MEASUREMENTS OF THE ^{90,91,92,93,94,96}Zr (n, γ) CROSS-SECTIONS AT n_TOF

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Neutron capture cross sections for zirconium isotopes have important implications in the field of nuclear astrophysics as well as in the nuclear technology. In particular they play a key role for the determination of the neutron density in the He burning zone of the Red Giant stars. Zirconium is also largely used as structural materials of traditional and advanced nuclear reactors. Neutron capture cross sections of the ^{90,91,92,93,94,96}Zr have been measured at the spallation neutron facility n_TOF at CERN. The innovative features of the neutron beam, in particular its high instantaneous flux, the high energy resolution and low background, together with improvements of the neutron sensitivity of the capture detectors make this facility unique for neutron-induced reaction cross section measurements with much improved accuracy.

1. Introduction

The measure of the neutron capture cross section of ^{90,91,92,93,94,96}Zr has relevance in nuclear astrophysics, since the Zr belongs to the first *s*-process peak in the solar abundances distribution around the neutron magic number N = 50. Accurate *s*-process analyses have recently attracted great interest over the last decade, thanks to the progresses in astronomical observations and in stellar interpretative models. The understanding of the *s*-process has advanced from a phenomenological description of the abundance distribution in the solar system towards a comprehensive picture, which includes the overall aspects of stellar and galactic evolution [1]. However the success of the stellar *s*-process models could only be achieved by significant improvements in the neutron capture cross section data, which reached uncertainties of only a few %. This accuracy turned out to be a prerequisite for this application of nuclear data in nuclear astrophysics. At present many cross section data are still missing, particularly in the mass region A≤100 as well as for neutron magic nuclei where cross sections are small and dominated by isolated resonances [2]. The ^{90,91,92,93,94}Zr are characterized by a low neutron capture cross section and are predominately of *s*-process origin. The ^{96}Zr is considered to be an *r*-only isotope with a small *s*-process admixture [1]. Branching in the reaction path, which occur at unstable isotopes, where neutron capture competes with β-decay, are a unique tool for constraining the physical conditions in the He burning zones near the stellar core. An important branching occurs at ^{95}Zr.

The (n, γ) cross section of stable Zr isotopes are also of interest for technological reasons, since zirconium is widely present in structural materials of nuclear reactors. Moreover, the unstable isotope ^{93}Zr is one of the major long-lived fission products.

Existing (n, γ) cross sections of Zr isotopes in the relevant energy range from 0.1 to 500 keV exhibit uncertainties larger than 10 % and discrepancies up to a factor 2 between different measurements have been observed. The necessary improvement of these data was achieved using the unique features of the n_TOF facility, which combines excellent resolution, high instantaneous neutron flux and low background [3].

2. Experimental set-up

The measurements were performed with the n_TOF pulsed neutron beam. 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 the surrounding 5 cm thick layer of cooling water. An evacuated neutron flight path with collimators at 135 and 175 m leads to the measuring station at a distance of 185 m from the spallation target. Background due to the fast charged particles is suppressed by a sweeping magnet, heavy concrete walls and a 3.5 m thick iron shielding.

The experimental set-up consists of two C_6D_6 detectors with minimized neutron sensitivity [4], placed perpendicular to the neutron beam at a distance of about 3 cm from the beam axis. The background due to in beam γ-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}Cs, ^{60}Co and Pu/C γ-ray sources.

The calibrated neutron time of flight was used to determine the neutron energy.

The detectors signals were recorded with fast flash ADC using the standard n_TOF data acquisition system [5].

Enriched Zr samples were prepared from oxide powder, pressed to pellets 22 mm in diameter, encapsulated in a thin aluminum can. Admixtures of Hf, Sn, Na, Mg, Al were also present in the samples. Even thought the presence of contaminants is very low, their influence can not be neglected. C, Au, Pb samples were used for flux and background measurements. The relevant sample characteristics are given in the Table.

