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Measurement of (n,γ) reaction cross sections at stellar energies for ^{58}Ni and ^{78}Se

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Abstract

The energy averaged (n,γ) cross sections of ^{58}Ni and ^{78}Se at a stellar energy of $kT = 25$ keV have been investigated with the so called activation technique, which represents a well established tool for measurements of stellar neutron cross sections. For both nuclides, off-line decay counting is prohibitive due to the long half-lives of the reaction products as well as the absence of suitable γ-ray transitions. The measurements reported here are, therefore, based on accelerator mass spectrometry (AMS) determination of the number of ^{59}Ni and ^{79}Se nuclei produced by irradiation of Ni and Se targets in a quasi-stellar neutron spectrum. We present in this paper preliminary results of $\sigma_{\mathrm{exp}}(kT = 25\ \mathrm{keV}) = 27.2 \pm 1.8$ mb and 61 ± 10 mb for neutron capture cross sections of ^{58}Ni and ^{78}Se, respectively.

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Keywords: Accelerator mass spectrometry; ^{59}Ni; ^{79}Se

1. Introduction

The origin of the elements heavier than iron can be almost completely ascribed to neutron capture processes characterized by much longer and much shorter time scales compared to average β decay half-lives. The respective nucleosynthesis processes, known as the slow (s) and the rapid (r) neutron capture process, contribute in approximately equal parts to the total elemental abundances in the mass range above iron.

Our measurements refer to the mass region, which is dominated by the so called "weak" component of the s-process. Next to ^{56}Fe, which is the most important seed nucleus for the s-process path, ^{58}Ni constitutes a significant fraction of the seed.

The reaction flow to the higher mass region depends not only on the abundances of the seed nuclei but also on their stellar neutron capture cross sections.

A large fraction of these cross sections can be determined directly by measuring the prompt γ-rays associated with the neutron capture reaction via the time-of-flight method (TOF), like e.g. with the Karlsruhe 4π BaF_2 detector [1]. A method to include also the "direct" component of the capture process is to measure the activity of the product nuclei – if unstable – after the irradiation, applying the so called activation method. Existing data for the cross sections of ^{58}Ni to ^{59}Ni ($T_{1/2} = (7.6 \pm 0.5) \times 10^4$ a [2]) are based on TOF measurements [3–5] with quoted systematic uncertainties of about 5%. Although the three data sets are

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in fair agreement, a comparison with the result of an alternative method could add confidence in the value.

For ^{78}Se, no experimental information on its stellar (n,γ) cross section exist, so far. In this case, experimental data are essential for evaluating the important branching in the *s*-process reaction path at ^{79}Se. The terrestrial half-life ($(2.80 \pm 0.36) \times 10^5$ a [6]) of ^{79}Se is drastically reduced under stellar conditions due to thermal population of the low-lying isomeric state ($I^\pi = 1/2^-$ at $E_\gamma = 96$ keV, $T_{1/2} = 3.9$ min) with a small beta-decay branch. The partial β^- half-life of the isomer is 4×10^5 s ([7]), thus β-decay competes with neutron capture at temperatures above 1.5×10^8 K. The strength of the resulting branching in the nucleosynthesis process is reflected in the abundance of the *s*-only isotopes ^{80,82}Kr. Analysis of the local abundance pattern in the mass region $80 \leqslant A \leqslant 82$ allows to deduce the effective half-life of ^{79}Se at the *s*-process site. Since the temperature dependence of the half-life is well known [7], the branching at ^{79}Se can be interpreted as an *s*-process thermometer [8].

The determination of neutron cross sections by means of the activation technique represents an important complement to measurements using the time-of-flight method since this independent approach implies different systematic uncertainties, and gives access to partial cross sections leading to isomeric states. In combination with AMS the activation technique can be extended to hitherto inaccessible cases, e.g. to reactions producing very long-lived nuclei with very weak or completely missing γ-transitions. The application of AMS counting in stellar neutron reactions, which has been demonstrated recently [9], has the further advantage of being independent of uncertainties in γ-ray intensities.

