β-delayed neutron emission of r-process nuclei at the $N = 82$ shell closure

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Abstract

Theoretical models of β-delayed neutron emission are used as crucial inputs in r-process calculations. Benchmarking the predictions of these models is a challenge due to a lack of currently available experimental data. In this work the β-delayed neutron emission probabilities of 33 nuclides in the important mass regions south and south-west of ^{132}Sn are presented, 16 for the first time. The measurements were performed at RIKEN using the Advanced Implantation Detector Array (AIDA) and the BRIKEN neutron detector array. The P_{1n} values presented constrain the predictions of theoretical models in the region, affecting the final abundance distribution of the second r-process peak at $A \approx 130$.

Keywords: β-delayed neutron emission, r-process

The astrophysical conditions for the r-process, i.e. the nucleosynthesis process responsible for the production of half the elements heavier than iron, are still a matter of debate [1–3]. Recent observations, such as the gravitational wave event GW170817 and its accompanying electromagnetic counterpart [4–8], point to binary neutron star mergers as a significant source of r-process material in the galaxy [8–10]. It is not yet determined whether these events are partially or entirely able to reproduce the r-process abundance pattern observed throughout the galaxy. Hydrodynamical models of these events [11] provide the astrophysical conditions present during these events, allowing reaction networks to simulate the nucleosynthesis taking place under explosive conditions [12]. Performing accurate reaction network calculations requires a precise knowledge of the nuclear properties of the nuclei involved. In particular, heavy element abundance predictions are sensitive to the values of nuclear masses, β-decay half-lives and β-delayed neutron emission probabilities P_n of very neutron-rich nuclei [13, 14]. r-process calculations are not only sensitive to P_n values of nuclei along the r-process path but also of nuclei encountered as they β-decay back to stability, where neutron emission causes branching along the decay chains modifying the final abundance distributions and act as a secondary source of neutrons during freeze-out.

Nuclear theory predictions of P_n values depend on the β-strength function S_β [15], and the masses of the nuclei used for the calculations. Theoretical models broadly fall into two categories: microscopic models and phenomenological models. Microscopic models aim to describe S_β based on microscopic theories, typically through some form of Quasiparticle Random Phase Approximation (QRPA) [16, 17]. Phenomenological models aim to provide a description of S_β based on the systematic trends of existing P_n values. The benchmarking of these theoretical models against new experimental data, as they are extrapolated far from stability, is critical for reliable modelling of the astrophysical r-process [2, 18]. When compared to the most recent evaluation of P_n values [19] microscopic models, such as the Finite Range Droplet Model with QRPA (FRDM+QRPA) [17, 20], systematically underpredict the P_n values of nuclei in the mass region south-west of ^{132}Sn, just below the $N = 82$ shell closure. Sensitivity studies have shown r-process abundances to be particularly sensitive to changes in P_n values in this region that shapes the second r-process peak [14]. In this region the total P_n value for most nuclei is equal to its $P_{1\mathrm{n}}$ value, the probability of a single delayed-neutron being emitted.

In this paper the β-delayed neutron emission probabilities and β-decay half-lives of 33 neutron-rich nuclei with $N \leq 82$ are presented. In particular, we report the first experimental $P_{1\mathrm{n}}$ measurements of 16 nuclides: $^{115-116}$Tc, $^{116-121}$Ru, $^{119-124}$Rh, ^{128}Pd and $^{127-129}$Cd. Also included, and often with higher precision than previous data, are measurements of $^{121-128}$Pd, $^{124-129}$Ag and ^{130}Cd that encompass the nuclides for which

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the current discrepancy between experiment and theory is observed.

