s-process nucleosynthesis in massive stars: new results on ^{60}Fe , ^{62}Ni and ^{64}Ni

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Abstract. The *s* process synthesizes the elements between Fe and Sr in massive stars during two major evolutionary stages, convective core He burning and C shell burning. This scenario implies fascinating consequences for the chemical evolution of the star. For instance, the neutron capture rate at each isotope can have a big influence on the production of many of the subsequent higher mass isotopes. Correspondingly, one needs to know the (n, γ) cross sections of the involved isotopes with high accuracy in order to determine the abundance pattern reliably and to obtain a consistent picture of this stage. This contribution gives an overview on recent and future experiments for the Fe/Ni nucleosynthesis in massive stars. New results on ^{60}Fe , ^{62}Ni and ^{64}Ni are reported. ^{60}Fe is mostly produced during the short convective C shell burning phase, where peak densities of $\sim 10^{11} \text{ cm}^{-3}$ are reached, prior to the SN explosion. The stellar (n, γ) cross section of ^{60}Fe could be measured with a 1 μg sample obtained at PSI (Switzerland), which was sufficient for an activation measurement using the intense, quasi-stellar neutron field for a thermal energy of 25 keV at the Karlsruhe Van de Graaff accelerator. The FZK accelerator was also used for an activation of ^{62}Ni , whereas in this case, the number of ^{63}Ni nuclei produced were determined via accelerator mass spectroscopy at the Maier-Leibnitz-Laboratorium in Garching/Munich. The (n, γ) cross section of ^{64}Ni at a stellar temperature equivalent to 50 keV has been measured in a collaboration between FZK Karlsruhe and PTB Braunschweig. Finally, complementary time of flight measurements on the Fe and Ni isotopes over a broad energy range are planned at the white neutron source n_TOF of CERN for the future campaign in 2009.

Keywords: nuclear astrophysics, neutron capture cross section, ^{60}Fe , ^{62}Ni , ^{64}Ni

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INTRODUCTION

By means of germanium spectrometers on board of the two satellites RHESSI [1] and INTEGRAL [2] it has been possible to measure, towards the inner radiant of our Galaxy, the γ -ray flux from the decay of ^{60}Fe ($t_{1/2} = 1.5 \times 10^6$ years) and ^{26}Al ($t_{1/2} = 7.16 \times 10^5$ years). The results from these observations can be compared to theoretical predictions, which include models for nucleosynthesis during stellar evolution and in the

SN explosion. The most promising scenario for the nucleosynthesis of these two isotopes has been described in a recent paper [3]. According to the latter calculations ^{26}Al is produced by means of proton captures in the SN explosion, whereas ^{60}Fe seems to be synthesized mostly by means of neutron capture reactions during shell Carbon burning and also, in the case of stars with $M > 40 M_{\odot}$, during shell He burning [3]. Therefore, the main nuclear data input needed in the prediction of the ^{60}Fe abundance is the (n, γ) cross section (CS). Indeed, the production and destruction of ^{60}Fe are determined by the CS of ^{59}Fe and ^{60}Fe , respectively. The CSs of these two isotopes were not measured before because of the difficulty to produce a sample of these radioactive materials. Recently a sample of ^{60}Fe could be made at PSI [4] thus allowing, for the first time, to experimentally determine its CS. This measurement is described in the first section.

Another long-standing problem in the nucleosynthesis of the Ni isotopes is that they seem to be excessively overproduced by current calculations based on state-of-the-art massive star models [5, 6]. Furthermore, during massive star evolution the neutron exposure is so small, that the reaction flow does not reach equilibrium. Due to this effect, the stellar reaction rate at one particular isotope affects the nucleosynthesis of many heavier elements (see for example Fig. 3 of Ref. [7]). This means that the CS of all the involved isotopes are needed in order to predict the observed abundances reliably.

The Maxwellian averaged cross sections (MACS) reported in the literature before 2008 for ^{62}Ni showed discrepancies of more than a factor of two, as discussed below in the second section. In order to improve the situation a new measurement of the ^{62}Ni CS was carried out in a collaboration between the Forschungszentrum Karlsruhe (FZK) and the Maier-Leibnitz-Laboratorium (MLL) in Garching. This measurement, which is described in the second section, combines the activation technique with Accelerator Mass Spectrometry (AMS), and it is similar to a measurement carried out in 2005 in a collaboration between FZK and Argonne National Laboratory [7].

