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The Role of Fe and Ni for S-process Nucleosynthesis and Innovative Nuclear Technologies

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The accurate measurement of neutron capture cross sections of all Fe and Ni isotopes is important for disentangling the contribution of the s-process and the r-process to the stellar nucleosynthesis of elements in the mass range $60 < A < 120$. At the same time, Fe and Ni are important components of structural materials and improved neutron cross section data is relevant in the design of new nuclear systems. With the aim of obtaining improved capture data on all stable iron and nickel isotopes, a program of measurements has been launched at the CERN Neutron Time of Flight Facility n_TOF.

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I. INTRODUCTION

Neutron capture (followed by β-decay) is the main process in the formation of elements heavier than iron in the universe. The so called slow neutron capture (s) process, which occurs in the interior of stars during the Red Giant phase, is characterized by neutron capture times of about a year, much longer than typical β-decay times. Accordingly, the reaction path of the s process runs along the valley of β-stability and the resulting abundances are directly correlated with the respective capture cross sections. The r-process rapidly builds very neutron-rich nuclei in explosive stellar scenarios (*e.g.*, supernovae), which then decay towards the stability valley. Both processes contribute in about equal amounts to the total heavy element budget although notable differences exist for individual isotopes or particular stars. In fact it has been observed that in Ultra Metal Poor (UMP) stars the total abundance of elements heavier than iron matches closely the scaled solar r-process abundance distribution. Since these UMP stars constitute some of the oldest stars in the universe, this remarkable fact has been interpreted as a signature of a very robust primary r-process, independent of the history of star formation in the Galaxy [1]. Further investigations revealed that the almost perfect match is restricted to elements above Ba, while systematically lower values are observed in UMP stars for elements below Ba. This observation has been used to propose a secondary r-process contributing later in Galactic evolution to the abundances in this mass region [2], which depends, however, on the reliability of the r-process abundances in this mass region.

Solar r-process abundances are determined (except for r-only isotopes) as the difference between total solar abundances and s-process abundances. The latter are obtained from astrophysical calculations where the main nuclear physics input consists of the (n,γ) cross sections of the involved isotopes, averaged over the stellar neutron spectrum. In the mass region below A = 90, the so called "weak" s-process determines the abundances. In the main s-process, occurring during the thermal pulsing phase of Asymptotic Giant Branch (AGB) stars, the isotopic abundances are inversely proportional to the neutron capture cross sections. In the weak s-process, which takes place in more massive stars, the neutron fluence is not sufficient to reach steady flow equilibrium. Therefore cross section uncertainties are affecting also the abundances of the following isotopes on the s-process path [3]. To avoid this propagation effect precise cross section data are required over a large range of isotopes, particularly at and near the Fe/Ni seed.

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The quality of neutron capture cross-section data as measured by the experimental uncertainties for nuclei below A = 120 is, in general, considerably worse than for heavier nuclei [4]. Moreover, there are indications coming both from activation measurements [3,5] and recent time-of-flight measurements [6] that some cross sections in this region are overestimated. It must be noticed that an overestimation of the capture cross section leads to an overestimation of the solar r-residuals.

Iron and nickel are also important constituents of structural materials utilized in nuclear technology, including advanced systems such as Accelerator Driven Systems (ADS) and Generation IV power reactors. Both elements are part of the cladding material for fuel elements and of the neutron reflector, for example. In the context of the ADS studies several steels are considered to build the window separating the proton accelerator and the spallation target. In line with what was mentioned above, relatively large uncertainties are found for capture cross section in data bases for important Fe and Ni isotopes [7], in particular in the higher keV neutron energy range. These uncertainties will have an impact on the assessment of the system parameters in ongoing and future design studies. Another reason for improved capture data comes from the fact that the neutron capture products ^{59}Ni ($t_{1/2} = 7.5 \times 10^4$ y) and ^{63}Ni ($t_{1/2} =$ 100 y) contribute significantly to the long and short term activation of materials. A more precise quantification of the expected activity is required also in the design studies.

In view of the above discussion a systematic review of capture cross section data in the mass region above A $\sim$ 60, starting with Fe and Ni isotopes is of great relevance.

