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2010 J. Phys.: Conf. Ser. 202 012020

(http://iopscience.iop.org/1742-6596/202/1/012020)

Precise Measurement of the Neutron Capture Reaction ^{54}Fe(n,γ)^{55}Fe via AMS

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Abstract. The measurement of cross sections relevant to nuclear astrophysics has become one main research topic at the VERA (Vienna Environmental Research Accelerator) facility. The technique applied, accelerator mass spectrometry (AMS), offers excellent sensitivity for the detection of long-lived radionuclides through ultra-low isotope ratio measurements. We discuss the potential and preliminary results of ongoing precision measurements of neutron-capture cross sections of ^{54}Fe. Such measurements might help to clarify the recently found discrepancy of s-process nucleosynthesis at lower-mass nuclei (A<120), which might be attributed to a systematic offset in previous experimental data. Samples were irradiated with neutrons from thermal to MeV energies. After the irradiations, the amount of produced long-lived ^{55}Fe ($t_{1/2}$ = 2.72 yr) was analyzed using AMS. At VERA, detection of ^{55}Fe was developed with a reproducibility of about 1%, which makes the ^{54}Fe(n,γ)^{55}Fe reaction a precise and unique laboratory measurement, which can serve as a reference to complementary techniques. In this regard a new ^{55}Fe standard for AMS measurements was produced. The final cross-section data are expected to be accurate to better than 3%. We report a preliminary, however, already significantly improved thermal neutron cross section value of (2.32 ± 0.10) barn, and a value of (6.3 ± 0.6) mbarn for E_n = (520 ± 50) keV.

1. Introduction

Recent accurate abundance observations in very rare, ultra metal-poor (UMP) stars in the galactic halo showed abundance patterns that scale exactly with the solar r component [1]. This holds for elements heavier than barium, however, for lighter elements significant discrepancies of the order of 20% were found from this observation. A similar discrepancy is found for s-only isotopes lighter than barium (see [2]). These facts hint to some deficiency in the standard description of s- or r-process nucleosynthesis or there is a systematic offset in the experimental data. The latter can be tested by precision measurements of neutron-capture cross sections, e.g. the ^{54}Fe(n,γ)^{55}Fe reaction: Via the weak s-process massive stars contribute dominantly to the abundance distribution between Fe and Zr. For those nuclides, however, neutron capture cross sections are poorly known compared to the required accuracy of 3-5% [3].

For specific reactions accelerator mass spectrometry (AMS) offers a powerful tool to measure cross sections independent of the half-lives of the reaction products. Activation with subsequent AMS measurements have been applied for isotopes where off-line decay counting is difficult or impossible due to long half-lives or due to the absence of suitable γ-ray transitions (see e.g. [4,5], for a detailed list see [6].) In particular, measurements on ^{55}Fe allow producing highly precise data. AMS will therefore allow one to identify possible hidden systematic uncertainties associated with different detection methods and, in special cases, to reduce the total uncertainty of cross section measurements to a few %. A summary of AMS measurements at the VERA facility (Vienna Environmental Research Accelerator) studying neutron-induced cross-sections of relevance in nuclear astrophysics is given in Tab. 1.

Table 1. Summary of AMS measurements at VERA related to neutron capture studies of interest to nuclear astrophysics. FZK, ATI and IKI denote neutron irradiations performed at Forschungszentrum Karlsruhe ("keV neutrons"), at the Atominstitut, Vienna ("thermal neutrons"), and at the Budapest Research Reactor ("cold neutrons"), respectively.