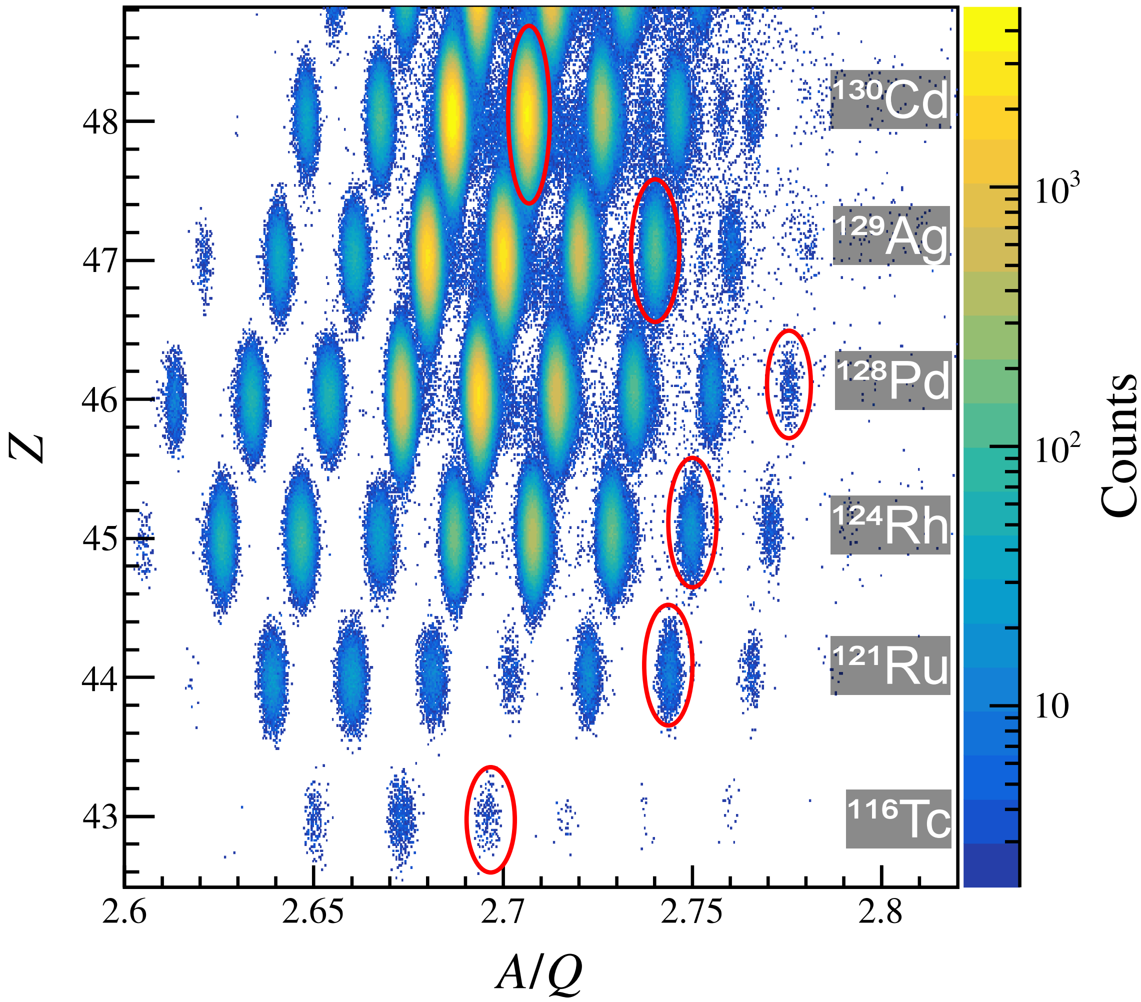

Figure 1: Particle identification plot obtained by BigRIPS showing the atomic number Z against A/Q ratio of ions implanted in the AIDA detector stack. Nuclide labels relate to the adjacent groups highlighted by red ellipses.

The experiment was performed at the Radioactive Isotope Beam Factory (RIBF) [21], located at the RIKEN Nishina Centre in Japan. Exotic neutron-rich nuclei were produced by in-flight fission of a 50 pnA primary beam of ^{238}U accelerated to an energy of 345 MeV/u impinging on a ^{9}Be target. The fission products of interest were analysed using the BigRIPS separator [22, 23]. Particle identification (PID) was performed using the $\Delta E - B\rho -$ TOF method [24] in the second stage of BigRIPS. The resulting PID plot is shown in Figure 1. Contaminant events such as hydrogen-like ions are clearly separated from the fully stripped ions of interest even for the most neutron-rich nuclei. The identified ions of interest were delivered to the F11 experimental area at a rate of around 100 ions per second via the ZeroDegree spectrometer [23].