The previous two activation measurements on ^{60}Fe and ^{62}Ni were carried out using a quasi-stellar neutron spectrum, which simulates the Maxwellian neutron exposure during core He- and shell He-burning in the star, at temperatures of $kT = 25$ keV. However, these isotopes are further synthesized during shell Carbon burning at higher stellar temperatures of $kT = 90 - 120$ keV. Extrapolations of the CS value measured at 25 keV to this higher energy range are uncertain because the CS of these isotopes at high neutron energies are dominated by resonances. In order to gain knowledge on the CS of ^{64}Ni at neutron energies higher than 25 keV, an activation measurement was made at the Physikalisch-Technische-Bundesanstalt (PTB) in Braunschweig. In the latter, a tritium target was used for the neutron production, which simulates a Maxwellian energy spectrum at $kT = 52$ keV. This experiment is described below in the third section.

In general, calculations of stellar nucleosynthesis in massive stars need the corresponding CSs over a broad energy range in order to determine the production yields during core He burning ($kT = 25$ keV) and during the subsequent shell C-burning stage ($kT = 90$ keV). By means of time of flight (TOF) measurements it is possible to cover completely the energy region of interest. However, in the measurement of the Fe and Ni isotopes there are two main experimental difficulties. On one hand, the highly resonance dominated cross section requires excellent TOF (or neutron energy) resolution. On the other hand, most of the resonances show a dominant neutron scattering channel. Those scattered neutrons can be easily captured in the materials surrounding the sample,

thus contaminating the measurement and artificially increasing the reaction rate. These two problems can be efficiently reduced at the CERN n_TOF neutron time of flight facility [8]. At n_TOF a very long flight path of 185 m is available, which ensures the required neutron energy resolution. A capture detection system at n_TOF is available, which consists of two home-made C₆D₆ detectors with a highly suppressed neutron sensitivity [9]. By means of such an improved setup, corrections for scattered neutrons are expected to be a few percent only. A proposal has been accepted at CERN in order to measure the neutron capture cross section of the Fe and Ni isotopes during the next campaign in 2009. More details about these experiments are discussed in the fourth section of this contribution.

⁶⁰Fe(*n*, γ)

A sample of ⁶⁰Fe ($t_{1/2} = 1.5(3) \times 10^6$ a) was made from about $7.8(1.6) \times 10^{15}$ atoms of this isotope, which could be chemically isolated from a copper proton-beam dump at PSI [4]. Apart from the ⁶⁰Fe atoms, the diluted HCl solution contained also some other spallation products, like ⁵⁵Fe, and ⁶⁰Co from the decay of ⁶⁰Fe. At present, in order to determine the ⁶⁰Fe (*n*, γ) cross section from such a challenging sample, only the highly selective and very sensitive activation technique can be applied.

For the neutron activations the quasi-stellar neutron field for a thermal energy of 25 keV at the FZK Van de Graaff accelerator was used [10]. A very intense neutron field of $\sim 10^9$ n/s was obtained by means of the ⁷Li(*p*, *n*)⁷Be reaction using a water cooled ⁷Li-target. Neutrons are emitted in a 120° cone, which was fully covered by the 6 mm diameter ⁶⁰Fe sample. The latter was sandwiched between two 30 μ m thick gold foils, which were used to determine the absolute value of the neutron fluence.

⁶¹Fe shows a half-life of only 6 minutes and hence, a large number (47) of activation runs were carried out, each irradiation lasting for 15 minutes. Time intervals between 30 seconds and 1 minute were needed to bring the sample from the experimental hall to the counting room, where the γ -rays from the decay of the excited states in the daughter nucleus ⁶¹Co were measured by means of two clover detectors, each with 120% relative efficiency. The irradiated sample was mounted in a holder, which ensured exact positioning with a precision of 0.1 mm. From both, the single spectrum of the 1205 keV γ -ray line, and the coincidence spectrum between 1027 keV and 298 keV γ -rays, the Maxwellian cross section at 25 keV was determined as $\langle \sigma v \rangle / v_T = 9.9 \pm 2.8$ (sys) ± 1.4 (stat) mb. Details about this measurement and the related analysis can be found in Ref. [11]. Several experimental effects like γ -ray absorption in the sample, γ -ray summing and backgrounds were addressed by means of detailed GEANT4 [12] Monte Carlo simulations. A theoretical (Hauser-Feshbach) description for the energy dependence of the ⁶⁰Fe cross section was needed in order to derive the Maxwellian cross section from the measured value, which corresponds to the experimental quasi-stellar neutron spectrum [10]. This correction factor was only 3%, and its uncertainty is much smaller than the rest of uncertainties in the measurement. The systematic uncertainty of almost 30% is dominated by the uncertainty on the half-life of ⁶⁰Fe, which is about 25%. This half-life was used to determine the initial ⁶⁰Fe atoms in the sample, which were calculated from the ingrowth of ⁶⁰Co. The accuracy of

the cross section result reported here will be improved when a more accurate value for this half-life becomes available.

