Light Charged-Particle Production Activation Cross Sections of Zr Isotopes from 14 to 20 MeV

V. Semkova¹,², A. J. M. Plompen¹, and D. L. Smith³)

¹) European Commission, Joint Research Center, Institute for Reference Materials and Measurements, B-2440 Geel, Belgium
²) Institute for Nuclear Research and Nuclear Energy, 1784 Sofia, Bulgaria
³) Nuclear Engineering Division, Argonne National Laboratory, Argonne, IL 60439, USA

Abstract. Results of new cross-section measurements are presented for the following neutron-induced reactions: $^{90}\text{Zr}(n,\alpha)^{87m}\text{Sr}$, $^{90}\text{Zr}(n,p)^{90m}\text{Y}$, $^{91}\text{Zr}(n,p)^{91m}\text{Y}$, $^{91}\text{Zr}(n,x)^{90m}\text{Y}$, $^{92}\text{Zr}(n,p)^{92}\text{Y}$, $^{92}\text{Zr}(n,x)^{91m}\text{Y}$, $^{94}\text{Zr}(n,\alpha)^{94}\text{Sr}$, and $^{94}\text{Zr}(n,p)^{94}\text{Y}$ in the energy range from 14 to 21 MeV. Use was made of the activation technique in combination with high-resolution $\gamma$-ray spectrometry. The irradiations were carried out at the 7-MV Van de Graaff accelerator at IRMM, Geel. Quasi-monoenergetic neutrons were produced via the $^3\text{H}(d,n)^4\text{He}$ reaction at 1, 2, 3, and 4 MeV incident deuteron energy. Both natural and samples enriched in $^{90}\text{Zr}$, $^{91}\text{Zr}$, and $^{92}\text{Zr}$ were used to enhance the reaction yield or to facilitate correction for interfering reactions leading to the same product. The measured results are compared with work by other authors, TALYS-0.57 and EMPIRE-II model calculations, and current evaluated data files. Cross sections for all of the investigated reactions have been measured for the first time above 16 MeV.

INTRODUCTION

Accurate measurements of neutron cross sections are needed for design calculations of nuclear technologies that involve free neutrons, and for safety and economy analysis of nuclear energy production. Zirconium is present in most innovative concepts and despite its use in current reactors, requirements for new cross-section data for Zr are included in the request lists for future reactor systems and Accelerator Driven Systems (ADS) [1-3]. The neutron spectrum in important current developments is extended above the traditional limits for light water reactors. Although the energy range of interest for ADS extends up to a few GeV, neutrons with energy below 20 MeV play a considerable role with regard to hydrogen and helium gas production. The hydrogen and helium production data are important for prediction of the evolution of radiation damage, which causes degradation of mechanical properties of irradiated materials.

Studies of excitation functions of fast neutron-induced reactions are also of considerable interest in testing nuclear models. The measurements of reaction cross sections on a series of isotopes of a particular target element are important for development of nuclear reaction model parameterizations in a systematic way and for improvement of reliability of the model predictions.

The production of quasi-monoenergetic fast neutrons with energies up to 20 MeV requires the use of few-neutron-emission charged particle reactions at different incident energies. Usually neutron sources cover the considered energy range only partly. This is reflected in the available experimental database, which is most complete around 14 MeV, while the cross section data for 15–20 MeV incident neutron energy are scarce and very often inconsistent. Although the $(n,xp)$ and $(n,\alpha)$ reaction cross sections on Zr isotopes have been measured by several experimental groups [4], no data have been reported above 16.6 MeV. Here, new measurements are presented for the $^{90}\text{Zr}(n,\alpha)^{87m}\text{Sr}$, $^{90}\text{Zr}(n,p)^{90m}\text{Y}$, $^{91}\text{Zr}(n,p)^{91m}\text{Y}$, $^{91}\text{Zr}(n,np+pn+d)^{90m}\text{Y}$, $^{92}\text{Zr}(np+pn+d)^{91m}\text{Y}$, $^{92}\text{Zr}(n,p)^{92}\text{Y}$, $^{94}\text{Zr}(\alpha)^{94}\text{Sr}$, and $^{94}\text{Zr}(n,p)^{94}\text{Y}$ reaction cross sections between 14.8 and 20.6 MeV. The present results are compared with the evaluated nuclear data files and also with TALYS-0.57 and EMPIRE-II model calculations.
MEASUREMENTS OF CROSS SECTIONS FOR THE REACTION Zr(n,α)Y

By A. J. M. de Graaff and H. G. van der Graaf

EXPERIMENTAL METHOD

Measured cross sections were determined by activation technique in combination with high-resolution γ-spectrometry. A detailed description of the measurement procedure has been given earlier [5].

The irradiations were carried out at the 7-MV Van de Graaff accelerator at IRMM, Geel. Quasi monenergetic neutrons with energies between 14.8 and 20.5 MeV were produced via the \( ^7\text{He}(d,n)^3\text{He} \) reaction (Q = 17.59 MeV) using a solid-state Ti/T target (2 mg/cm\(^2\) thick) on a silver backing (0.4 mm thick) at incident deuteron energies of 1, 2, 3, and 4 MeV. The samples, each sandwiched between monitor foils, were placed at angles between 0° and 75° relative to the incident deuteron beam and at a distance of about 4 cm between the center of the target and the front of the sample stack.

