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β-delayed neutron emission studies

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Abstract The study of β-delayed neutron emission plays a major role in different fields such as nuclear technology, nuclear astrophysics and nuclear structure. However the quality of the existing experimental data nowadays is not sufficient for the various technical and scientific applications and new high precision measurements are necessary to improve the data bases. One key aspect to the success of these high precission measurements is the use of a very pure ion beam that ensures that only the

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ion of interest is produced. The combination of the IGISOL mass separator with the JYFLTRAP Penning trap is an excellent tool for this type of measurement because of the ability to deliver isobarically and even isomerically clean beams. Another key feature of the installation is the non-chemical selectivity of the IGISOL ion source which allows measurements in the important region of refractory elements. This paper summarises the β-delayed neutron emission studies that have been carried out at the IGISOL facility with two different neutron detectors based on ^{3}He counters in a polyethylene moderator: the Mainz neutron detector and the BEta deLayEd Neutron detector.

Keywords Beta delayed · Neutron emission · ^{3}He counter

1 Introduction

β-delayed neutron emission takes place when a precursor nucleus β-decays and the resulting daughter-nucleus emits a neutron. This neutron emission is energetically allowed if the excitation energy of the state populated in the β-decay is larger than the neutron separation energy of the daughter nucleus, S_n.

The study of β-delayed neutron emission probabilities, P_n, is of interest in different fields, such as nuclear technology applications, nuclear astrophysics and nuclear structure.

The technological interest of this type of study is related to nuclear power generation. In nuclear fission β-delayed neutron emission plays an essential role in safely controlling the sustainability of the fission reaction. Research into such nuclei is, therefore, fundamental for the design of safer and more efficient nuclear reactors. The Nuclear Energy Agency (NEA) highlights the importance of experimental measurements and data evaluation of delayed neutron emission in its working group 6, "Delayed neutron data" [1]. Furthermore, recently the International Atomic Energy Agency (IAEA) has held a consultants meeting on beta delayed neutron emission in order to boost further work on this field [2].

In nuclear astrophysics, the delayed neutron emission modulates the element abundance curve of stellar nucleosynthesis [3, 4]. Improved experimental data from delayed neutron emission represents an important input to r-process model calculations since properties of nuclei on the expected r-process path can be predicted by extrapolation on the basis of systematics of experimental $T_{1/2}$ and P_n values.

Furthermore, in nuclear structure, β-delayed neutron emission constitutes an important probe for the structure of neutron-rich nuclei far away from the valley of stability where other measurements are not yet possible [5–7]. The probability of neutron emission after β-decay, P_n, carries information on the β-strength just above the neutron separation energy, S_n.

However the quality of the existing experimental data nowadays is not sufficient for the various technical and scientific applications and it is necessary to perform new high precision measurements. This paper presents the studies of neutron emission probability after β-decay, P_n, carried out at IGISOL. The work with two different detectors, the Mainz neutron detector and the BEta deLayEd Neutron detector, BELEN, is summarised here. Both detectors consist of ^{3}He counters embedded in a polyethylene matrix. In the case of the Mainz neutron detector, experiments were carried out in the late 90s before the Penning trap was available and therefore the ion beams delivered by IGISOL were mass separated with relatively low mass resolution, typically M/δM of about several hundred. In the experiments with BELEN the Penning trap was fully functional and it allowed additional Z separation thus delivering a pure beam of the ion of interest.

The high purity of the ion beam is a key feature for this type of neutron detector since this measurement technique with ^{3}He cannot discriminate contaminant neutrons from those originating from the ion of interest. Neither does this technique allow discrimination of background neutrons. Another key advantage of IGISOL is the non-chemical selectivity of the ion-guide method which allows the production of radioactive beams of any element including the refractory elements which are very difficult to produce at other mass separators equipped with more conventional ion sources.