From the point of view of the nuclear physics input, the main uncertainty on the determination of the ^{60}Fe abundance seems to be now related to other reaction rates like the cross section of the preceeding nucleus ^{59}Fe and also the reactions connected with the neutron source, $^{22}\text{Ne}(\alpha, n)$ and $^{22}\text{Ne}(\alpha, \gamma)$.

$^{62}\text{Ni}(n, \gamma)$

Until 2005 the values for the ^{62}Ni Maxwellian cross section at 25 keV reported in the literature ranged between ~ 12 mb and ~ 35 mb [13–15]. In 2005 a new measurement was reported, which was the first of its kind by combining activation with AMS [7]. However, about one month later only, the result from another TOF measurement was published [16], which was not in agreement with the result of Ref. [7]. Recently, a TOF measurement [17] has been published, which is in agreement with the activation value [7] but not with the TOF result of Ref. [16].

In view of these discrepancies a new activation measurement, similar to that of Nassar *et al.* [7], was carried out in a collaboration between Forschungszentrum Karlsruhe and the Maier-Leibnitz-Laboratorium (MLL) in Garching. This measurement was part of a PhD-Thesis [18] and more details on the measurement and analysis can be found in that reference. Two samples of about 100 mg each, with ^{62}Ni enrichments of 95.0% and 97.3% and small copper impurities (< 10 ppm and 0.002%) were irradiated in the quasi-stellar 25 keV neutron spectrum [10] using the FZK Van de Graaff accelerator in combination with a ^7Li target. The conventional approach of measuring the γ -rays from the decay of the daughter nucleus is hindered in this case, because ^{63}Ni is a pure beta emitter. Therefore, the number of activated ^{63}Ni nuclei was determined by means of Accelerator Mass Spectrometry (AMS) at MLL. The tandem accelerator at MLL was used to produce a beam of sample atoms with an energy of 165 MeV. The isotopic background can be reduced down to a negligible level by using a Wien filter and concomitant TOF measurements. The main difficulty in this measurement was connected with the isobaric ^{63}Cu background component. A technique has been developed at MLL in order to suppress this type of background effectively. The latter is based on a Gas Filled dipole Magnet System (GAMS) [19, 20] and a multi differential energy-loss and position sensitive ionization chamber. By applying several “software cuts” in the ΔE values, a sensitivity of 3×10^{-13} was achieved in this particular measurement. This corresponds to a $^{63}\text{Ni}/^{62}\text{Ni}$ ratio, which is two orders of magnitude smaller than the ratios of 5×10^{-11} and 6×10^{-11} measured with the two activated Ni-samples. From the latter two AMS measurements, an average Maxwellian cross section at $kT = 30$ keV of 23.4(4.6) mb was obtained [18].

In summary, there are at present four independent measurements [7, 13, 17, 18], two of them made with the TOF method and the other two via the Activation/AMS technique, which show good agreement with each other, and yield a weighted average Maxwellian cross section value of 26 ± 4 mb.

$^{64}\text{Ni}(n, \gamma)$

An important contribution to the nucleosynthesis of the Fe and Ni isotopes occurs during the shell Carbon burning phase of evolution in massive stars ($M > 8M_{\odot}$). At this stage, peak neutron densities of about 10^{12} n/cm^3 are reached at stellar temperatures of $T = 1\text{--}1.5 \times 10^9 \text{ K}$, which correspond to thermal neutron energies of $kT = 90\text{--}120 \text{ keV}$.

Information about the neutron capture cross section near this higher neutron energy range can be gained by means of a tritium neutron source [21]. By using a tritium target in combination with a proton beam of 1099 keV a neutron energy distribution is obtained, which simulates a Maxwellian energy spectrum at $kT = 52 \text{ keV}$. Combined with a similar measurement at 25 keV [22], information on the energy dependence of the cross section can be obtained, which allows to extrapolate the MACS more reliably to the 90-100 keV energy range.

The measurement of the ^{64}Ni CS at 52 keV was performed at the Physikalisch Technische Bundesanstalt (PTB) in Braunschweig. Using a Van de Graaff, protons were accelerated to an energy of 1099 keV. An air cooled tritium target was used in order to produce the 52 keV quasi-stellar neutron spectrum [21]. Two enriched ^{64}Ni samples 6 and 10 mm in diameter were used. Four single activations of 4 to 6 hours each were carried out. After each neutron irradiation run, the 1482 keV γ -rays from the decay of excited states in ^{65}Cu were measured with a high purity Ge-detector. In this way the number of activated ^{65}Ni nuclei was determined with high selectivity.