Sample characteristics. All samples were in the form of ZrO_2

Sample	Isotopic composition, %						Mass (g)
	^{90}Zr	^{91}Zr	^{92}Zr	^{93}Zr	^{94}Zr	^{96}Zr	
^{90}Zr	97.7	0.87	0.60	-	0.67	0.16	2.717
^{91}Zr	5.43	89.9	2.68	-	1.75	0.24	1.404
^{92}Zr	4.65	1.62	91.4	-	2.03	0.30	1.349
^{93}Zr	1.5	19.	20.	20.	20.	19.	4.88
^{94}Zr	4.05	1.18	1.93	-	91.8	1.04	2.015
^{96}Zr	19.41	5.21	8.2	-	8.68	58.5	3.398

3. Data analysis

The quantity to be determined in a neutron capture experiment is the yield, which is defined as the fraction of incident neutrons undergoing (n, γ) reactions in the sample. The capture yield is directly linked to the capture and the total cross sections. Since the capture detectors have low efficiency and cover a restricted solid angle, the overall efficiency for the detection of capture events was ≈ 20 %. The absolute normalization of the capture yields has then been made referring to the well-known 4.9 eV saturated resonance of ^{197}Au.

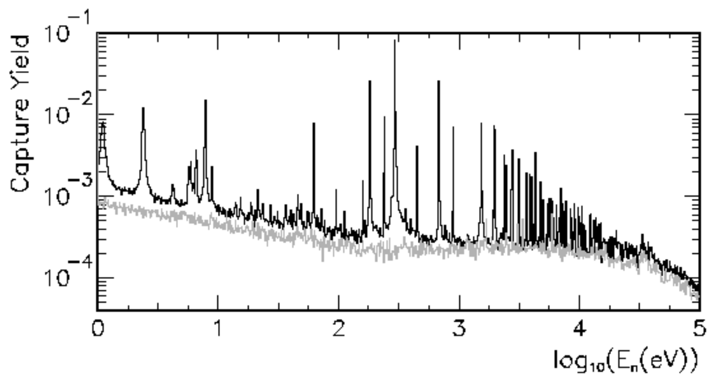

Fig. 1. Capture yield (black) and overall background (grey) for the ^{91}Zr isotope; most of the resonances up to 150 eV are due to Hf and Sn contamination in the sample.

The efficiency correction via the Pulse Height Weighting Technique (PHWT) [6] is based on the application of a pulse height dependent weight for each single registered γ-ray to ensure that the detection efficiency is independent on the multiplicity distribution of the capture cascade. The respective weighting functions are very sensitive to the experimental set-up including the investigated sample. These functions were derived by detailed Monte Carlo simulations.

Ambient and sample related backgrounds were subtracted by means of the spectra measured with an empty Al can and with the Pb sample.

Fig. 1 shows the capture yield for the ^{91}Zr compared to the total background.

Observed resonances were analyzed in the Reich-Moore approximation with the R-matrix code SAMMY [7]. For each resonance three parameters have been extracted: the resonance energy E_R, the capture width Γ_γ and the neutron width Γ_n. The correction for Doppler broadening of resonance widths due to the thermal motion, for the energy resolution of the neutron beam, for isotopic and chemical sample impurities, and for self-shielding and neutron multiple scattering, are taken into account inside the code. The effect of potential scattering was theoretically calculated [8].

The neutron capture kernels obtained from the resonance parameters measured at n_TOF were compared to other experimental data. Previous experiments are in general old (mid-1970) and provide scarce information on resonance parameters. The capture kernels measured at n_TOF are in general (but the ^{94}Zr) 10 - 20 % smaller than previous reported [8, 9]. This can be explained in terms of the systematic uncertainties that are significantly reduced by the improved n_TOF set-up. As an example one has to note that the use of fast flash ADCs provides an efficient way for n/γ discrimination. Other developments in favor of the present data are related to the very low neutron sensitivity of the n_TOF set-up and of the use of the well tested R-matrix code [7]. In this context, it should be noted that the low neutron induced background, obtained with the optimized experimental set-up and with the extremely small duty factor of the n_TOF facility, represents a strong improvements in measurements of small capture cross sections as in the case of Zr isotopes.

4. Results

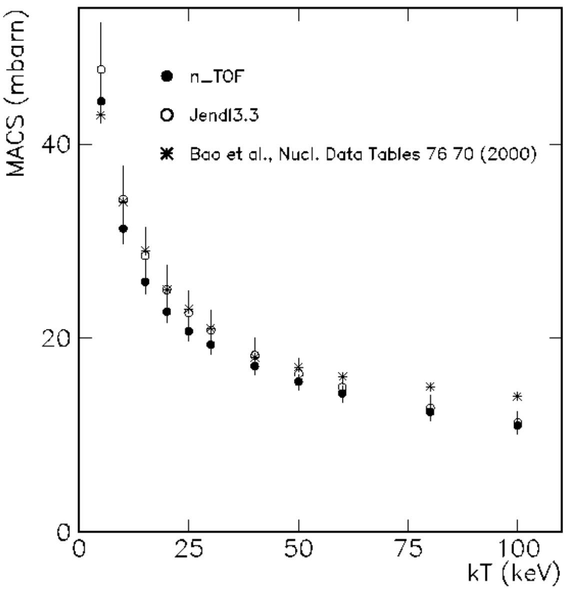

Fig. 2. Comparison of MACS calculated with present ^{90}Zr experimental results (full circles), with values obtained from Ref. [11] (open circles) and with the compilation of Ref. [2] (asterisks).