The Advanced Implantation Detector Array (AIDA) [25] was installed in the F11 experimental area and used for the measurements of implanted ions and their subsequent decays. AIDA comprised six 128×128 strip, 1 mm thick Double-sided Silicon Strip Detectors (DSSDs). High resolution positional information was obtained for implanted ions via energy signal matching from the strips of the front and rear sides of the detector. When the energy was deposited across multiple adjacent strips, total deposited energy was calculated summing the individual strip contributions. The overlapping area between the front and rear strips in which energies were recorded form a cluster localising the event, typically to a region of ~ 1 mm^2 in the x and y planes of the detector. Decay events in the detector were localised using the same methodology although clusters were observed to vary in size due to the higher penetrability of β particles. Correlations between implantation and decay events were performed by identifying events in which the area of the β-decay event cluster was overlapping with or adjacent to the area of an implantation event cluster. This definition of a correlation was found to maximise the β-detection efficiency while minimising random correlations [26, 27].

β-delayed neutrons were detected using the BRIKEN neutron counter array [28, 29], which consisted of 140 ^{3}He proportional counters embedded in a High-Density Polyethylene (HDPE) matrix. A nominal neutron detection efficiency of 66.8(20)% was used for β-delayed neutrons in this region of interest. The efficiency was determined via the use of Monte Carlo simulations [28], and verified through measurements of the well-known neutron energy spectrum of ^{252}Cf [30].Theoretical predictions of the neutron-energy spectra expected were obtained for two of the most neutron-rich nuclides studied, ^{124}Rh and ^{129}Ag. The spectra were generated utilising the model detailed in Ref. [31] and took S_β from Ref. [17]. These spectra showed that the majority of neutrons are emitted in the energy range of $0-2$ MeV with average neutron energies of less than 1 MeV. Across this energy range the neutron-detection efficiency of BRIKEN is "flat" allowing the same nominal neutron-detection efficiency to be used for all nuclides [28, 30].

Half-lives and $P_{1\mathrm{n}}$ values were obtained through Bateman equation fits [32] of the β-decay and neutron-gated β-decay activities, which included the contributions of all decay products along the path to stability. The fits accounted for the contributions of random neutrons and random β-decay correlations. Figure 2 shows an example fit of the neutron-gated activity of ^{121}Rh. A detailed description of the full analysis methodology used can be found in Ref. [30]. All values that were not measured in this experiment were taken from the Evaluated Nuclear Structure Data File (ENSDF) database [33].

The $P_{1\mathrm{n}}$ values and half-lives for nuclides measured in this work are presented in Table 1. Where upper limits have been assigned to a $P_{1\mathrm{n}}$ value, it is calculated with a 95% confidence limit assuming a Gaussian estimator. Estimated masses extrapolated from the mass surface [34] indicate β-delayed two neutron emission is energetically possible for $^{128-129}$Ag and $^{127-128}$Pd. However, no evidence of two neutron emission was observed in this work.

Figure 3 shows our measured $P_{1\mathrm{n}}$ values grouped by element as a function of neutron number. Recommended $P_{1\mathrm{n}}$ values from the recent evaluation [19] are also shown in Figure 3. Predictions of four theoretical

