

## Determination of the stellar ( $n, \gamma$ ) cross section of $^{54}\text{Fe}$ with accelerator mass spectrometry

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The activation technique has been combined with accelerator mass spectrometry (AMS) for investigating the stellar ( $n, \gamma$ ) cross section of  $^{54}\text{Fe}$  at  $kT=25$  keV. Our measurement is based on the detection of  $^{55}\text{Fe}$  atoms directly via AMS, which has the advantage of being independent of half-lives and decay intensities. The neutron activations were performed at the Karlsruhe 3.7 MV Van de Graaff accelerator using the quasi-stellar neutron spectrum of  $kT=25$  keV produced by the  $^7\text{Li}(p, n)^7\text{Be}$  reaction. The subsequent AMS measurements were done at the Vienna Environmental Research Accelerator (VERA). The determination of the stellar neutron cross section via the combination of the activation technique and AMS represents an important complement to previous TOF measurements, since this independent approach implies different systematic uncertainties, much higher sensitivity, and includes the contribution from the direct capture channel as well. Our preliminary Maxwellian averaged cross section at  $kT=30$  keV yielded  $\langle \sigma \rangle_{30 \text{ keV}} = 30.3 \pm 3.0$  mbarn, in good agreement with previous values.

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## 1. Introduction

The production of the iron group elements was already assumed by Burbidge, Burbidge, Fowler and Hoyle [1] to occur in a high density, high temperature environment, the so-called  $e$ -process. This picture was detailed later and involves now the more general concept of nuclear statistical equilibrium (NSE). In silicon burning, the last hydrostatic burning stage in a pre-supernova star [2],  $^{28}\text{Si}$  is not directly fused to  $^{56}\text{Fe}$ , because this would require temperatures of  $>3.5$  GK for penetrating the Coulomb barrier. Instead, photodisintegration plays a dominant role, since the number of photons scales with  $T^4$ . Nuclei are partly decomposed into  $p$ ,  $n$ , and  $\alpha$  particles, which are eventually recaptured to build heavier elements up to the iron region. Consequently, the production of  $^{54}\text{Fe}$  takes mainly place during explosive oxygen and silicon burning at low entropies, since otherwise  $^{58}\text{Ni}$  is produced by  $\alpha$  capture [3].

Beyond iron no more energy can be gained by fusion, and further charged-particle reactions are hindered by the large Coulomb barriers. The elements heavier than iron are thus mainly produced by neutron captures in the "slow" ( $s$ ) and "rapid" ( $r$ ) processes.

In this context, neutron capture cross sections are required in the neutron energy range  $1 \leq E_n \leq 300$  keV [7]. These cross sections can be determined by means of different experimental techniques, either directly by measuring the prompt  $\gamma$ -rays associated with the neutron capture reaction or via the induced activity of the product nuclei. The first technique is particularly suited in connection with the time-of-flight (TOF) method [4, 5, 6]. 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 as well as access to partial cross sections leading to isomeric states.

The present measurement on  $^{54}\text{Fe}$  was performed by counting the product nuclei via accelerator mass spectrometry (AMS), which allows one to extend the activation technique to hitherto inaccessible cases, e.g. to reactions producing very long-lived nuclei with correspondingly low activities or to reactions leading to nuclei with very weak or completely missing  $\gamma$ -transitions. The AMS technique has the further advantage of being independent of uncertainties related to  $\gamma$ -ray intensities and half-lives. It combines the high efficiency of mass spectrometry with excellent discrimination against isobaric and molecular interferences. This is achieved by using negative ions, by dissociation of molecular ions and by stripping ions to high positive charges after a first stage of acceleration, and by using a second stage of acceleration for the final identification of individual ions (see also the contribution of A. Wallner *et al.*).

We report on an independent approach to verify the recommended cross section [7] by measuring this cross section with the powerful combination of the activation technique and AMS.

## 2. Experimental technique

The iron samples were activated at the 3.7 MV Van de Graaff accelerator of Forschungszentrum Karlsruhe. Since  $^{55}\text{Fe}$  decays into the ground-state of  $^{55}\text{Mn}$  ( $t_{1/2} = 2.73$  yr) without  $\gamma$ -ray emission, AMS was required to count the number of activated nuclei [8]. This part has been done at the VERA laboratory in Vienna.