II. MEASUREMENTS

A campaign of measurements to determine the (n,γ) cross sections of all stable Fe and Ni isotopes in the neutron energy region between 0.1 keV and 1 MeV has been started at the n_TOF facility [8]. A large "instantaneous" number of neutrons from spallation reactions are produced by the impact of intense proton bunches from the CERN PS (7×10^{12} protons in a 7 ns wide bunch) with an energy of 20 GeV on a massive lead target. The initial neutron bursts are further moderated in a water volume at the exit towards the evacuated neutron beam pipe, which connects the target with the experimental area at a distance of about 185 m. The long flight path provides high resolution in neutron energy, which allows resolving closely spaced neutron resonances.

During the experiments the neutron flux at the sample position was monitored by counting the number of ^{6}Li(n,t)^{4}He reactions produced in a thin ^{6}Li deposit on

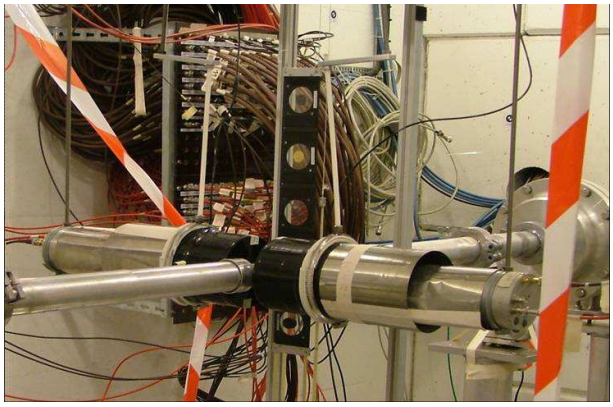

Fig. 1. (Color online) The experimental setup, showing the neutron beam pipes, C_6D_6 detectors and the sample holder. The neutron beam enters from the left.

a Mylar foil with 4 large area Si detectors outside of the neutron beam [9]. The prompt capture γ-rays were detected with two optimized deuterated benzene (C_6D_6) liquid scintillation detectors [10]. Figure 1 shows a view of the experimental setup. The choice of deuterium, the utilization of a thin carbon fibre canning, and the thorough reduction of dead materials in and near the detector resulted in a reduced sensitivity to background from sample scattered neutrons. This type of background had been the cause of undetected problems in previous work, because the probability for neutron scattering may well exceed that for capture by more than a factor of 1000 in many Fe and Ni resonances.

In order to properly account for the γ-ray energy dependence of the detection efficiency, the Pulse Height Weighting Technique (PHWT) is used. The technique can be applied because the probability for detecting more than one γ-ray of a capture γ-ray cascade in the C_6D_6 detectors is sufficiently small. The PHWT requires the precise knowledge of the detector response as a function of γ energy. The latter is obtained from very detailed Monte Carlo simulations using Geant4 [11] with a complete description of the experimental setup. From this information a counting weight as a function of deposited energy can be calculated. It has been shown that an accuracy of 2% can be achieved in this way [12].

The metallic samples (⌀20 mm, ∼2 g) are placed in a narrow gap between two thin KAPTON windows in the beam pipes. The samples are glued onto a KAPTON foil stretched over a carbon fibre frame which is not hit by the neutron beam, reducing unwanted scattering backgrounds. It also enables the easy exchange of samples for calibration and background measurements. In addition to the highly enriched Fe and Ni samples listed in Table 1, samples of Au, Pb, and C served for the determination of the neutron flux, of the background related to in-beam γ-rays, and of the background due to sample scattered neutron, respectively.

The detectors are placed 9 cm upstream of the sample position in order to reduce the influence of in-beam γ-rays, which are scattered in the samples preferentially in forward direction. This position also minimizes the angular distribution effect of the primary capture γ-rays. Neutron backgrounds are quantified using black resonances in W and Al absorption filters in the beam. All measurements are normalized to the saturated 4.9 eV resonance in ^{197}Au in order to obtain absolute yields. This method requires an accurate knowledge of the energy dependence of the flux as a function of neutron energy, which has been obtained from dedicated measurements using several monitors and Monte Carlo simulations with FLUKA [13].

Table 1. Degree of enrichment of the Fe and Ni samples used in the measurements.