Reaction	Irradiation facility	neutron energy
^{9}Be(n,γ)^{10}Be	FZK, ATI	thermal, 25 keV, 520 keV
^{13}C(n,γ)^{14}C	FZK, ATI	thermal, 25 keV, 128 keV, 167 keV
^{14}N(n,p)^{14}C	FZK, ATI	thermal, 25 keV, 123 keV, 178 keV, 14 MeV
^{40}Ca(n,γ)^{41}Ca	FZK	25 keV
^{54}Fe(n,γ)^{55}Fe	FZK, ATI, IKI	cold, thermal, 25 keV, 520 keV, 14 MeV
^{209}Bi(n,γ)^{210m}Bi	FZK, ATI	thermal, 25 keV

2. Comparison of AMS measurements to other techniques

Cross section measurements can be classified into two complementary techniques: direct and indirect methods. The direct method makes use of the detection of the prompt and characteristic radiation associated with the production of a specific nuclide, or selectively detects the reaction product itself. A second and independent method makes use of the activation technique, with sample irradiation and subsequent measurement of the reaction product. After the irradiation the number of produced radioactive nuclei can be quantified either by decay counting or by mass spectrometric methods. This method is mostly restricted to radioactive products; however, it represents a very sensitive technique due to potential long irradiation periods. With the dedicated activation setup at Karlsruhe, stellar conditions can directly be simulated by generating a quasi-stellar spectrum (see e.g. [7,8]), and consequently, that reaction rate directly scales with stellar rates.

The determination of the stellar cross sections via the combination of the activation technique and AMS is complementary to direct measurements, since this independent approach implies different systematic uncertainties. In this regard, precision measurements of neutron-capture cross sections of ^{54}Fe are in progress to clarify the recently found discrepancy of s-process nucleosynthesis at lower-mass nuclei (A<120; see above). The potential and power of AMS for detecting the product nuclei ^{55}Fe has been demonstrated recently in a first irradiation campaign [9,10].

3. Activation and AMS measurement

Several neutron activations in combination with subsequent AMS measurements at VERA were performed recently for studying neutron capture on ^{54}Fe: Thermal neutrons and cold neutrons were produced at the Atominstitut of the Vienna University of Technology, and the Budapest Research Reactor (IKI, Institute of Isotopes, Hungarian Academy of Sciences) [11]. "keV neutrons" were produced at Forschungszentrum Karlsruhe (FZK) [7] via the ^{7}Li(p,n)^{7}Be reaction: By choosing a proper irradiation geometry, a quasi-stellar neutron spectrum was generated (by simulating a Maxwellian-Boltzmann distribution) of kT = 25 keV; and by higher-energetic protons neutron energies of 520 keV were produced as well. In addition, neutron activations in the fast neutron energy range (14-MeV) were performed in cooperation with TU Dresden. Depending on the cross-sections, isotope ratios ^{55}Fe/^{54}Fe between 10^{-11} and 10^{-12} were produced in such activations. After the neutron irradiations, the subsequent AMS measurements were performed at the VERA facility in Vienna.

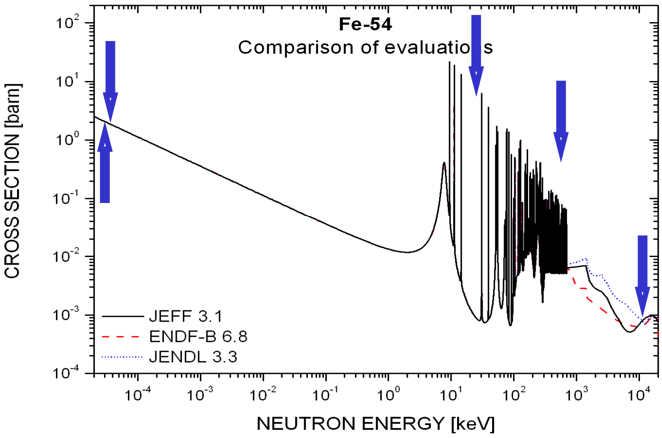

Figure 1. Neutron capture cross section for ^{54}Fe(n,γ)^{55}Fe. Plotted are evaluations [12]; the energies of new activation/AMS data are indicated by arrows.

In Fig. 1 these neutron energies are schematically indicated by arrows. Also plotted are results from semi-empirical evaluations [12], which indicate discrepancies in the energy region above a few hundred keV. With these "activation"-anchors, distributed over a wide energy range, AMS data can be compared to continuous data obtained with time-of-flight techniques.

