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Investigation of ^{54}Fe(n,γ)^{55}Fe and ^{35}Cl(n,γ)^{36}Cl reaction cross sections at keV energies by Accelerator Mass Spectrometry

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Abstract.

Activations with neutrons in the keV energy range were routinely performed at the Karlsruhe Institute of Technology (KIT) in Germany in order to simulate stellar conditions for neutron-capture cross sections. A quasi-Maxwell-Boltzmann neutron spectrum of $kT = 25$ keV, being of interest for the astrophysical s-process, was produced by the ^{7}Li(p,n) reaction utilizing a 1912 keV proton beam at the Karlsruhe Van de Graaff accelerator. Activated samples resulting in long-lived nuclear reaction products with half-lives in the order of yr - 100 Myr were analyzed by Accelerator Mass Spectrometry (AMS). Comparison of the obtained reaction cross sections to literature data from previous Time-of-Flight (ToF) measurements showed that the selected AMS data are systematically lower than the ToF data. To investigate this discrepancy, ^{54}Fe(n,γ)^{55}Fe and ^{35}Cl(n,γ)^{36}Cl reaction cross sections were newly measured at the Frankfurt Neutron Source (FRANZ) in Germany. To complement the existing data, an additional neutron activation of ^{54}Fe and ^{35}Cl at a proton energy of 2 MeV was performed. The results will give implications for the stellar environment at $kT = 90$ keV, reaching the not yet experimentally explored high-energy s-process range. AMS measurements of the activated samples are scheduled.

1 Introduction

The astrophysical slow neutron-capture process (s-process) is responsible for the production of about half of the heavy elements above iron in late stellar burning phases. Typical s-process neutron energies are in the range of 10 - 100 keV. To understand the nucleosynthesis, it is essential to obtain reaction cross sections under conditions corresponding to the respective astrophysical site. As particles in stellar environment follow the Maxwell-Boltzmann distribution, the corresponding cross section is referred to as Maxwellian-Averaged Cross Section (MACS) [1, 2].

MACS can be experimentally determined measuring the total energy of prompt gammas following irradiation with a pulsed high-intensity neutron beam with a wide energy range. The energy of the individual neutrons in the pulse is determined by measuring the Time-of-Flight of the neutrons over a well-defined distance (ToF technique). The MACS can then be calculated by folding the measured energy-dependent cross sections with an appropriate Maxwell-Boltzmann distribution. The activation technique [3–5] offers a complementary approach to determine MACS. It is a two-step technique. In the first step, a sample is irradiated with a quasi-Maxwell-Boltzmann neutron spectrum. In the second step, off-line analysis of the reaction product is performed. If the reaction product is radioactive with a suitable half-life and decay characteristics, it can be counted using an adequate detector. If the reaction product is long-lived or has no measurable gamma transitions, the reaction cross section can be determined using Accelerator Mass Spectrometry (AMS) [5–8]. AMS is a unique technique offering sensitive single-atom counting. Just milligramms of target material provide sufficient amount for a measurement. Advantages of the activation technique are that very thin targets reducing scattering corrections can be used and that the direct capture component is included.

The neutron-capture cross sections of ^{54}Fe and ^{35}Cl investigated within this work are of interest for the understanding of the s-process nucleosynthesis. ^{54}Fe is depleted in the astrophysical s-process by neutron-capture and ^{35}Cl is a so-called neutron poison. It reduces the availability of neutrons for the s-process nucleosynthesis and therefore the ^{35}Cl(n,γ) reaction cross section needs to be known to constrain the significance of ^{35}Cl as neutron poison.

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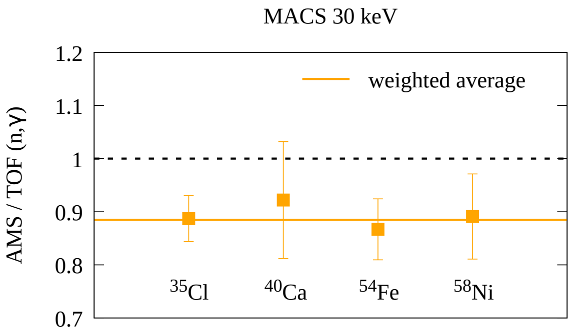

Figure 1. Ratio between the Maxwellian-averaged neutron-capture cross sections of ^{35}Cl, ^{40}Ca, ^{54}Fe and ^{58}Ni at 30 keV measured using AMS [10–13] and those measured using ToF [14–18]. The AMS data are on average 12% lower than the ToF data, for more details see [19].