2 Mainz 4π neutron detector at IGISOL

Two experiments have been performed at the IGISOL facility to measure β-delayed neutron emission with the Mainz 4π neutron long counter [8]. This detector is formed by 42 ^{3}He tubes embedded in a polyethylene matrix and arranged in two crowns around the beam hole. The efficiency of this counter was determined to be (24.9 $\pm$ 0.2)% using a combination of two calibrated sources, Am/Li and ^{252}Cf, and online with the well-known neutron emitter ^{95}Rb [9].

The experimental technique at IGISOL was as follows. A beam of mass-separated short-lived ions was implanted directly onto a collection tape placed inside the neutron long counter. At the implantation position there was also a plastic scintillator for the detection of the β-particles and a Germanium detector that was used for the identification of the implanted ions via their corresponding γ-rays. The collection tape was moved periodically in order to remove the contamination from longer lived species.

2.1 P_n-values of very neutron rich Y to Tc isotopes

A first measurement was carried out by Mehren et al. [10] and it aimed at the measurement of very neutron rich isotopes from yttrium to technetium.

The ions were produced in fusion-fission reactions by bombarding a uranium target with a 50 MeV beam of H_2^+ of 6–8 μA intensity from the K = 130 MeV cyclotron at the University of Jyväskylä. The IGISOL separator was used to produce and separate isobarically pure beams of ions with a delay in the order of milliseconds. This experiment was carried out before the Penning trap was available, therefore there was no Z-separation of the different species.

The data was recorded as delayed coincidences between the neutron detection and the β-decay [6] in order to minimize the background. In this technique the coincidence rate was measured by an overlap technique in which the β-signal was stretched up to 40 μs by a gate-delay generator. In order to measure the random coincidences from the background a second coincidence was performed with the same stretched β-signal and a neutron signal which had been delayed by 45 μs. The use of this technique removes the dependence on the β-detection efficiency which is usually a source of error.

Table 1 Experimental values from this work [10, 11] compared to QRPA predictions [12]

Nuclide	$T_{1/2}$ (s)		P_n (%)	
	Experiment	Theory	Experiment	Theory
^{94}Kr	0.3 ± 0.1	0.44		0.55
^{99}Y	1.48 ± 0.02	0.93	2.5 ± 0.5	3.7
^{100g}Y	0.71 ± 0.03	0.29	1.8 ± 0.6	0.9
^{101}Y	0.40 ± 0.02	0.14	1.5 ± 0.5	1.4
^{102g}Y	0.29 ± 0.02	0.18	4.0 ± 1.5	1.6
^{103}Y	0.23 ± 0.02	0.08	8 ± 3	4.6
^{104}Y	0.18 ± 0.06	0.035		
^{105}Zr	0.6 ± 0.1	0.095		0
^{104g}Nb	5.0 ± 0.4	2.07	0.06 ± 0.03	0
^{105}Nb	2.8 ± 0.1	2.73	1.7 ± 0.9	0.26
^{106}Nb	0.9 ± 0.02	0.15	4.5 ± 0.3	0.27
^{107}Nb	0.30 ± 0.03	0.45	6.0 ± 1.5	3.7
^{108}Nb	0.19 ± 0.02	0.23	6.2 ± 0.5	11
^{109}Nb	0.19 ± 0.03	0.28	31 ± 5	26
^{110}Nb	0.17 ± 0.02	0.19	40 ± 8	20
^{109}Tc	0.8 ± 0.1	0.42	0.08 ± 0.02	0.02
^{110}Tc	0.78 ± 0.15	0.25	0.04 ± 0.02	0.075
^{111}Tc	0.29 ± 0.02	0.235	0.85 ± 0.2	0.53
^{112}Tc	0.29 ± 0.02	0.26	1.5 ± 0.2	1.1
^{113}Tc	0.17 ± 0.02	0.13	2.1 ± 0.3	
^{114}Tc	0.15 ± 0.03	0.08	1.3 ± 0.4	

The QRPA predictions for ^{94}Kr and ^{99}Y take into account the shape coexistance in these nuclei

If the neutron efficiency of the detector ϵ_n is known, the probability of β-delayed neutron emission P_n, can be expressed in terms of the number of β-neutron coincidences $n_{\beta n}$ and the number of β decays of the precursor, n_β, using the following equation,

$$P_n = \frac{n_{\beta n}}{\epsilon_n \cdot n_\beta} \tag{1}$$

The data from this experiment was also used to obtain the half-lives, $T_{1/2}$, of the β-delayed neutron precursors via a fit to the growth and decay curves. The β-decay half-lives of ^{103}Y, $^{108-110}$Nb were reported for the first time. For the isotopes for which previous experimental values existed there was good agreement with the values obtained in this experiment.