2. Neutron irradiation of the samples at the FZK

Both samples were activated at the Karlsruhe 3.7 MV Van de Graaff accelerator at the Forschungszentrum Karlsruhe (FZK). Neutrons were produced with the ^{7}Li$(p,n)^7$Be source by bombarding 30 μm thick layers of metallic Li on a water-cooled Cu backing with protons of 1912 keV, 30 keV above the reaction threshold, typical beam currents were about 100 μA. The resulting quasi-stellar neutron spectrum approximates a Maxwellian distribution for $kT = 25 \pm 0.5$ keV [10] with a maximum neutron energy of 108 keV (see Fig. 1). The neutron flux is kinematically collimated in a forward cone with 120° opening angle. Neutron scattering through the Cu backing is negligible since the transmission is about 98% in the energy range of interest. To ensure homogeneous illumination of the entire surface, the beam was wobbled across the Li target.

Both samples were sandwiched at the activation position between 10–30 μm thick Au foils. In this way the neutron flux can be determined via the induced ^{198}Au activity relative to the well-known neutron capture cross section of ^{197}Au [10] after the irradiation. The decay parameters for

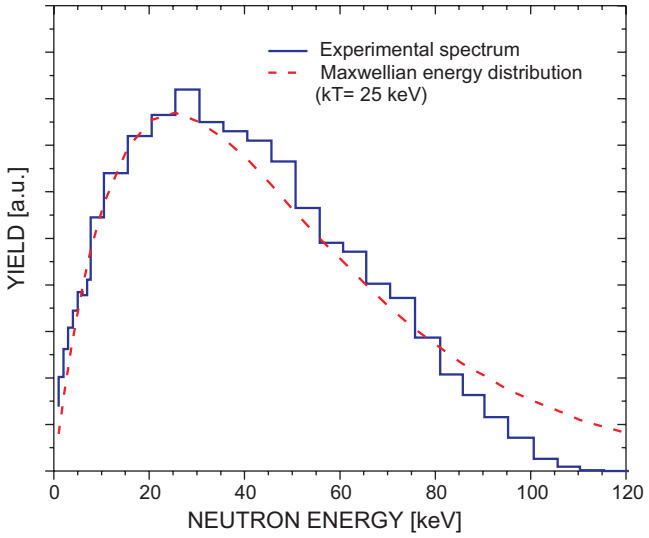

Fig. 1. Comparison of the experimental neutron distribution and the Maxwellian energy distribution spectrum, which approximates a Maxwellian distribution for $kT = 25 \pm 0.5$ keV [10].

^{198}Au were taken from [11]. The mean neutron flux over the activation period was $1.5–2.0 \times 10^9$ *n*/s at the position of the samples, which were placed in close geometry to the Li target. Throughout the irradiation the neutron flux history was recorded in intervals of 1 min using a ^{6}Li-glass detector to account for neutron flux variations and nuclei decaying during the activation. This factor can be neglected for very long-lived isotopes, but has to be taken into account in the determination of the total neutron flux via the Au foils.

The sample material used for the irradiation was prepared from material assuming natural isotopic composition (23.77% for ^{78}Se and 68.077% for ^{58}Ni, respectively [12]). In case of the activation of the nickel samples, two equivalent nickel foils were sandwiched by Au foils in front and after, respectively. Two pellets, one from CdSe and the other from pure Se metal powder were prepared and simultaneously activated in double sandwich geometry with three Au foils.