Zirconium plays an important role in the determination of the *s*-process abundances of heavy elements. The *s*-process occurs predominantly in thermally pulsing low mass Asymptotic Giant Branch (AGB) stars, where neutrons are produced by a (γ, n) reactions on ^{13}C and ^{22}Ne. Methods of converting experimental cross section data in stellar properties were suggested by Macklyn and Gibbons [10]. Among them, the definition of the Maxwellian averaged capture cross section (MACS) is strictly connected to the stellar neutron capture rate [2]. The MACS can then give information on the nucleosynthesis probability along the *s*-path. The calculation of MACS at typical *s*-process temperatures has to be carried out by folding the capture cross section with the thermalized stellar spectra over a sufficiently wide neutron energy range, starting at about 100 eV and extending to about 500 keV at the highest temperatures reached during carbon shell burning in massive stars.

The MACS calculated with the present experimental data are in general lower than that listed in evaluated libraries [11]; this fact is a direct consequence of the weaker capture kernel values obtained in the present measurements. Fig. 2 illustrates the ^{90}Zr example, where the MACS of these experiments are up to 10% lower than those obtained with previous available data.

5. Conclusions

The neutron capture cross sections of ^{90,91,92,93,94,96}Zr have been measured at the CERN n_TOF facility. The optimized experimental conditions at n_TOF allowed to significantly improve the accuracy in the results, allowing for better face the *s*-process nucleosynthesis studies. The capture kernels obtained in this work are for all samples but the ^{94}Zr weaker than what reported in previous experiments. Consequently the MACS calculated with the present experimental data are in general lower than the values extracted by evaluation studies. This result can be explained in terms of the top performances of the facility, experimental set-up data acquisition and more accurate data analysis tools that allow a reduction of systematic uncertainties.

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REFERENCES

1. *Käppeler F.* The origin of the heavy elements: the s-process // Prog. Nucl. Part. Phys. - 1999. - Vol. 43. - P. 419 - 483.
2. *Bao Z.Y. et al.* Neutron cross sections for nucleosynthesis studies // Atomic Nuclear Data Tables - 2000. - Vol. 76. - P. 70 - 154.

3. *Borcea C. et al.* Results from the commissioning of the n_TOF spallation neutron source at CERN // Nucl. Instr. Meth. - 2003. - Vol. A 513. - P. 524 - 537.
4. *Plag R. et al.* An optimized C_6D_6 detector for studies of resonance-dominated (n, γ) cross-sections // Nucl. Instr. Meth. - 2003. - Vol. A 496. - P. 425 - 436.
5. *Plag R. et al.* The data acquisition system of the neutron time-of-flight facility n_TOF at CERN // Nucl. Instr. Meth. - 2005. - Vol. A 538. - P. 692 - 702.
6. *Abbondanno U. et al*. New experimental validation of the pulse height weighting technique for capture cross-section measurements // Nucl. Instr. Meth. - 2004. - Vol. A 521. - P. 454 - 467.
7. *Larson N.M*. Updated users' guide for SAMMY: Multi-level R-matrix fits to neutron data using Bayes equations, SAMMY computer code // Report ORNL/TM-9179/R7, Oak Ridge National Laboratory.
8. *Mughabghab S.F*. Atlas of neutron resonances, resonance parameters and thermal cross sections Z = 1 - 100. - Amsterdam: Elsevier Science, 2006.
9. *Tagliente G. et al*. Neutron capture cross section of ^{90}Zr: bottleneck in the s-process reaction flow // Phys. Rev. C. - 2008. - Vol. 77. - P. 035802.
10. *Macklyn R*., *Gibbons J*., Capture cross-section studies for 30 - 220 keV neutrons using a new technique // Phys Rev. - 1967. - Vol. 159. - P. 1007 - 1012.
11. *For results* compiled in evaluation nuclear data libraries see for example at www-nds.iaea.org