Neutrons were produced via the  $^7\text{Li}(p, n)^7\text{Be}$  reaction by bombarding 30  $\mu\text{m}$  thick layers of metallic Li (or 10  $\mu\text{m}$  layers of LiF) on a water-cooled Cu backing with protons of 1912 keV, 31 keV above the  $(p, n)$  reaction threshold at 1881 keV. The produced neutrons are kinematically collimated in a forward cone with 120° opening angle. With these parameters we obtain a quasi-stellar spectrum for a thermal energy of  $kT=25\pm 0.5$  keV [9]. The mean neutron flux during the irradiations was  $\approx 10^9$  n/s at the position of the samples, which were placed in close geometry to the Li target. The samples were sandwiched between 10-30  $\mu\text{m}$  thick gold foils of the same diameter. The cross section of  $^{197}\text{Au}(n, \gamma)$  is used as reference, since it is known for this experimental neutron spectrum with high accuracy [9].

Two different types of commercially available metallic Fe powder from Merck ( $\text{Fe}_M$ ) and Alfa Aesar ( $\text{Fe}_\alpha$ ) with natural abundances (5.8%  $^{54}\text{Fe}$ ) were used for the two irradiations. The activation measurements were carried out with the Van de Graaff accelerator operated in DC mode with a beam current of 80-90  $\mu\text{A}$ . During the irradiations, the neutron flux history was registered in intervals of 90 s by a  $^6\text{Li}$ -glass detector in 1 m distance from the neutron target. With this information it is possible to correct the fraction of decays during irradiations properly, including the fact that the Li and LiF targets degrade during the activation.

The isotopic ratio  $\frac{Z(^{55}\text{Fe})}{N(^{54}\text{Fe})}$  was measured by AMS at the Vienna Environmental Research Accelerator (VERA). The typical measurement procedure is the following: negatively charged ions are produced in a cesium sputter source and are pre-accelerated before they pass a low energy mass spectrometer, which analyses a specific mass. The ions are further injected into a tandem accelerator (terminal voltage: 3 MV for Fe). Any molecules that might contribute to a molecular interference are completely destroyed in the terminal stripper of the accelerator. Due to this stripping process, only positively charged ions leave the tandem accelerator. A specific charge state (e.g.  $^{55}\text{Fe}^{3+}$ ) is selected by a second analysing magnet. Stable isotopes will be measured as currents using Faraday cups, while radionuclides are counted directly with a particle detector. In the case of  $^{54}\text{Fe}$ , the isotopic ratio  $\frac{Z(^{55}\text{Fe})}{N(^{54}\text{Fe})}$  has so far been measured relative to an AMS standard produced by irradiation of Fe powder samples with thermal neutrons at the TRIGA Mark-II reactor of the Atominstitut der Österreichischen Universitäten in Vienna to obtain  $^{55}\text{Fe}$  via the  $^{54}\text{Fe}(n, \gamma)$  reaction. Therewith, reference materials with  $\frac{Z(^{55}\text{Fe})}{N(^{54}\text{Fe})}$  isotope ratios between  $10^{-10}$  and  $10^{-12}$  were obtained. Usually, the range of isotope ratios measured with AMS is of the order of  $10^{-10}$  to  $10^{-15}$  (for further details, see the contribution of A. Wallner to these proceedings, "AMS measurements of stellar cross sections across the nuclear chart").

### 3. Preliminary results

Our experimental cross section  $\sigma_{exp}$  is deduced from the isotopic ratio  $\frac{Z(^{55}\text{Fe})}{N(^{54}\text{Fe})} = \sigma_{exp} \Phi_{tot}$ , where  $N$  is the number of  $^{54}\text{Fe}$  atoms in the sample and  $\Phi_{tot} = \int \phi(t) dt$  the time-integrated total neutron flux. The isotopic ratio of the two samples has been determined by AMS. The neutron flux is determined by the activity of the gold foils.

The experimental neutron spectrum of the  $^7\text{Li}(p, n)^7\text{Be}$  reaction approximates a Maxwellian distribution with  $kT=25$  keV almost perfectly [9]. However, the cut-off at  $E_n = 106$  keV and small differences compared to the true thermal spectrum require a correction that is determined in the following way: The neutron energy-dependent cross section  $\sigma_{eval}(E_n)$  from databases like