3. AMS measurement of the irradiated Ni and Se samples

^{59}Ni and ^{79}Se, have long half-lives and no suitable γ-ray transitions for offline decay counting. Therefore we have measured the number of produced nuclei via AMS. The AMS system at Munich consists of a sputter ion source, a 90° injector magnet, an 18° electrostatic deflection, a 14 MV tandem accelerator and a Wien filter. To separate the radioisotopes from the stable isobars we use the sensitive combination of a gas-filled magnet with a multi-ΔE ionization chamber [13,14]. With AMS we determine the concentration ratio of the radioisotope relative to a stable isotope by measuring the number of radionuclides relative to the current of the isotopic ions in front of the detector, after adjusting the injector magnet, terminal and Wien

filter voltage appropriately. By measuring relative to a standard sample of known isotopic ratio, factors like stripping yields and transmissions mostly cancel.

3.1. ^{59}Ni-measurements

For the determination of ^{59}Ni/Ni ratios the tandem was operated at a terminal voltage of 12.58 MV and the charge state 12^+ was selected. Although ^{59}Co is suppressed in total by 9 orders of magnitude, the background in the spectra is due to tails of ^{59}Co. In a first measurement the nickel sample had a high Co contamination. Therefore, the second irradiated foil was dissolved in 10 N HCl and purified by anion exchange chromatography with DOWEX AG1 resin. In a subsequent step ammonia was added until pH 9 and Ni was precipitated with dimethylglyoxim. This complex was centrifuged and washed with pure water, then ashed to nickel(II)oxide which serves as sample material. Due to this procedure the isobar ^{59}Co is suppressed by two orders of magnitude.

The ^{59}Ni standard material was produced via irradiation of natural Ni powder with thermal neutrons. The simultaneously produced ^{65}Ni ($T_{1/2} = 2.52$ h) served as a neutron flux monitor for the neutron fluence. The corrections for the epithermal flux are in the order of 0.5% and cancel each other. The activity of ^{65}Ni was measured with a γ-ray detector by the two well known γ-ray lines of 1481.8 keV and 1115.5 keV. With the thermal neutron cross section of ^{64}Ni ($\sigma_\gamma = 1.52 \pm 0.03$ b [15]) and the recently remeasured thermal cross section of ^{58}Ni ($\sigma_\gamma = 4.13 \pm 0.05$ b [16]), we calculate a ^{59}Ni/Ni ratio for our standard of $(9.1 \pm 0.4) \times 10^{-11}$. This error also includes the uncertainties from the geometry of the sample (2%), the statistical uncertainty of the γ-ray measurement (1.8%) and the uncertainty of the calibration (^{152}Eu with 2.0%). Fig. 2 shows a spectrum of the differential energy loss signal $\Delta E4$ versus the position in the ionization chamber with software conditions on all other measured parameters for a standard and a blank sample. The remaining counts from the blank sample are due to ^{59}Co ions. This background contribution (less than 1%) is subtracted from the observed events of a real sample according to the total ^{59}Co counts.