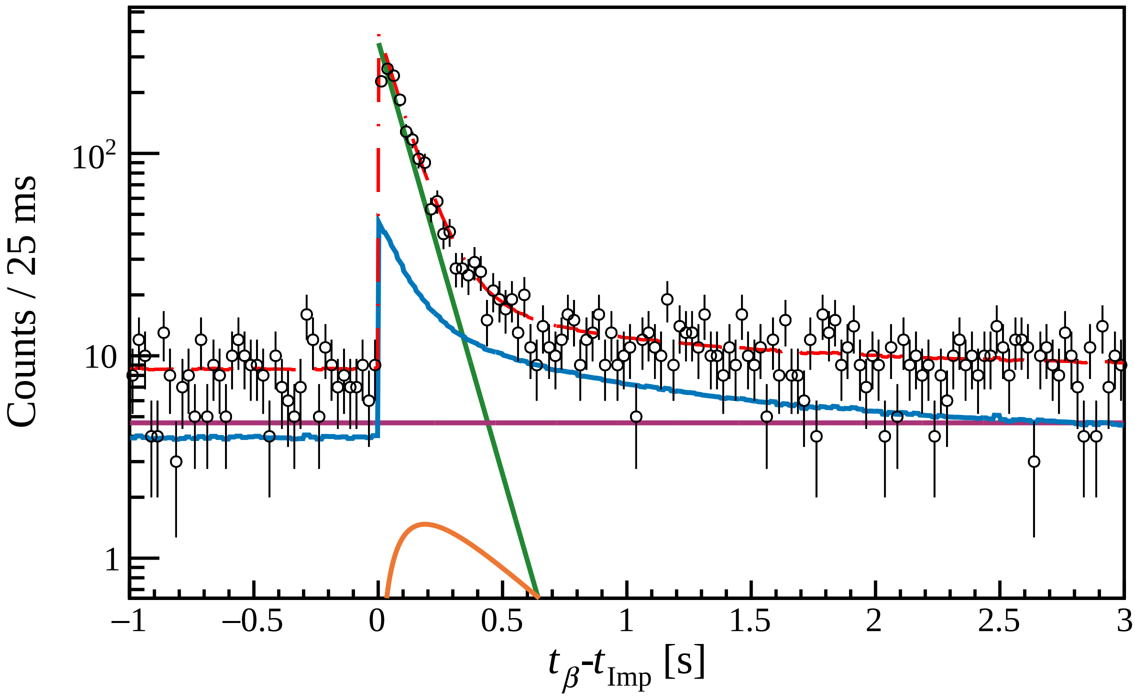

Figure 2: Time distribution of neutron-gated ^{121}Rh β-decay events fitted as part of the analysis. The fitted function (red dashed line) includes contributions of the parent decay (green line), β-delayed neutron emitting daughters and granddaughters (orange line), randomly correlated neutrons (blue line) and a linear random background (purple line).

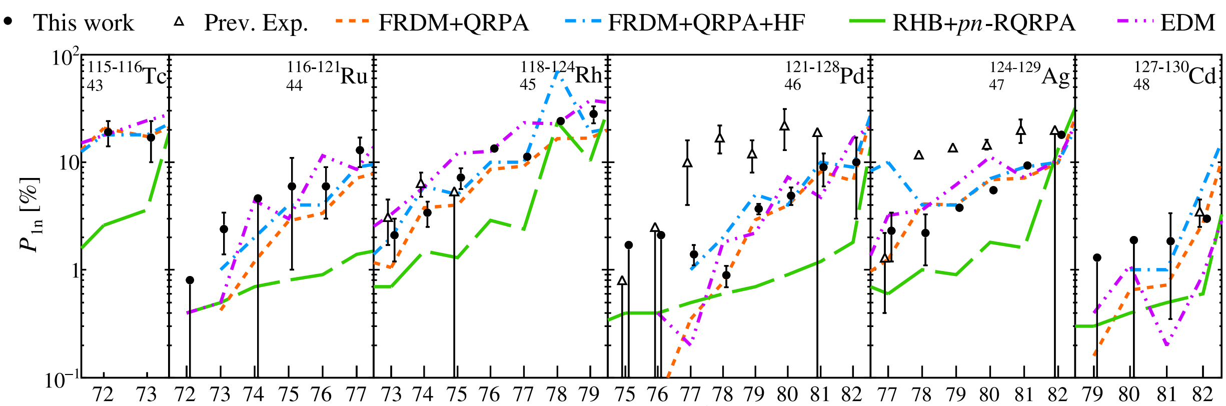

Figure 3: Experimental P_{1n} values (symbols) from both this work (circles) and the current recommended values from the most recent evaluation [19] (triangles). Lines are used to show the published theoretical P_{1n}-values: FRDM+QRPA (orange line) [20], FRDM+QRPA+HF (blue line) [17], RHB+pn-RQRPA (green line) [35] and the EDM (purple line) [36, 37]

model calculations are included. These include the Finite Range Droplet Model [38] with the Quasiparticle Random Phase Approximation (FRDM+QRPA) [20], the FRDM+QRPA with the inclusion of a Hauser-Feshbach framework (FRDM+QRPA+HF) [17], the Relativistic Hartree-Bogoliubov mass model with the proton-neutron Relativistic QRPA [35] (RHB+pn-RQRPA) and the semi-empirical Effective Density Model

[36, 37].