2 Previous measurements

Activation and Time-of-Flight are two independent complementary techniques. Reaction cross sections of nuclides ^{35}Cl, ^{40}Ca, ^{54}Fe and ^{58}Ni were measured by both techniques, allowing comparison between them. Figure 1 shows experimentally-obtained MACS at 30 keV of the nuclides ^{35}Cl, ^{40}Ca, ^{54}Fe and ^{58}Ni measured using AMS over those using ToF. The AMS data are systematically lower, on average by 12%.

The AMS data in Fig.1 originate from activations at Karlsruhe Institute of Technology (KIT) [9] in Germany with subsequent AMS measurements at the following facilities: ^{35}Cl, ^{40}Ca and ^{54}Fe at the 3 MV Vienna Environmental Research Accelerator (VERA) in Austria [10–12] and ^{58}Ni at the 14 UD tandem accelerator of the Maier-Leibnitz-Laboratory in Garching, Germany [13].

The ToF values for neutron-capture cross sections of ^{35}Cl and ^{40}Ca were obtained using the Oak Ridge Electron Linear Accelerator in USA [14, 15] and the cross section of ^{54}Fe at the n_TOF (neutron Time-of-Flight) facility at CERN in Switzerland [16]. The cross section of ^{58}Ni was measured both at n_TOF [17] and using the Oak Ridge Electron Linear Accelerator [18] and the plotted value is based on both measurements.

3 Sample material for new measurements

3.1 ^{54}Fe

A 0.5 mm thick iron foil with a mass of 200 mg enriched in ^{54}Fe to 94.3% from ISOFLEX USA was rolled out to increase its area from 0.5 cm^2 to 6 cm^2. In this way, two foils with a radius of 1 cm, a thickness of about 45 µm and a mass of 35 mg could be produced. This procedure was needed as no foil with a sufficient size was commercially available.

3.2 ^{35}Cl

NaCl powder (CertiPUR®) with a purity of $(99.92 \pm 0.05)\%$ from Merck KGaA with a natural ^{35}Cl and ^{37}Cl composition was used to press two pellets with a diameter of 1 cm and a thickness of approximately 500 µm.

4 Experimental technique

4.1 Activation at FRANZ

The ^{54}Fe and NaCl samples were irradiated with neutrons at the Frankfurt Neutron Source (FRANZ) in Germany. Figure 2 shows a sketch of the neutron production target at FRANZ. A metallic lithium layer evaporated on a copper backing was bombarded with a continuous MeV proton beam of 10 µA using the Frankfurt 2.5 MV Van de Graaff accelerator. Neutrons were produced via the ^{7}Li(p,n) reaction. To prevent lithium from melting, the target backing was water cooled. The ^{54}Fe and ^{35}Cl samples were positioned 0.5 mm and 1 mm, respectively, from the lithium target and perpendicular to the proton beam axis.

Two independent irradiations were performed. First, at a proton energy of 1912 keV, a neutron spectrum closely approximating the Maxwell-Boltzmann distribution at $kT = 30$ keV was produced. About $1.5 \cdot 10^8$ neutrons per second were emitted and kinematically focused into a neutron cone with an opening angle of about 120° at this energy. With the chosen irradiation geometry, nearly the whole neutron cone was covered by the samples. With an irradiation time of 45 h, the neutron fluence on the ^{54}Fe and NaCl samples positioned in the neutron cone was of the order of 10^{13} cm^{-2}.

The second irradiation was performed at a proton energy of 2 MeV. At this energy, the produced neutrons are emitted under an angle larger than 90°, therefore only a part of the distribution reaches the samples. The irradiation geometry was chosen in a way to reach the not yet experimentally explored high energy s-process range around $kT = 90$ keV. About $3 \cdot 10^8$ neutrons per second were emitted and approximately 75% of them hit the samples. At 2 MeV, the neutron fluence on the targets in 50 h activation time was of the order of 10^{13} cm^{-2}.

Each sample was placed between gold foils with a thickness of 25 µm and the exact same size as the samples. The well-known neutron-capture cross section of ^{197}Au [2] was used to determine the neutron fluence on the respective sample. The activity of the Au foils was counted after the activation using a high-purity germanium detector.

4.2 AMS measurements at HIAF and HZDR

4.2.1 Chemical sample preparation

After irradiation, the samples were dissolved and converted to a chemical form suitable for AMS measurements (iron oxide Fe_2O_3 in case of iron, and silver chloride AgCl in case of chlorine [21, 22]). The reaction product of the neutron-capture on ^{54}Fe is the radioisotope ^{55}Fe with a half-life of (2.7563 ± 0.0004) yr [23]. The stable isobar of iron, ^{55}Mn, is orders of magnitude more abundant than ^{55}Fe. But in this case, the AMS measurement is not interfered by ^{55}Mn, because it does not form negative ions