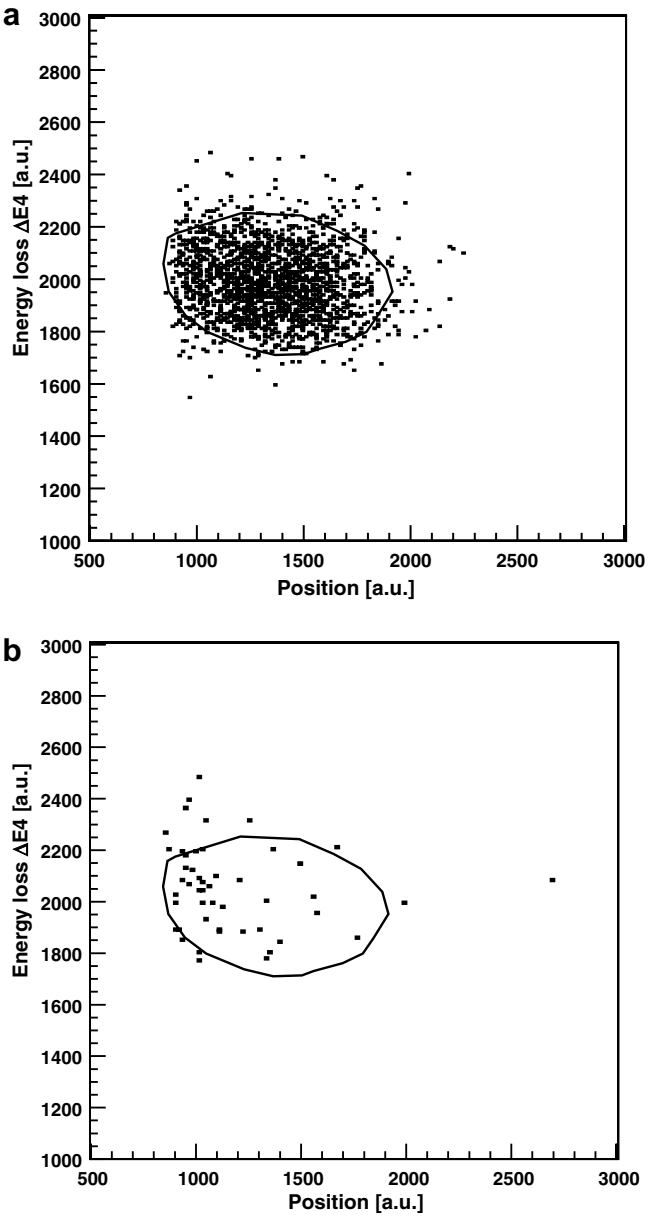

Fig. 2. Spectrum of the energy loss signal $\Delta E4$ versus position in the ionization chamber. Software cuts are applied on the other measured parameters. The cut in the spectra defines the accepted counts. (a) Spectrum of the standard; with software cuts this sample yields 2038 counts, corresponding to a transmission of 34% from the cup in front of the gas-filled magnet to ^{59}Ni counts the detector. (b) Blank sample with 16 times more integrated current at the detector. With the applied software cuts 32 counts survive, what corresponds to a ^{59}Ni/Ni ratio of 3×10^{-14}.

3.2. ^{79}Se-measurements

The determination of ^{79}Se/Se ratios at low levels is demanding by several reasons. (i) Because of the higher nuclear charges ($Z = 34$ and $Z = 35$ for the stable isobar Br) and the lower velocities for the higher masses, the isobars are not well separated in the gas-filled magnet and the ionization chamber. (ii) Bromine is rather volatile and easily contaminates a sample. In addition, it forms readily negative ions. (iii) The neighboring isotopes ^{78}Se and ^{80}Se are stable and highly abundant (23.8% and 49.6%, respectively).

Therefore we chose the rather high charge state 15+ (stripping yield: 0.64%) at a terminal voltage of 12.55 MV leading to an energy of 200 MeV. The rate of the isobar ^{79}Br is reduced by the aperture after the gas-filled magnet and in front of the ionization chamber by roughly two orders of magnitude leading to tolerable count rates. Approximately 1 kHz resulting from ^{79}Br particles were registered with the ionization chamber for ^{78}Se currents of a few particle nA in front of the gas-filled magnet. Stringent software cuts had to be applied in order to extract the

^{79}Se signals from the vast ^{79}Br background. Under such conditions, a sample containing bromine at a level of 10^{-5} can be quantified for ^{79}Se/^{78}Se ratios of about 10^{-11}. Fig. 3 shows spectra of a standard and a blank sample with all software cuts applied to identify ^{79}Se.

The ^{79}Se standard material was produced via the irradiation of a Se target with thermal neutrons. To reduce the induced activity of ^{75}Se ($T_{1/2} = 120$ d), we used a target enriched in ^{78}Se (98.8%) while depleted in ^{74}Se (about 0.01% instead of 0.87% natural abundance). The neutron flux was determined with a Au standard with an uncertainty of 3%. The cross section of 0.43 ± 0.02 b [17] for the thermal ^{78}Se$(n,\gamma)^{79}$Se reaction yields a ^{79}Se/^{78}Se ratio of $(1.47 \pm 0.09) \times 10^{-8}$.

