton beam. The pressure was limited to 400 mbar due to incomplete sealing of the cyclotron beam line to the IGISOL target chamber via the connection with the neutron converter. The increase in ion guide pressure thus resulted in a steady increase in the baseline pressure of the cyclotron beam line. The extracted current and beta decay rate of fission products both show a steady increase as a function of pressure.

In Fig. 5 the dependence of the extracted current and silicon detector count rate on the primary proton beam current is shown. The extracted current shows a nonlinear increase as a function of primary beam intensity. The transport capacity of the SPIG which is located before the Faraday cup (FC) has been experimentally determined to be $\sim 10^{12}$ ions/s before space charge effects start to take effect [16]. Therefore, we believe that the nonlinear dependence reflects recombination effects within the ion guide. The recombination time scale of the He ions is inversely proportional to the square root of the ionization rate density, and is well within the evacuation time of the ion guide volume.

The β-count rate of fission products also increases as a function of primary beam intensity however the interpretation of the trend is more complicated. This reflects a number of competing factors in the gas cell resulting in an elemental dependence in the ion survival. Such effects have been studied previously at the LISOL laser ion source in experiments with a ^{252}Cf spontaneous fission source [17]. The beta activity measured at the highest proton intensity of 11 μA cannot be explained, however was seen to be reproducible. Future tests of the ion guide will be needed to confirm these measurements. In proton-induced fission, the mass-separated yield of ^{112}Rh has been studied at IGISOL-3 as a function of proton beam intensity and was shown to be linear up to a maximum intensity of ~10 μA [16]. Due to the increase in radiation levels around the mass separator, higher beam intensities were not attempted. Nevertheless, in a similar experiment performed with the skimmer electrode rather than the SPIG, deviations from a linear trend were seen at even higher intensities, interpreted due to space charge effects between the ion guide and skimmer. It will be important to test the neutron-induced fission at intensities well above ~10 μA in the future.

At the highest primary beam intensity an effort was made to mass separate the beam of fission products. The decay of mass-separated fission products was detected with a silicon detector beta counter in position (B) in the switchyard (Fig. 1). In mass number $A = 99$ a decay rate of 0.6 β-particles/s per 1 μA of proton beam was obtained. The typical decay rate of mass 99 in proton-induced fission with proton beam intensity of 1 μA is $5 \cdot 10^4$ β-particles per second. This decay rate has been deduced from the measured cumulative yield of ^{112}Rh, which is ~1500 atoms/(μC·mbarn).

Such a low yield for the neutron-induced fission cannot be explained solely due to the loss factor in proton-to-neutron conversion and experimental geometry. We suspect the mass separa-

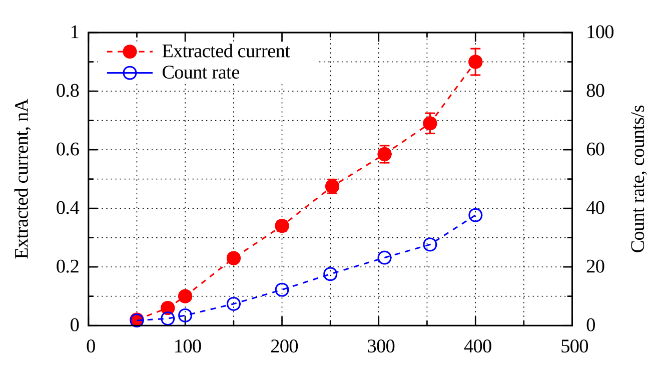

Fig. 4. Dependences of the extracted current and β-count rate on the buffer gas pressure. The proton beam current is 1 μA. Red dots are extracted current and blue open circles are Si count rates. Some error bars are smaller than the size of symbols. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