Comparing the P_{1n} values presented here to the values from the most recent evaluation [19], significant systematic differences can be seen in Figure 3. This discrepancy is also present between the theory predictions and this evaluation. The evaluation values which show the largest systematic differences, $^{123-127}$Pd and $^{125-128}$Ag, are all taken from a single source, corresponding to a PhD thesis [39] representing the only available source of measurements for these nuclides and labelled as "preliminary" in [19]. The two other sources that make up the evaluation in the region — providing P_{1n} values for $^{118-121}$Rh, $^{121-122}$Pd and ^{124}Ag [40]; and ^{130}Cd [41] — are from peer reviewed sources and are consistent with the present, often more precise, values.

The P_{1n} values reported in this work show a regular trend for most elements, of increasing neutron emission probability as neutron number increases. Some odd-even staggering in the P_{1n} values is observed for the lighter elements, such as Tc, Ru and Rh, though this is seen to diminish for nuclei close to $Z = 50$ where a smoother increase is observed. The predictions of the FRDM+QRPA and FRDM+QRPA+HF calculations reproduce this trend well across all isotopic chains, matching much of the staggering that is observed in the experimental values. The P_{1n} values predicted by FRDM+QRPA (2003) are calculated using the "cutoff" method [20], making the assumption that if a state above the neutron-separation energy S_n, the energy required to remove a single neutron from the nucleus, of the β-decay daughter is populated a neutron will be emitted. With the inclusion of the HF framework, de-excitation of the daughter is handled statistically, including γ-ray emission explicitly at every stage [31, 42]. The semi-empirical EDM calculations reproduce the general trend of the data well. Large odd-even staggering in the predicted P_{1n} values though result in the calculations fluctuating above and below the experimental values. The predictions of the RHB+pn-RQRPA are seen to be systematically smaller than both the predictions of the other models and the P_{1n} values measured here for almost all nuclides.

The impact of the newly measured P_{1n} values on r-process abundances was explored by estimating their effect during the decay to stability following the freeze-out of neutron-capture reactions. The calculation assumes that the r-process path passes through ^{128}Pd and ^{127}Rh, which act as classical waiting points with their abundances weighted by their respective half-lives, and that the decay to stability follows an instantaneous freeze-out. These isotones lying on the $N = 82$ shell closure are part of the r-process path in many calculations [43, 44]. The resulting isobaric abundance distribution of the stable nuclei produced after the progenitor ^{128}Pd and ^{127}Rh abundances decay back to stability is shown in Figure 4. Abundance uncertainties were calculated using a Monte Carlo approach where the experimental P_{1n} values were varied within their uncertainties. As it was not measured during the experiment, the P_{1n} value for ^{127}Rh was taken from the FRDM+QRPA+HF calculations due to their good agreement with the measured values of other nuclei in the region. This agreement between the theoretical P_{1n} values of FRDM+QRPA+HF and those presented in this work is reflected in the similar calculated abundances shown in Figure 4. In contrast the

