

## Measurement of Capture Cross Sections of $^{90,91,92,94,96}\text{Zr}$ Isotopes at n\_TOF

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**Abstract.** The neutron capture cross sections for Zr isotopes have important implications in nuclear technologies as well in the field of nuclear astrophysics. The innovative features of the n\_TOF CERN facility allowed for an accurate determination of the capture cross sections for these isotopes. The preliminary results of the measurements show capture-resonance strengths systematically smaller than in previously available data.

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## INTRODUCTION

Since the baryonic freeze-out in the Big Bang, stars have been identified as the sites for nucleosynthesis. Beyond Iron, the only method of production of isotopes is via neutron capture, through two dominant processes, which differ by the time scale on which they occur: the *r* (rapid) process, related to extremely hot ( $T > 10^9$  K), neutron rich ( $n_n \gg 10^{20} \text{ cm}^{-3}$ ) environments; the *s* (slow) process, which operates at lower temperatures ( $T \approx 10^8$  K) and densities ( $n_n \approx 10^8 \text{ cm}^{-3}$ ) [1]. Realistic stellar models have been developed, which can account for the actual stellar conditions under which the *s* process is taking place [2]. However, the reliability of the stellar *s* process models can only be tested by new developments in neutron-capture cross-section measurements, with uncertainties of only a few %. This improved accuracy is a pre-requisite for the application in Nuclear Astrophysics. Moreover, recent developments have emphasized the importance of neutron-capture nucleosynthesis for probing the deep interior of Red Giant stars and for following the enrichment of heavy elements during galactic evolution. These data, required in the energy range from 0.3 to 300 keV, are largely missing, particularly in the mass region  $A \leq 100$ , where cross sections are small and dominated by single resonances [3].

The Zr isotopes represent an important example to illustrate this situation.  $^{90}\text{Zr}$ ,  $^{91}\text{Zr}$ ,  $^{92}\text{Zr}$ , and  $^{94}\text{Zr}$  are predominantly of *s*-process origin. However, the small cross section of the neutron magic  $^{90}\text{Zr}$  isotope acts as a bottleneck for the reaction flow towards heavier elements. Moreover, even if the heaviest isotope,  $^{96}\text{Zr}$ , is traditionally considered to be an *r*-only isotope, a non-negligible *s*-process admixture must be considered in their formation [4].

Apart from the impact on problems of Nuclear Astrophysics, the  $(n, \gamma)$  cross sections of the Zr isotopes are of interest for technological reasons also. Zr constitutes, in fact, an important component in alloys used as structural material and in fuel assemblies in nuclear reactors. In particular the NEA "High Priority Nuclear Data Request List" asks for a fine  $(n, \gamma)$  cross section determination from thermal to 1-MeV neutron energy [5].

Existing data in the range between 0.1 and 500 keV exhibit uncertainties larger than 10% and discrepancies up to a factor 2 between different measurements.

## EXPERIMENTAL SET-UP

The measurements were performed over the energy range between 1 eV to 1 MeV at the spallation neutron facility n\_TOF at CERN [6]. The low repetition frequency of the proton beam driver, the extremely high instantaneous neutron flux, the low background conditions in the experimental area, together with the low neutron sensitivity of the capture detector make this facility unique for neutron-induced reaction cross-section measurements. The neutrons are generated by spallation reactions induced by a pulsed beam of 20-GeV protons on a massive Pb target. Neutrons are slowed down in the lead and moderated in a surrounding 5-cm-thick layer of cooling water. An evacuated neutron flight path with collimators leads to the measuring station at a distance of 185 m from the spallation target. Background due to fast charged particles is suppressed by means of a sweeping magnet, heavy concrete walls, and a 3.5-m-thick iron shielding [7].

The experimental set-up consists of two  $\text{C}_6\text{D}_6$  (deuterated benzene) detectors with minimized neutron sensitivity [8], placed perpendicular to the neutron beam at a distance of about 3 cm from the beam axis. The background due to in-beam  $\gamma$ -rays was reduced by placing the detectors 9.2 cm upstream of the sample position. The light output of the detectors was calibrated by means of  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ , and Pu/C  $\gamma$ -ray sources. The calibrated neutron time of flight was used to determine the neutron energy. The detector signals were recorded with fast flash ADC using the standard n\_TOF data acquisition system [9].

All zirconium samples were prepared from oxide powder, which was pressed to pellets 22 mm in diameter and encapsulated in a thin aluminum can. Additional Au and Pb samples of the same diameter were used for flux and background measurements.

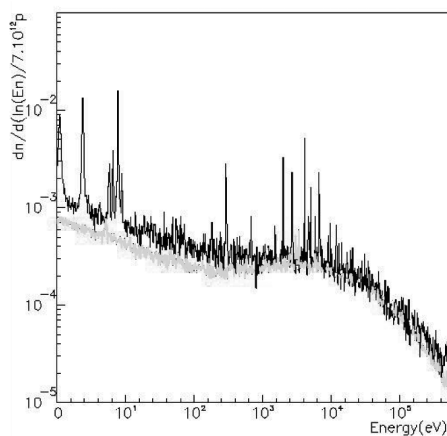
The relative neutron flux was measured 1.5 m upstream of the capture samples with a low mass flux monitor consisting of a thin layer of  $^6\text{Li}$  surrounded by four silicon detectors outside the neutron beam [10].