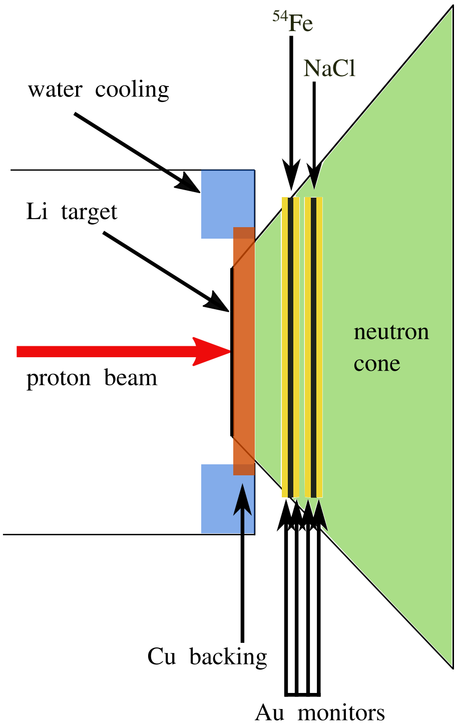

Figure 2. Schematic setup for the neutron production target at the FRANZ facility and the NaCl and ^{54}Fe irradiation geometry.

[24, 25]. In such a case, the chemical preparation of the AMS samples is straightforward: Fe foils are dissolved in HCl with a few drops of H_2O_2 to accelerate the oxidation process and subsequent addition of $NH_{3(aq)}$ for hydroxide precipitation. Finally, the solution is centrifuged and the precipitate washed with deionised water and dried.

However, there is a strong isobaric interference from ^{36}S, the stable isobar of ^{36}Cl (half-life $(3.013 \pm 0.015) \cdot 10^5$ yr [26]), which is the product of neutron-capture on ^{35}Cl. In this case, chemical reduction of ^{36}S is needed to lower the background and enable AMS measurements of ^{36}Cl. First, the irradiated NaCl pellet is dissolved in deionised H_2O and AgCl is precipitated adding slightly acidic $AgNO_3$ solution. To purify the AgCl from sulfur, AgCl is first dissolved in $NH_{3(aq)}$. $Ba(NO_3)_2$ is added and $BaSO_4$ co-precipitates with $BaCO_3$ (CO_2 from air). $BaSO_4$ is then removed by filtration. Afterwards, AgCl is reprecipitated by adding HNO_3 and the precipitates are washed and dried.

4.2.2 Future AMS measurements

Subsequent AMS measurements of the chemically-processed samples will be performed at the 14 UD tandem accelerator at the Heavy Ion Accelerator Facility (HIAF) at the Australian National University in Canberra [27] and at the Helmholtz-Zentrum Dresden-Rossendorf (HZDR) in Germany [28, 29].

The expected isotopic ratios are both in the order of $\frac{^{55}\mathrm{Fe}}{^{54}\mathrm{Fe}} \sim \frac{^{36}\mathrm{Cl}}{^{35}\mathrm{Cl}} \sim 10^{-13}$. The neutron-capture cross sections $\sigma_{\mathrm{reaction}}$ will be derived from the isotopic ratio and the neutron fluence (see Section 4.1) [6] using the following equation

$$\sigma_{\mathrm{reaction}} = \frac{N_{\mathrm{prod}}}{N_{\mathrm{target}}} \cdot \frac{1}{\Phi_{\mathrm{n}}}, \qquad (1)$$

with N_{prod} being the number of produced atoms, N_{target} the number of atoms in the target and Φ_{n} the neutron fluence.

5 Summary

The neutron-capture cross sections of $^{54}\mathrm{Fe}(\mathrm{n},\gamma)^{55}\mathrm{Fe}$ and $^{35}\mathrm{Cl}(\mathrm{n},\gamma)^{36}\mathrm{Cl}$ reactions were measured using activation at the Frankfurt Neutron Source (FRANZ) in Germany. Subsequent single-atom counting via AMS is scheduled at the Heavy Ion Accelerator Facility at the Australian National University in Canberra and Helmholtz-Zentrum Dresden-Rossendorf in Germany. The aim of this experiment is to investigate the discrepancy between previous measurements performed using the Time-of-Flight (ToF) technique and the activation technique followed by Accelerator Mass Spectrometry (AMS) as the results of the ToF measurements are on average 12% higher. Additionally, neutron-capture cross sections of ^{54}Fe and ^{35}Cl in the not yet experimentally explored high-energy range of the s-process of about $kT = 90$ keV were measured.

The activation setup for this experiment was analogous to the previous setup at the Karlsruhe Van de Graaff accelerator which led to discrepant cross section values compared to ToF measurements. Using the same activation technique and the same setup at a different facility might help to reveal the reason for the not-understood discrepancy between the ToF and the AMS data. The activated samples have been chemically processed and AMS measurements of the samples are scheduled to take place.

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