P_n values were obtained for ^{99}Y, ^{100g}Y, ^{101}Y, ^{102g}Y, ^{103}Y, ^{104g}Nb, ^{104m}Nb, ^{105}Nb, ^{106}Nb, ^{107}Nb, ^{108}Nb, ^{109}Nb, ^{110}Nb, ^{109}Tc, ^{110}Tc, ^{111}Tc and ^{112}Tc, out of which there were 13 new P_n values and several isotopes of yttrium whose P_n values, already known, were remeasured with improved statistics and higher reliability. The results are presented in Table 1. The higher reliability in the measurement is due to the fact that previously the yttrium isotopes had been obtained as decay products of rubidium or strontium, which introduced some uncertainty. However at IGISOL, refractory elements, such as yttrium can be obtained directly.

This experiment produced isotopes in the mass range from A = 99(Y) to A = 112(Tc) which allowed the authors to obtain the isotopic yield curves for fission products which can be used to test and improve theoretical models.

2.2 β-delayed neutron decay of ^{104}Y and $^{112-114}$Tc

A second measurement with the Mainz neutron detector was carried out by Wang et al. [11] and used the same experimental setup described above.

This experiment was a continuation of the systematic investigation started in the previous measurement. In this later experiment more neutron-rich nuclei were studied with a significant yield increase due to improvements in the ion guide and to an increased accelerator beam intensity resulting in isobaric yields of the order of 10^5 ions/s for medium mass fission-product beams.

The ions were produced in 25 MeV proton-induced fission of ^{238}U and were mass-separated by the IGISOL separator magnet. Signals from the γ-rays and from the observation of neutrons and β-particles were time-stamped within the measurement. These time-gated spectra were used to obtain the half-lives and production rates from the growth and decay curves.

The half-lives of the β-delayed neutron precursors were determined from the neutron time spectra by fitting the growth and decay periods. Only one component and a constant background were required for a satisfactory fit. The half-lives of ^{104}Y and ^{114}Tc were obtained for the first time. For ^{112}Tc and ^{113}Tc there existed previous experimental values that were in good agreement with the values found by Wang et al.

The β-delayed neutron branching ratios of ^{112}Tc, ^{113}Tc and ^{114}Tc were determined from the ratio of the neutron- and β-intensities obtained from the growth and decay curves. The low counting rate of ^{104}Y did not allow for an extraction of the P_n value. The values for ^{113}Tc and ^{114}Tc were reported for the first time in this work. The good statistics for ^{112}Tc resulted in a more precise value than in the previous attempt [10].

This data set also provided values for production yields of neutron-rich nuclei in proton-induced fission of ^{238}U.

2.3 Discussion of experimental results

The experimental values obtained by Mehren [10] and Wang [11] are presented in Table 1. The $T_{1/2}$ and P_n values were compared to predictions of an unpublished QRPA model [12] based on the FRDM masses. This QRPA calculation included Gamow–Teller and First Forbidden transitions. According to this model a trend of decreasing $T_{1/2}$ and increasing P_n value with increasing neutron number is expected. In general the P_n-values were larger than expected which could be due to a more abrupt decrease of the S_n values for $N \geq 60$ than predicted by the models. The large P_n values obtained indicate the importance of β-transitions to high-lying levels with large neutron excess. The fluctuations of the QRPA predictions from the experimental values can be explained taking into account the rapid changes in energies and ordering of the single particle orbitals that give rise to deformations and shape coexistance phenomena. For example, the β-decay daughters of ^{94}Kr and ^{99}Y have spherical-prolate shape coexistence. Furthermore $^{106-108}$Nb and $^{109-111}$Tc are predicted by Möller et al. [13] to be triaxial. While in the QRPA used in this calculation it is possible to account for shape coexistence, the model cannot account for triaxial shapes, therefore, discrepancies with respect to experimental data are expected in the triaxal nuclei.