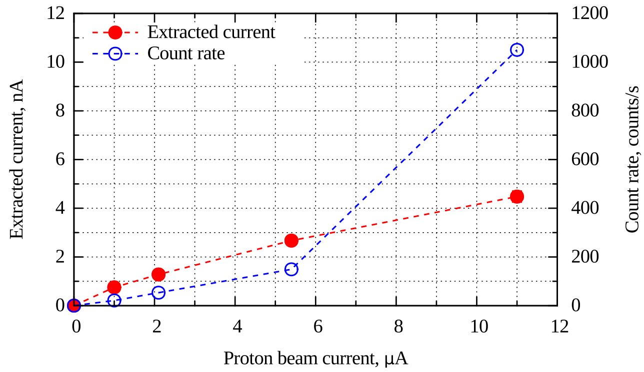

Fig. 5. Dependence of the extracted current and β-count rate on the proton beam intensity. Buffer gas pressure was 400 mbar. Red dots are measured extracted current and blue open circles are count rates. Some error bars are smaller than the size of symbols. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

tor tuning was poor. Usually the mass separator is tuned on-line using well known mass peaks associated with buffer gas contaminants, or with trace amounts of a calibrant gas such as Xe or Kr which can be introduced and monitored with Faraday cups [18]. Following this, the tuning of the radioactive species is optimized. In this experiment we did not see mass peaks with the separator Faraday cups, thus the tuning was far from optimal.

At the end of the on-line tests the aluminium foil in front of the silicon detector in position (A) was taken to the low background shield and a γ-ray spectrum of the implanted radioactive species was measured, see Fig. 6. Several days later the fission ion guide was dismantled and γ-spectra of the uranium target and the titanium foil were measured with the same setup.

The amount of fission products left in the targets and implanted in the collection foil could be deduced from the γ-ray intensities of the long-lived fission products. On the other hand, the production of the long lived nuclides can be approximated from the calculated fission rate ($5 \cdot 10^6$ fissions/s $\cdot$ µA), cumulative fission yields and the total proton beam dose. The cumulative yields of long-lived isotopes in fast fission of ^{238}U from [19] can be considered as a sufficiently close approximation. From the analysis of the gamma-ray spectra it can be estimated that about 12% of fission products with mass number $A = 99$ are stopped in the uranium target and ~0.09% are found from the implantation foil in the position (A), see Fig. 1. This value is comparable with the total efficiency of the proton-induced fission ion guide of 0.05% for $A = 99$, which is a combination of ~0.8% stopping efficiency [20] and the rest (about 6%) is an extraction and transport efficiency.

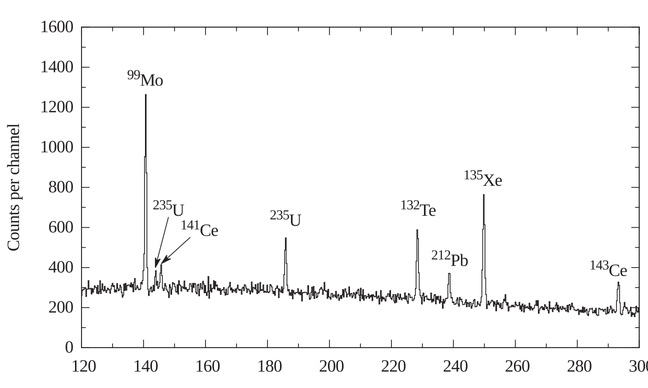

Fig. 6. A γ-ray spectrum from the aluminium foil in front of the silicon detector in position (A) collected 18 h after 30 min of implantation.

5. Summary and outlook

A neutron production target based on Be(p,n) reactions has been designed and built for the IGISOL facility. The experimentally obtained neutron spectra have a satisfactory agreement with the simulated ones.

The applicability of the neutron-induced fission to produce neutron-rich nuclei was tested with a new, specifically modified ion guide. Fission products were extracted from the ion guide and separated by the dipole magnet. Two different numbers of the neutron-induced fission ion guide efficiency were obtained by different methods. In our opinion, the value obtained by the off-line γ-spectroscopy is more trustable and it is comparable to that of proton-induced fission ion guide.

Further developments and experimental tests are needed before the neutron-induced fission can be used to produce neutron rich nuclei in a sufficient amount for studying their properties. It is clear that the neutron-induced fission cannot be fully utilized without a properly designed and constructed ion guide. Modeling calculations, which are going on, will help to improve the present construction of the ion guide. A different approach based on lower gas pressure and larger volume with electric fields to guide ions is also concidered as an option.

Another important step for the future development will also be a test at the full (~100 µA) intensity of the proton beam, performed preferably with on-line monitoring of the mass-separated fission products by means of γ-ray spectroscopy.

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