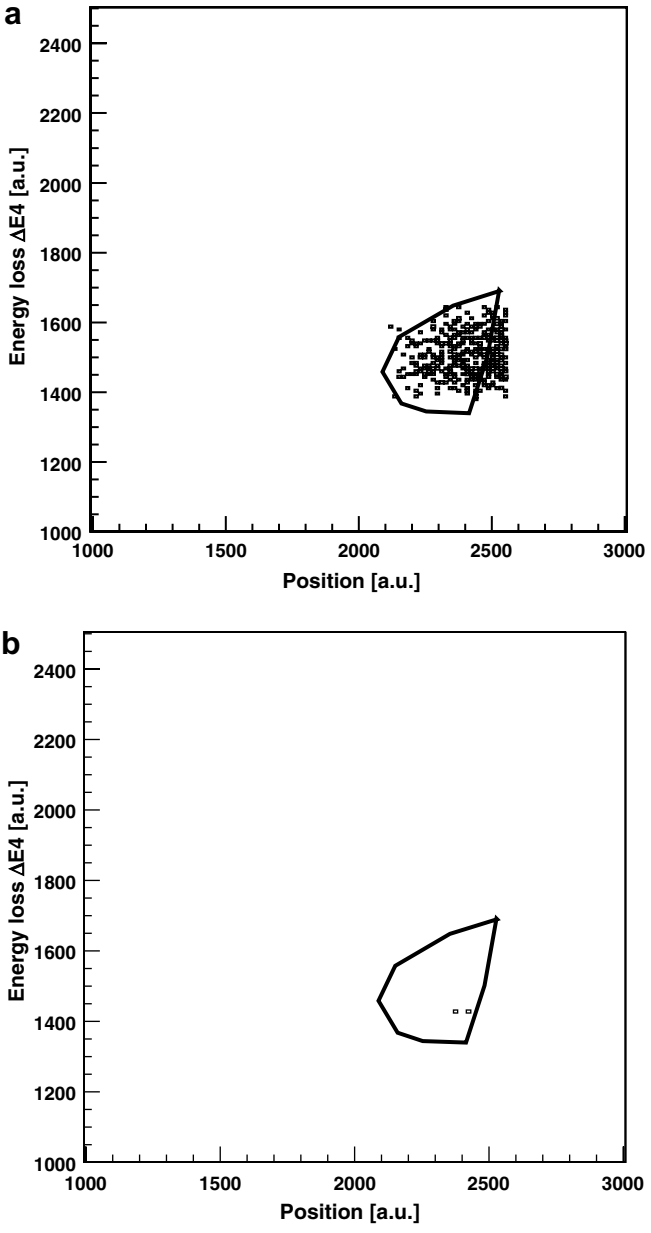

Fig. 3. Spectrum of the energy loss signal $\Delta E4$ versus position in the ionization chamber. Stringent software cuts are applied on the other measured parameters. The cut in the spectra defines the accepted counts. (a) Spectrum of the standard, 266 in the cut corresponds to a transmission of 1% from the last cup to the detector. (b) Spectrum of the blank sample, corresponding to a ^{79}Br suppression of roughly 10^6.

4. Results and discussion

The total number of activated nuclei N_{act} after the irradiation is given by

$$N_{\mathrm{act}} = N\Phi_{\mathrm{tot}}\sigma(1 - \mathrm{e}^{-\lambda t_{\mathrm{a}}}) \tag{1}$$

with Φ_{tot} being the time-integrated neutron flux and t_a the activation time. Φ_{tot} can be determined from the activity measurement of the respective gold foils (for a detailed description, see [18]). The values for Φ_{tot} with an estimated uncertainty of 3% are shown in Table 1. Due to the long half-lives of ^{59}Ni and ^{79}Se the correction factor $(1 - \mathrm{e}^{-\lambda t_{\mathrm{a}}})$ for the decay during the time of activation can be neglected. AMS determines directly the isotopic ratio N_{act}/N, thus the respective cross section for the experimental neutron spectrum at $kT = 25$ keV (σ_{25}) can be calculated from

$$\sigma_{25} = \frac{N_{\mathrm{act}}}{N}\frac{1}{\Phi_{\mathrm{tot}}} \tag{2}$$