JEFF 3.1, JENDL 3.3, or ENDF-B/VI.8 is used to calculate a normalization factor  $NF$  to the experimental cross section. To obtain this normalization factor  $NF = \frac{\langle \sigma_{eval} \rangle}{\sigma_{exp}}$  the evaluated cross section  $\sigma_{eval}(E_n)$  is folded with the experimental neutron spectrum of [9]. The proper normalized Maxwellian averaged cross section  $\langle \sigma \rangle_{kT}$  is then:

$$\langle \sigma \rangle_{kT} = \frac{2}{\sqrt{\pi}} \cdot \frac{\int_0^\infty \frac{\sigma_{eval}(E_n)}{NF} \cdot E_n \cdot e^{-E_n/kT} \cdot dE_n}{\int_0^\infty E_n \cdot e^{-E_n/kT} \cdot dE_n},$$

where  $E_n$  is the neutron energy, and  $E_n \cdot e^{-E_n/kT}$  is the Maxwellian distribution corresponding to the thermal energy  $kT$ . In the astrophysically relevant energy range the evaluated data files for  $^{54}\text{Fe}$  are identical. The respective cross section derived with the evaluated data is  $\sigma_{eval} = 22.2$  mbarn. This yields normalization factors close to  $NF = 0.7$  (Table 1). The preliminary mean value for the MACS at 30 keV is  $\langle \sigma \rangle_{30 \text{ keV}} = 30.3$  mbarn.

**Table 1:** Activation parameters and preliminary results from the  $^{54}\text{Fe}$  measurement.

Sample	$t_{act}$ [d]	$\Phi_{tot}$	Isotopic ratio $^{55}\text{Fe} / ^{54}\text{Fe}$	$\sigma_{exp}$ [mbarn]	$NF$	$\langle \sigma \rangle_{30 \text{ keV}}$ [mbarn]
$\text{Fe}_M$	10.4	$0.80 \times 10^{15}$	$2.52 \times 10^{-11}$	31.3	0.709	30.8
$\text{Fe}_\alpha$	15.4	$1.37 \times 10^{15}$	$4.15 \times 10^{-11}$	30.3	0.733	29.8
mean value:						$30.3 \pm 3.0$

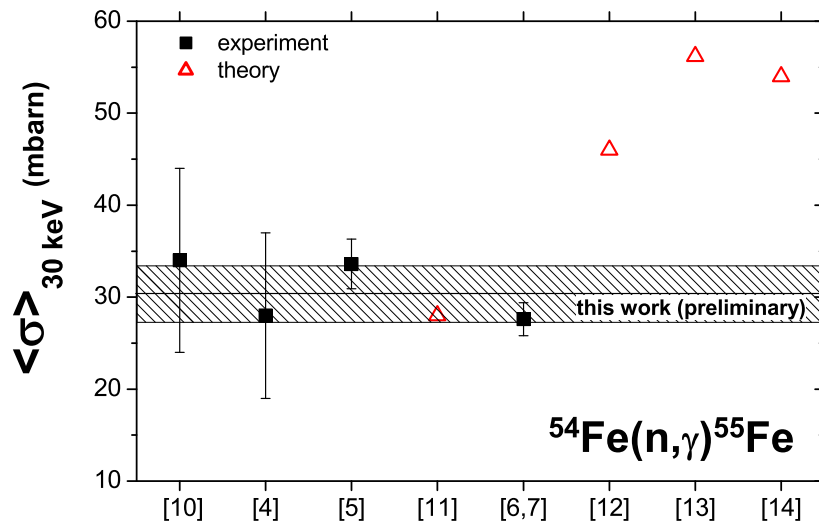
This preliminary experimental cross section is compared with previous experimental and theoretical values in Fig 1. The uncertainty of our new value is largely dominated by the uncertainty of the absolute value of our current Fe standard material. Our result is in good agreement with all previous TOF values [4, 5, 6], and the "older" theoretical predictions from Allen et al. [10] and Woosley et al. [11], whereas more recent calculations using Hauser-Feshbach codes, NON-SMOKER [12] and MOST [13, 14] deviate considerably. Moreover, the accuracy of our data will be further improved to  $<10\%$  by using a new  $^{55}\text{Fe}$  standard produced via nuclear reactions.

#### 4. Conclusion

The combination of the highly sensitive AMS technique with the activation method allowed us to measure the MACS at 30 keV of the reaction  $^{54}\text{Fe}(n, \gamma)^{55}\text{Fe}$ . Our preliminary result of  $\langle \sigma \rangle_{30 \text{ keV}} = 30.3 \pm 3.0$  mbarn is in good agreement with previous experimental data, although some significant discrepancies remain compared to recent theoretical calculations. The AMS technique is a powerful tool for the detection of long-lived radionuclides through ultra-low isotope ratio measurements. In combination with the activation technique this method will be used in future measurements for determining the stellar  $(n, \gamma)$  cross sections of  $^9\text{Be}$ ,  $^{13}\text{C}$ ,  $^{35}\text{Cl}$ , and  $^{62}\text{Ni}$  as well.

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**Figure 1:** Comparison of the experimental result of the  $^{54}\text{Fe}(n, \gamma)^{55}\text{Fe}$  cross section with previous data (ordered according to date of publication).

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