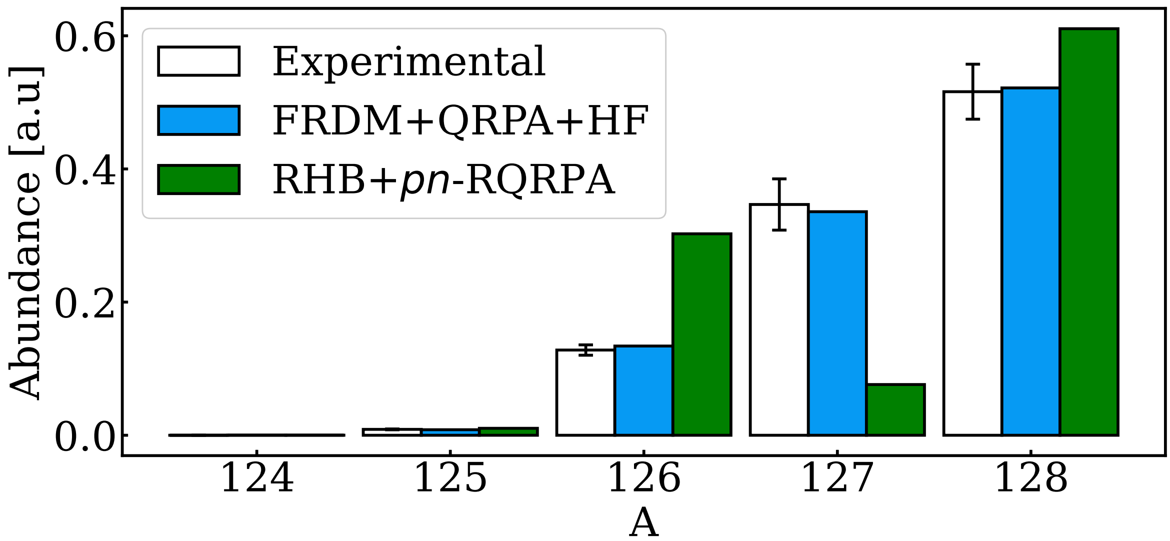

Figure 4: Resulting r-process abundance following an instantaneous freeze out starting with an initial abundance distribution of ^{128}Pd and ^{127}Rh weighted by their literature half-lives.

large differences between the theoretical RHB+pn-RQRPA P_{1n} values and experiment are seen to have a significant impact on the abundance distribution, with large differences seen across all values of A.

Comparisons from our present calculations can be made with solar r-process abundances by taking the ratio of isobaric abundances Y. For example the $Y_{A=128}/Y_{A=127}$ ratio obtained with our experimental P_{1n} values, 1.5(2), compares with observations of the solar r-process abundance distribution which vary from $1.73 - 1.77$ [45–47]. The difference between the calculated and observed abundance ratios may be explained by the absence of $A = 129$ progenitor nuclei in the calculation. The $A = 129$ isobars ^{129}Ag and ^{129}Cd have P_{1n} values of 17.9(14)% and 1.84(15)%, respectively, resulting in around 18% of the final $A = 128$ abundance originating from the $A = 129$ decay chain. Accounting for this contribution in the abundances of $A = 128$ increases the ratio of $Y_{A=128}/Y_{A=127}$ to 1.83(25) in very good agreement with the observed solar ratio. In contrast, calculations using the predicted P_{1n} values of RHB+pn-RQRPA result in a significantly larger ratio of 8.0, much larger than the observed solar ratio. These calculations show the importance of having precise P_{1n} values for use in r-process calculations, particularly in regions such as the $N = 82$ shell closure where large amounts of matter accumulate during the r-process allowing the P_{1n} values of relatively few nuclei to have a large impact on the final r-process abundance distribution.

Figure 5 shows our measured β-decay half-lives grouped by element and plotted against neutron number. Recent literature half-lives from Lorusso *et al.* [48] are also shown for comparison. Excellent agreement is observed between the two data sets, with almost all values falling within uncertainties. When comparing these values with the predictions of theoretical models in Figure 5, it is seen that the FRDM+QRPA cal-

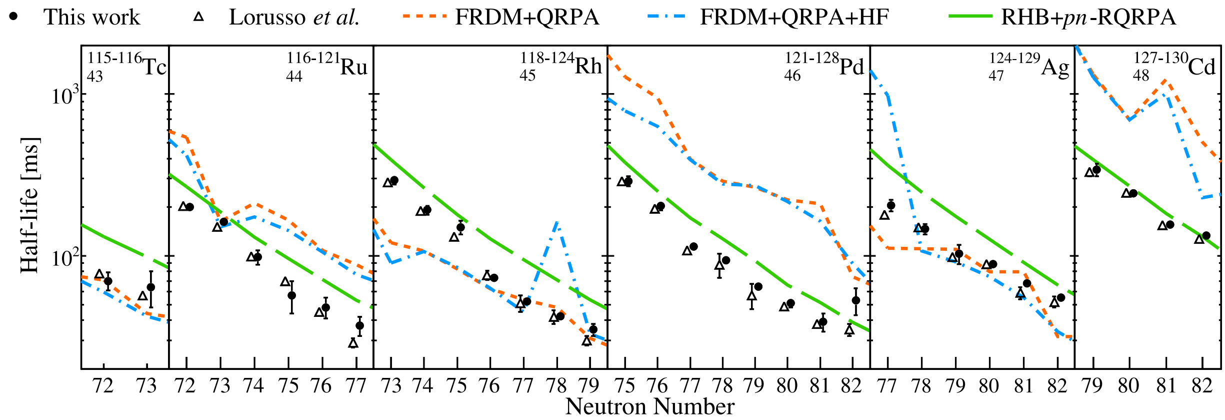

Figure 5: Experimental half-lives (symbols) from both this work (circles) and Lorusso *et al.* [48] (triangles). Lines are used to show the published values of theoretical half-lives: FRDM+QRPA (orange line) [20], FRDM+QRPA+HF (blue line) [17] and RHB+pn-RQRPA (green line) [35].