In an astrophysical environment with temperature T, the neutron spectrum corresponds to a Maxwell-Boltzmann distribution $\Phi \approx E_n \mathrm{e}^{-E_n/kT}$. The experimental neutron spectrum of the ^{7}Li$(p,n)^7$Be reaction approximates a Maxwellian distribution with $kT = 25$ keV almost perfectly (see Fig. 1) [10]. However, to obtain the exact Maxwellian averaged cross sections for the temperature T, the energy-dependent cross section $\sigma(E)$ has to be folded with the associated Maxwellian energy distribution. A more detailed analysis is in progress, thus the experimental values presented here are no Maxwellian averaged cross sections, but only the values derived with our experimental neutron distribution.

The AMS result includes statistical uncertainty, standard uncertainty and a systematic error. The error weighted mean of three measurements results in a ^{59}Ni/^{58}Ni ratio of $(2.53 \pm 0.15) \times 10^{-11}$, corresponding to a σ_{25} (^{58}Ni) of 27.2 ± 1.8 mb (see Table 2). A blank sample of similar Co content gave an isotopic ratio almost three orders of magnitude lower than the irradiated samples.

In the case of ^{79}Se only the CdSe sample has been used since the Se metal sample could not be used because of a

Table 1
Activation parameters and sample characteristics

Sample	Mass (mg)	Atoms ^{58}Ni or ^{78}Se	t_a (d)	Φ_{tot} (neutrons/cm^2)
Ni	47.3	3.30×10^{20}	6.0	$(9.30 \pm 0.28) \times 10^{14}$
CdSe	216.8	1.62×10^{20}	13.0	$(1.96 \pm 0.06) \times 10^{15}$

Φ_{tot} gives the total neutron exposure of the sample during the activation; the uncertainty was estimated to be 3%.

Table 2
Preliminary results from our measurements, in comparison with the recommended Maxwellian averaged cross section [19] and theoretical predictions from the Hauser-Feshbach code NON-SMOKER [20]

Sample	N_{act}/N	σ (exp) (mb)	Bao et al. [19]	NON SMOKER [20]
Ni	$(2.53 \pm 0.15) \times 10^{-11}$	27.2 ± 2.1	41 ± 2	50.5
CdSe	$(1.19 \pm 0.19) \times 10^{-10}$	61 ± 10	109 ± 41[a]	74

The AMS results are corrected for isobaric background. For ^{59}Ni this correction is less than 1%, for ^{79}Se about 10%.
[a] Semi-empirical prediction.

too high ^{79}Br background. After correction of a 10% ^{79}Br background we get a ^{79}Se/^{78}Se ratio of $(1.19 \pm 0.19) \times 10^{-10}$, corresponding to a value for σ_{25} (^{78}Se) of 61 ± 10 mb (see Table 2). The uncertainty includes also a 10% systematic uncertainty for the stringent conditions applied for the analysis.

Additional measurements for both isotopes are planned, therefore the results should be considered to be preliminary. Both results are lower than the presently recommended values from the Bao et al. compilation [19] (41 ± 2 mb and 109 ± 41 mb, respectively). The result of the ^{78}Se cross section is close to the value of 74 mb from the Hauser-Feshbach calculation NON-SMOKER [20]. The NON-SMOKER calculation for ^{58}Ni yields 50.5 mb, higher than the three time-of-flight measurements [2–4], which agree all within 5% (41 ± 2 mb).

Our AMS measurement shows an important contribution of AMS in the field of nuclear astrophysics, where we extended the activation technique to hitherto inaccessible cases, like ^{79}Se.

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