3 BEta deLayEd Neutron detector at IGISOL

A prototype version of the BEta deLayEd Neutron detector, which is being developed for the FAIR/DESPEC experiment, has been used for the first time in an experiment at JYFL. This detector is based on ^{3}He counters and its first run was primarily intended to commission the detector and verify the working principles for future experiments. A new triggerless data acquisition has been developed for these measurements. This DACQ time-stamps the events and allows complete flexibility to construct correlations offline.

In the version employed at JYFL the detector consisted of 20 ^{3}He proportional counters with an effective length of 60 cm, a diameter of 2.54 cm and a gas pressure of 20 atm. The counters were embedded in a polyethylene block of dimensions $90 \times 90 \times 80$ cm^{3} and placed in two concentric crowns around the beam hole.

There are two polyethylene matrices for BELEN-20 whose difference is the radial position of the crowns and the radius of the beam hole. In the first version the counters are placed at a radius of 11 cm (8 counters) and 20 cm (12 counters) around a central longitudinal hole of 10 cm diameter. The detection efficiency for this version calculated with MCNPX (https://mcnpx.lanl.gov/) simulations is nearly constant below 1 MeV reaching 30% and decreases for higher energies (24% at 5 MeV). In the second version the first crown of detectors is placed at a radius of 9.5 cm and the second crown at a radius of 14.5 cm around a beam hole of 11 cm diameter. The average detection efficiency according to MCNPX simulations is 46% in the range from 1 keV to 1 MeV and it decreases to 29% at 5 MeV. Further details about the simulation work performed for this detector can be found in [14].

An isotopically pure beam was obtained using the JYFLTRAP Penning trap setup at the IGISOL facility and it was implanted on a movable tape placed in the centre of the BELEN-20 detector.

In this experiment, as well as with the Mainz detector, a measurement of the coincidence of a β-decay of the precursor and the neutron is required. However the main difficulty is the long moderation time of the neutron in the polyethylene (around 200 μs) which in a conventionally triggered system will require opening a long time window for the correlation. Such a long correlation window would cause a large dead time in the system. Therefore the BELEN-20 detector works with a purpose-built triggerless DACQ [15] where for energy signals above a certain threshold, time-energy pairs are registered independently for every channel. The GasificTL DACQ software builds the β-neutron coincidence with the desired correlation time. With this system the probability of neutron emission after a β-decay can be obtained with (1) and it is not subject to the uncertainty on the β-detection efficiency.

Two different experimental runs have been performed at IGISOL with the BELEN-20 detector. Each experiment was carried out with a different version of the polyethylene matrix.

3.1 First measurement with the BELEN detector at JYFL

In this measurement the first version of the BELEN-20 neutron detector was used and the known delayed neutron emitters of interest for nuclear power generation ^{88}Br, ^{94,95}Rb and ^{138}I were studied [16].