4. AMS measurement of ^{55}Fe

AMS represents a mass spectrometric method based on a tandem accelerator. Negatively charged ions are produced in an ion source requiring mg mass of solid sample. Extraction of Fe^{-} completely suppressed the stable isobar of ^{55}Fe, because ^{55}Mn does not form stable negative ions [13]. At VERA ^{55}Fe-measurements on the level of 1% reproducibility with a background ^{55}Fe/^{56}Fe < 2×10^{-15} can be performed (for more details see [10]). In this regard, an important aspect is the availability of accurate ^{55}Fe reference materials for AMS. In general, a typical iron sample with an isotopic ratio ^{55}Fe/^{56}Fe of 10^{-12} will result in a ^{55}Fe count rate of typically one event per 10 second. Accordingly, a background ratio ^{55}Fe/^{56}Fe of 1×10^{-15}, corresponds to a rate of ≈ 1 count per 3 hours measuring time.

Important ingredients for AMS measurements are reference samples to which the measured results of unknowns are normalized. AMS does not allow deducing accurate data directly from the measured quantities, particle count rate (^{55}Fe) and ion currents (^{54,56}Fe), because mass fractionation, machine instabilities etc. are difficult to quantify at levels better than 5 – 10%. For ^{55}Fe measurements no well-known reference material was available. Therefore, we produced our own in-house standard. Best results were obtained from a ^{55}Fe activity solution, certified by PTB Braunschweig, Germany [14]. This solution was stepwise diluted with Fe of natural isotopic composition. From the certified activity solution we produced ^{55}Fe AMS reference materials with an isotope ratio known to ±1.5%, and basically limited by the uncertainty of the certified value.

5. Results

The neutron capture cross section of the ^{54}Fe(n,γ)^{55}Fe reaction can directly be calculated from the neutron fluence and the isotope ratio measured with AMS. Interestingly, the recommended value for the thermal capture cross section of (2.25±0.18) barn [15] is known with an accuracy of only 8%. Our preliminary result for the thermal cross section is based on samples activated at the NIPS station [11] of the Budapest Research Reactor and at the TRIGA Mk II research reactor at the Atominstitut, Vienna. Fe metal pellets of natural isotopic composition, and metallic Fe foils, consisting of highly enriched ^{54}Fe, were exposed to a neutron fluence of $10^{13} – 10^{14}$ n cm^{-2}. From the AMS measurements, we obtained a thermal value of (2.33 ± 0.10) barn, in good agreement with the recommended value for ^{54}Fe(n,γ)^{55}Fe of (2.25 ± 0.18) barn [15]. Full data analysis will result in a final uncertainty of ≈ 2 %.

At Forschungszentrum Karlsruhe Fe pellets were activated with neutrons of kT ≈ 25 keV; and with neutrons of 520 keV energy. In a previous publication [9] we presented first preliminary results for 25 keV. Here, we add new data from AMS measurements performed relative to the new ^{55}Fe reference material. Our new results for 520 keV indicate a cross section value of (6.3 ± 0.6) mbarn. These preliminary results confirm previous ones, however, already with improved accuracy. The full set of acquired data is being analyzed now, with the goal to reduce uncertainties to 2-3%.

6. Conclusion

The neutron capture reaction ^{54}Fe(n,γ)^{55}Fe represents an excellent candidate for comparing different and independent methods for cross section measurements. AMS detection of ^{55}Fe combined with neutron activations has been performed at the VERA laboratory, where ^{55}Fe detection was demonstrated to be precise at a level of 1%. The preparation of a well-known reference material allows now to deduce accurate cross-section data over a wide neutron energy range. While activation allows one only to gain information on cross section values for selected neutron energies, time-of-flight based techniques will provide continuous data over a wide energy range. However, this more complicated technique can be checked and normalized with AMS data. In particular, such comparisons are important to clarify the recently debated discrepancy of measured solar-system abundances and observed abundances in ultra-metal poor stars.

Acknowledgement: Part of this work was funded by the Austrian Science Foundation (FWF), No. P20434.

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