FIGURE 1. (Left) Experimental cross section value for each activation made at PTB. The average value is shown together with the statistical and total uncertainties (dashed lines). (Right) MACS result at 50 keV (square) and at 25 keV (triangle) [22]. The evaluated data files are also shown. The latter have been normalized to the experimental value measured here.

The result from this measurement is shown in Fig. 1, together with a similar previous measurement at 25 keV [22]. The MACS obtained from different evaluated data files [23] are also shown in this figure, normalized to the experimental value measured here. It is obvious that the ENDF and JEFF libraries reproduce the CS energy trend perfectly, whereas JENDL and BROND show a completely different behavior.

By combining the two activation measurements at 25 keV [22] and at 52 keV (this work) the correct energy trend of the CS can be inferred. This lends more confidence to the MACS extrapolation to the energy range of 90 keV-100 keV, relevant for stellar nucleosynthesis during shell C-burning.

IRON AND NICKEL (n, γ) MEASUREMENTS AT CERN N_TOF

The n_TOF facility is the CERN time of flight installation for the measurement of neutron reaction rates, like fission and capture. A commissioning of the facility was carried out in 2001, where the key features were accurately characterized [24], like resolution function [25], neutron flux [26], spatial neutron beam profile [27] and backgrounds (activation, in beam γ -rays, prompt and neutron sensitivity) [28]. A substantial amount of beam-time was also employed in the study and experimental validation [29] of the so-called Pulse Height Weighting Technique (PHWT) for the measurement of (n, γ) cross sections with high accuracy employing low-efficiency C₆D₆ detectors. Capture and fission measurements, relevant for both nuclear astrophysics and technology, were performed during 2001, 2002 and 2003. For an overview of those measurements see e.g. Refs. [30, 31] and references therein.

At present an upgrade of the spallation source is being made, which consists basically of *i*) a new cylindrical lead target, and *ii*) a neutron beam moderator made from heavy water or borated water. These two changes on the target will allow for a more efficient cooling of the spallation source, and much lower in-beam γ -ray background levels in the measuring station. The latter type of background arose in previous measurements from neutron captures in the hydrogen of the water moderator and affected mostly the 10-100 keV neutron energy region.

The excellent neutron energy resolution of n_TOF and the low neutron sensitivity of the capture setup [9], together with the improved background conditions, will allow measurements of the (n, γ) cross section of the Fe and Ni isotopes under optimal conditions and with high accuracy in the neutron energy range from 1 eV up to almost 1 MeV.

As it is illustrated in Fig. 2, for many of the resonances at high neutron energy the scattering channel becomes 3 to 5 orders of magnitude larger than the capture channel. Those scattered neutrons can be easily captured in the materials surrounding the sample and the emitted γ -rays represent a prompt background, which is difficult to correct properly in the data analysis. This type of background can be efficiently suppressed at n_TOF, where the capture setup has been specially developed in order to overcome this problem. Indeed, CS corrections of only a few percent have been estimated for those resonances in Fig. 2 with the largest Γ_n/Γ_γ -ratios.

SUMMARY

The present activation measurement [11] at $kT = 25$ keV for ^{60}Fe corresponds to the first experimental determination of the neutron capture cross section for this isotope. Therefore, this result substantially improves the uncertainty on the theoretical prediction of the ^{60}Fe abundance in our Galaxy. However there are still many reactions connected with the ^{60}Fe problem, which need to be measured, like ^{59}Fe (n, γ), $^{22}\text{Ne}(\alpha, n)$, etc.

The activation measurement of ^{62}Ni at $kT = 25$ keV [18] has provided a result of key importance for disentangling a long-standing problem concerning the cross section value of this isotope and therefore, to determine its abundance more reliably. However, the extrapolation of the present result to the higher temperature of C-shell burning remains problematic, because of the resonance contribution to the cross section in that energy

FIGURE 2. Ratio of the neutron width (Γ_n) to capture width (Γ_γ) versus the resonance energy for the iron (top) and the nickel isotopes (bottom).

range. Therefore, it becomes mandatory to perform complementary TOF measurements and/or activation measurements at $kT = 52$ keV via the tritium source.

The measurement of ^{64}Ni at $kT = 52$ keV constitutes an excellent complement of the previous measurement at $kT = 25$ keV [22]. These two independent measurements show consistency with each other, and allow one to extrapolate more reliably the MACS to the energy range of shell C-burning at 90 keV.

Finally, since during the weak s process the reaction flow does not reach equilibrium, the accurate knowledge of the cross sections in the entire energy range (from 1 eV up to 100-500 keV) and of all the involved isotopes is needed in order to predict the stellar abundances reliably. Therefore, complementary TOF measurements with state of the art techniques are planned at n_TOF for the future capture campaign in 2009.

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