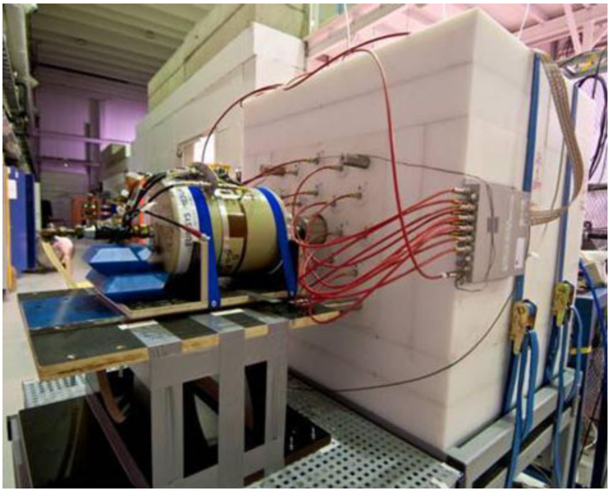

Fig. 1 BELEN neutron detector and Ge detector at JYFL

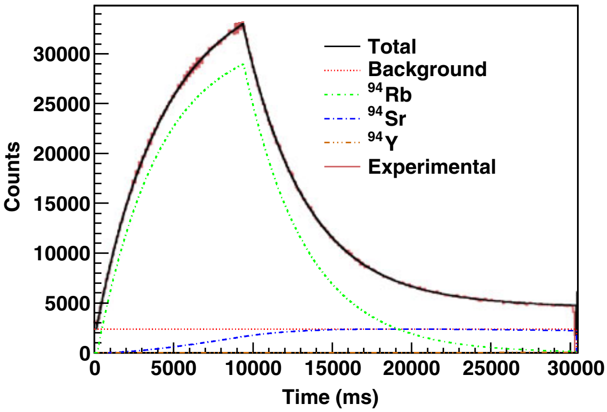

Fig. 2 Spectrum of the Si detector. Implantation and decay curve for ^{94}Rb and the fit of the Bateman equation

The radioactive species were produced at IGISOL by deuteron-induced fission ($E_d = 30$ MeV) on a uranium target. The IGISOL separator magnet was used for mass separation of the IGISOL beam which was then introduced into the Penning trap. The Penning trap was used as a very high resolution mass separator which removed all isotopes other than the one of interest. The isotopically pure beams extracted from the trap were delivered via a vacuum tube to the centre of the detector, where they were implanted on a movable tape. Two collimators were used to define the implantation position. The trap and tape transport cycles were adjusted in such a way that the radioactivity was accumulated during a period of $3 \cdot T_{1/2}$, while the measurement period was extended up to $10 \cdot T_{1/2}$ before removing the activity. A Si detector (0.9 mm thick, 25.2 mm diameter) for the detection of β-particles was placed closely behind the implantation position (distance 3 mm) also in vacuum. The instrumentation was complemented with an 80% efficiency HPGe detector for the detection of γ-rays, situated inside the central hole of the counter at a distance of 9 cm from the tape. The picture of the setup is presented in Fig. 1.

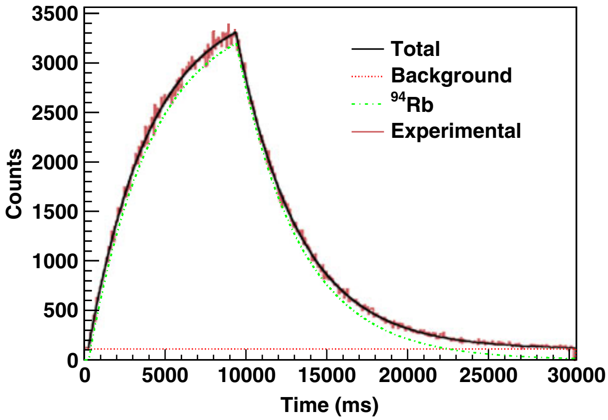

Fig. 3 Spectrum of the neutron detector. Implantation and decay curve for ^{94}Rb and the fit of the Bateman equation

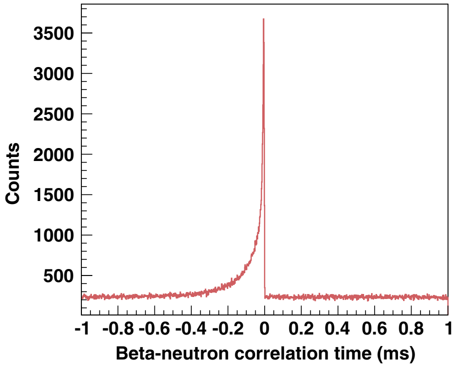

Fig. 4 Beta-neutron correlation spectrum

The measurements were performed for known delayed neutron emitters ^{88}Br, ^{94,95}Rb and ^{138}I. These emitters, with well known P_n values, together with measurements using a ^{252}Cf source were used to obtain the counter detection efficiency and to tune the Monte Carlo simulations.