culations differ significantly from the measured half-lives, particularly for even-Z nuclides, in stark contrast to their good agreement with the measured P_{1n} values. The RHB+pn-RQRPA model best reproduces the nuclides presented here, despite systematically underpredicting the P_{1n} values of all nuclides. In particular, the RHB model calculations accurately reproduce the values for the high-Z Cd nuclides. The differences between the various models' abilities to predict P_n values and half-lives shows the importance of having experimental measurements of both quantities to test the validity of these theoretical models as they are extrapolated far from stability.

In summary, we have presented β-delayed neutron emission probabilities and β-decay half-lives of 33 neutron-rich nuclei around the N=82 shell closure of importance for the astrophysical r-process. Our new P_{1n} values are generally well reproduced by theoretical models. This agreement is in contrast with a significant discrepancy between the very recently published evaluation of P_n values [19] and the predictions of these theoretical models in the same region. Furthermore, we showed that while FRDM+QRPA calculations are able to reproduce the present P_{1n} values well, they are unable to reproduce the measured half-lives, in particular those of even-Z nuclides. In contrast RHB+pn-RQRPA calculations systematically underpredict P_{1n} values in this region, but are best able to reproduce the measured half-lives. This showcases the strengths and weaknesses of different theoretical approaches, and reinforces the importance of having experimental measurements. In particular, the benefit of having experimental measurements of multiple properties predicted by the models is evident, with just experimental half-lives or P_n values to compare one could falsely conclude that a model performs well in a region. Calculations performed exploring the impact of P_{1n} values on the local astrophysical r-process abundance distribution shows that the present P_{1n} values well explain the observed solar $A = 127$ and 128 abundances that form part of the second r-process peak.

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Table 1: β-delayed neutron emission probabilities P_{1n} and half-lives measured in the present work. The nuclides for which a $P1n$ value is reported for the first time are indicated by an asterisk (*)

Nuclide	P_{1n} [%]	Half-life [ms]	Nuclide	P_{1n} [%]	Half-life [ms]	Nuclide	P_{1n} [%]	Half-life [ms]
^{115}Tc*	19(5)	70(9)	^{121}Rh*	13.4(8)	73(2)	^{128}Pd*	10(7)	52(10)
^{116}Tc*	17(7)	64(16)	^{122}Rh*	11.3(7)	52.4(15)	^{124}Ag	2.3(11)	205(17)
^{116}Ru*	< 0.8	200(11)	^{123}Rh*	24.2(14)	42.2(18)	^{125}Ag	2.2(11)	146(11)
^{117}Ru*	2.4(10)	162(9)	^{124}Rh*	28(5)	35(3)	^{126}Ag	3.8(2)	103.2(14)
^{118}Ru*	< 4.6	98(10)	^{121}Pd	< 1.7	290(20)	^{127}Ag	5.5(2)	89.1(9)
^{119}Ru*	6(5)	57(13)	^{122}Pd	< 2.2	203(12)	^{128}Ag	9.3(5)	67.4(16)
^{120}Ru*	6(3)	48(7)	^{123}Pd	1.4(3)	114(2)	^{129}Ag	17.9(14)	55(3)
^{121}Ru*	13(4)	37(5)	^{124}Pd	0.89(20)	94(3)	^{127}Cd*	< 1.2	340(30)
^{118}Rh	2.1(9)	294(17)	^{125}Pd	3.7(4)	64.4(17)	^{128}Cd*	< 1.9	243(11)
^{119}Rh	3.4(9)	192(12)	^{126}Pd	4.9(9)	51(3)	^{129}Cd*	1.84(15)	155.9(13)
^{120}Rh	7.2(16)	150(15)	^{127}Pd	9(3)	39(5)	^{130}Cd	3.0(2)	134(3)