## DATA ANALYSIS

The quantity determined in a neutron capture experiment is the capture yield, i.e., the fraction of incident neutrons undergoing  $(n, \gamma)$  interaction on a defined sample. The capture yield is directly linked to the capture and total cross sections. The data analysis was then based on an accurate measurement of the capture yield.

One has to consider that due to the small solid angle coverage and the low intrinsic efficiency of the  $C_6D_6$  detectors, which results in an overall efficiency of  $\approx 10\%$ , only one  $\gamma$ -ray per event is detected from the de-excitation cascade following neutron capture. For an accurate cross-section determination, the efficiency of the set-up has to be made independently on the detail of the de-excitation cascade, in particular of the  $\gamma$ -ray multiplicity. To this end, a pulse height weighting function (PHWF) was used [11]. It consists of suitably modifying via software the detector response so that the efficiency  $\varepsilon_\gamma$  is proportional to the photon energy  $E_\gamma$ . Under these conditions the efficiency for detecting a cascade becomes proportional to the known cascade energy  $E_c$  and independent of the cascade path. The proportionality of the efficiency with the  $\gamma$ -ray energy is achieved by modifying the detector energy response distribution  $R(E)$  with a pulse-height (deposited energy) dependent weighting function ( $W(E)$ ), applied to the recorded spectrum. A check on the reliability of the weighting function technique has been performed by measuring isotopes with well-known capture cross sections ( $^{nat}Fe$ ,  $^{nat}Ag$ ,  $^{197}Au$ ) [12].

As a further step, ambient and sample related backgrounds were subtracted by means of the spectra measured with an empty Al can (the Zr samples were lodged inside this support) and with the Pb sample. For the five measured isotopes, only the 90, 92, and 96 are completely analyzed; 94 is well in progress and 91 has still to be analyzed. Figure 1 shows the capture yield for  $^{92}Zr$  with the overall background.

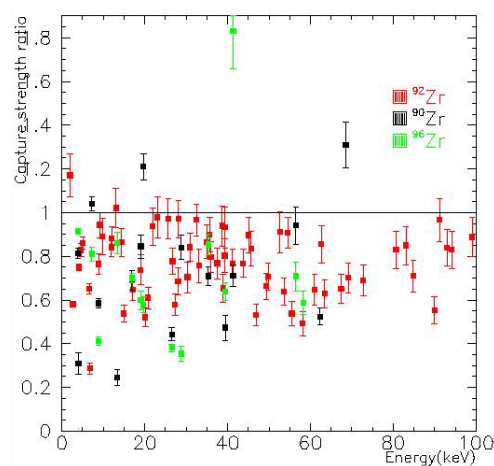


**FIGURE 1.** Capture yield of the  $^{92}Zr(n, \gamma)^{93}Zr$  reaction (black) and of the overall background (gray).

The absolute normalization of the capture yields has been made via the spectrum measured with the Au sample. In particular, the normalization factor was determined with accuracy better than 1.5% by analyzing the saturated standard 4.9-eV resonance.

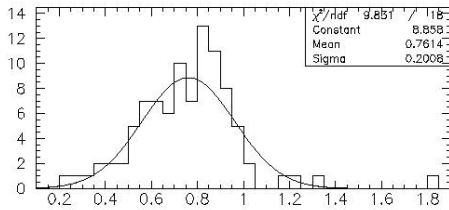
Once extracted, the experimental capture yields and the analysis of the resonances were done by means of the multi-level analysis code SAMMY [13].

The Energy  $E$ , the  $\Gamma_\gamma$  and  $\Gamma_n$  widths are extracted from the fit on the  $n\_TOF$  data given by SAMMY. The neutron capture strengths  $g\Gamma_\gamma\Gamma_n/\Gamma_{Tot}$ , where  $g$  is the spin statistical factor and  $\Gamma_{Tot}$  the total width are therefore calculated [14]. Figure 2 shows the comparison between the neutron capture strengths for the  $^{90}Zr$ ,  $^{92}Zr$ , and  $^{96}Zr$  ( $n, \gamma$ ) measurements with those determined in previous experiments [14].



**FIGURE 2.** Ratio between the  $n\_TOF$  capture measured strengths and previously observed values [14].

On this plot, the errors are proceeding from three independent sources: flux determination (1.5%), pulse height weighting function (2%), and SAMMY fit, which varies with the resonance and increases with the energy (5-10%). Except for a very few resonances, the  $n\_TOF$  results are systematically lower, for all the measured isotopes. Figure 3 shows the distribution of the ratio between the  $n\_TOF$  and the available previously measured data concerning the neutron capture strengths.



**FIGURE 3.** Distribution of the ratio between the n\_TOF capture measured strengths and previous observed values, for different Zr isotopes shown in Fig. 2 [14].

## CONCLUSIONS

The capture cross sections of  $^{90,91,92,94,96}\text{Zr}$  isotopes have been measured at the CERN n\_TOF facility in the energy range from 1 eV to 1 MeV. The innovative features of this facility allow performance of high-accuracy measurements. The obtained results concerning the capture strengths are systematically lower than those presented in previous experiment; this fact could be related to the lower neutron sensitivity of the experimental set-up used at n\_TOF.

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