In the preliminary analysis the growth and decay curves for the β-particles and the neutrons were fitted with the Bateman equation [17], see Figs. 2 and 3. These growth and decay curves were obtained by plotting the counts corresponding to the beta particles from the Si spectrum with respect to the cycle time and similarly with the neutron counts. The fit to the Bateman equation disentangles the contribution from each nucleus within the decay chain from the background and singles out the counts of the nucleus of interest.

The β-neutron coincidence spectrum, presented in Fig. 4, was built from the coincidences between the neutrons and all the beta particles in a 1 ms time window, forward and backward from the detection of a neutron. The true coincidences are the counts on left half of the spectrum on top of the flat background of random coincidences which is defined by the counts on the right half of the spectrum.

The P_n values of ^{88}Br and ^{95}Rb were used as references to calculate the efficiency of BELEN-20 since their values in references [9] and [18] have good agreement and small uncertainties. From these two values the average detection efficiency for BELEN-20 is (27.1 ± 0.8)% (see Table 2).

Table 2 BELEN-20 detection efficiency obtained using ^{88}Br and ^{95}Rb [9] as calibration

Isotope	P_n (%) [9]	N_β	$N_{\beta n}$	ϵ_n
^{88}Br	6.58 ± 0.18	867,701	14,350	27.6 ± 0.7
^{95}Rb	8.73 ± 0.20	588,116	13,301	26.6 ± 0.8

Table 3 P_n values for ^{94}Rb and ^{138}I obtained in this work, compared to other authors [9] and [18]

Isotope	N_β	$N_{\beta n}$	P_n (%)	Author
^{94}Rb	3,005,635	83,768	10.28 ± 0.31	This work
			10.01 ± 0.23	Rudstam [9]
			9.1 ± 1.1	Pfeiffer [18]
^{138}I	343,890	4,955	5.32 ± 0.2	This work
			5.46 ± 0.18	Rudstam [9]
			5.17 ± 0.36	Pfeiffer [18]

Using the above average efficiency and (1), the P_n values were obtained for ^{94}Rb and ^{138}I. They were in good agreement with the values reported in [9] and [18] (see Table 3).

3.2 Current and future work

Further measurements have been planned at the IGISOL facility with the motivations of technological applications, nuclear structure and astrophysics.

Some of the nuclei that we plan to measure in the future are fission fragments that have a major contribution to the number of delayed neutrons in nuclear power reactors [19]. We will focus on the new data requirements for reactor technology because of the use of high burn-up fuels or the burning of minor actinides where the fission product inventory differs from that of conventional reactors.

These and other nuclei that we plan to study are also important from the nuclear structure point of view. This will be the case for nuclei around the doubly magic ^{78}Ni and ^{132}Sn where the P_n values are sensitive to differences in the beta strength distribution. The shell reordering for these very neutron rich nuclei is actively discussed and information can be gained from our new measurements since the P_n values are sensitive to the structure of the daughter and parent nuclei.

Finally we will also target nuclei along the *r*-process path, where the decay towards the line of stability happens by a series of β-decays, accompanied by the emission of delayed neutrons in the case of the most neutron-rich isotopes. This is also the case of the above mentioned regions around ^{78}Ni and ^{132}Sn.

Some of these nuclei were already measured in June 2010. A primary beam of protons at 25 MeV with an intensity of 7 μA produced fission products from a ^{232}Th target. The BELEN-20 detector had the second version of the polyethylene matrix. The rest of the experimental setup and acquisition system was identical to the one described in the previous section.

The isotopes measured were ^{85}Ge, ^{86}Ge, ^{85}As, ^{91}Br and ^{137}I. ^{88}Br and ^{95}Rb were also measured in order to obtain the neutron detector efficiency. The data analysis process is being carried out and it is similar to that described in Section 3.1. Results will be published in the future.

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