Isotope	Enrichment (%)
^{54}Fe	99.84
^{56}Fe	99.93
^{57}Fe	96.06
^{58}Ni	99.5
^{60}Ni	99.31
^{62}Ni	97.95

During the 2009 campaign C_6D_6 capture data was taken for ^{56}Fe and ^{62}Ni samples. The signals from both detectors are continuously sampled using fast digitizers (500 MS/s) for a time length equivalent to 16 ms starting with the impact of the proton pulse on the spallation target. The detector noise was suppressed by the data acquisition software [14] and the waveform of the remaining true signals was stored for later processing by a pulse fitting procedure, which provided precise values for the amplitude and time of flight of each signal, even in conditions of moderate pileup. From the energy calibrated amplitudes the weight of each count is calculated using the sample specific weighting functions described before. Examples for time of flight spectra of weighted counts for both samples are shown in Fig. 2. These spectra represent a preliminary stage of data reduction, while the comprehensive analysis of all data sets is still in progress.

III. FUTURE MEASUREMENTS

In 2008 a new spallation target was build and installed at the n_TOF facility. One of the improvements in the new design is the provision of a liquid moderation volume at the neutron exit face of the target, independent of the cooling water circuit. The purpose of this modification is to use borated water as moderator in order to reduce the number of in-beam γ-rays arriving to the sample. In the previous design these γ-rays were produced mainly by capture of the neutrons on ^{1}H (E_γ = 2.2 MeV) in the water moderator/cooling. After scattering in the sample they constitute the major source of background in the region E_n = 0.5 – 500 keV. Addition of ^{10}B to the water moderator volume

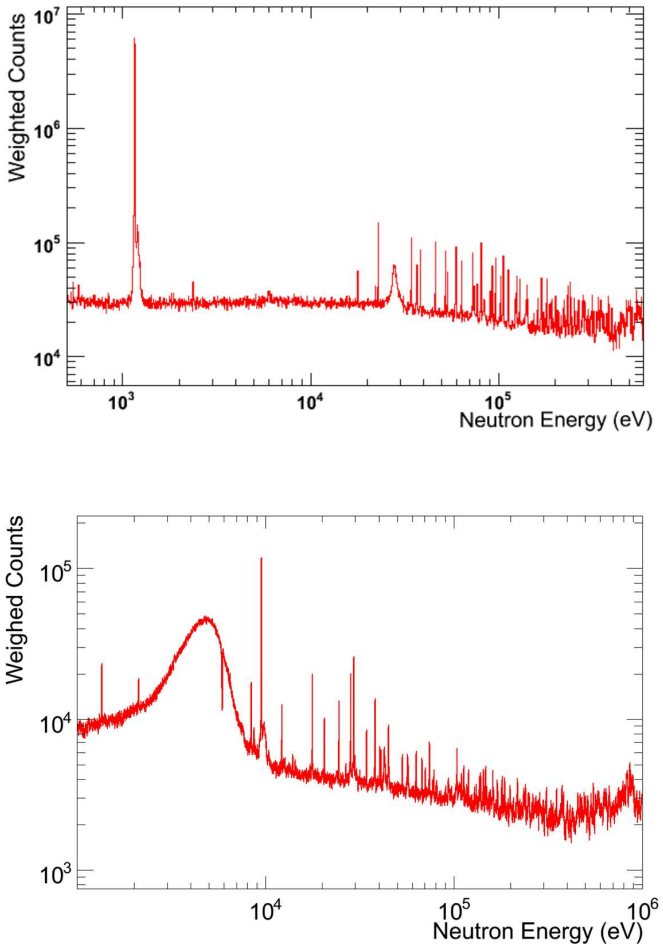

Fig. 2. (Color online) Weighted γ-spectrum obtained for ^{56}Fe (upper panel) and ^{62}Ni (lower panel).

diminishes drastically the capture on hydrogen while the energy of the γ-ray ($E_\gamma = 0.48$ MeV) from ^{10}B(n,γ) is low enough to escape detection in the C_6D_6 after scattering. Over one order of magnitude reduction of this type of background is expected according to Monte Carlo simulations. Starting with 2010 this modification will be implemented and the quality of the new neutron capture measurements in Fe and Ni isotopes will be greatly enhanced.

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