High-purity (99.8%) metallic zirconium sample foils, 0.12 mm thick, supplied by Goodfellow Metals, Cambridge, England, were punched into small disks 1.3 cm in diameter. A set of enriched ZrO\(_2\) sample materials in powder form was borrowed from the Japanese Atomic Energy Research Institute, Tokai-mura, Japan. Individual samples were formed by wrapping powder in small square paper envelopes of 1 cm\(^2\) size. The isotopic composition of the samples that were used is given in Table 1. Use of samples with different isotopic compositions allowed determination of activities produced by (n,p) and (n,p+pn+d) reactions on the neighbor Zr isotopes leading to the production of the same radionuclide.

TABLE 1. Isotopic abundances a (%) of the used natural and enriched Zr samples.

<table>
<thead>
<tr>
<th>Element</th>
<th>(^{90}\text{ZrO}_2)</th>
<th>(^{91}\text{ZrO}_2)</th>
<th>(^{92}\text{ZrO}_2)</th>
<th>(^{93}\text{ZrO}_2)</th>
<th>(^{94}\text{ZrO}_2)</th>
<th>(^{95}\text{ZrO}_2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>90</td>
<td>99.36±0.1</td>
<td>3.24±0.01</td>
<td>2.38±0.01</td>
<td>0.54±0.01</td>
<td>0.36±0.01</td>
<td>0.12±0.01</td>
</tr>
<tr>
<td>91</td>
<td>0.30±0.01</td>
<td>94.69±0.1</td>
<td>1.08±0.01</td>
<td>11.25±0.5</td>
<td>1.12±0.01</td>
<td>0.12±0.01</td>
</tr>
<tr>
<td>92</td>
<td>0.17±0.01</td>
<td>1.63±0.01</td>
<td>95.36±0.1</td>
<td>17.15±0.8</td>
<td>9.63±0.1</td>
<td>1.54±0.01</td>
</tr>
<tr>
<td>93</td>
<td>0.12±0.01</td>
<td>0.46±0.01</td>
<td>1.06±0.01</td>
<td>17.38±0.8</td>
<td>17.38±0.8</td>
<td>20.5±0.5</td>
</tr>
</tbody>
</table>

Given by supplier.

The mean neutron energies and resolutions at each sample for the primary \(^7\text{He}(d,n)^3\text{He}\) neutrons were calculated both by the program code EnergySet, which is based on the cross sections of DROSG-2000 of IAEA, version 2.1 and the stopping powers of Ziegler, and by the Monte Carlo code TARGET.

Above about 2 MeV deuteron energy, the DT neutron spectrum from solid tritium targets shows a significant component of low-energy secondary neutrons. The neutron flux density spectra were determined using neutron-spectrum information obtained by the time-of-flight method and the spectral index method that involves various monitor reactions with distinct energy thresholds. The following standard reaction cross sections were used for the unfolding: \(^{115}\text{In}(n,n)^{115m}\text{In}\), \(^{58}\text{Ni}(n,p)^{58}\text{Co}\), \(^{27}\text{Al}(n,p)^{27}\text{Mg}\), \(^{27}\text{Al}(n,α)^{27}\text{Na}\), \(^{56}\text{Fe}(n,p)^{56}\text{Mn}\), and \(^{61}\text{Nb}(n,2n)^{62}\text{mNb}\). Corrections for low-energy neutrons were significant in the case of \(^{90}\text{Zr}(n,α)^{87}\text{mSr}\), \(^{90}\text{Zr}(n,p)^{90}\text{mY}\), and \(^{91}\text{Zr}(n,p)^{91}\text{mY}\) reaction cross-section measurements. The correction factor was calculated as the ratio of the reaction rates produced by the neutrons below the cutoff energy to those produced by the entire neutron spectrum. Estimates for the needed excitation functions were obtained through fit to raw experimental data.

The neutron fluence rate was determined by the \(^{27}\text{Al}(n,α)^{24}\text{Na\text{ENDF/B-VI.6}}\) standard cross section.

The radioactivity of the samples was measured by γ-ray spectrometry. Two HPGe detectors with 24% and 100% relative efficiency were used. The photopeaks and total efficiencies were determined using standard sources. The measured calibration points were fitted with an analytical function [6]. The values for the decay constants and for the γ-ray branching factors used in data analysis are included in Table 2. Corrections were applied for γ-ray self-attenuation, measurement geometry, and coincidence summing.

TABLE 2. Decay data of measured reaction products [10].

<table>
<thead>
<tr>
<th>Reaction Product</th>
<th>Half-Life</th>
<th>γ-ray Energy (keV)</th>
<th>γ-ray Branch (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{87}\text{mSr})</td>
<td>2.815 h/2</td>
<td>388.5±3</td>
<td>82.4±3</td>
</tr>
<tr>
<td>(^{91}\text{Sr})</td>
<td>9.63 h</td>
<td>1024.3±1</td>
<td>33.5±11</td>
</tr>
<tr>
<td>(^{87}\text{mY})</td>
<td>3.19 h</td>
<td>202.5±1</td>
<td>97.3±4</td>
</tr>
<tr>
<td>(^{91}\text{mY})</td>
<td>49.71 m</td>
<td>555.7±5</td>
<td>90.7±4</td>
</tr>
<tr>
<td>(^{92}\text{Y})</td>
<td>3.54 h</td>
<td>934.4±7</td>
<td>13.9±15</td>
</tr>
<tr>
<td>(^{94}\text{Y})</td>
<td>18.7 m</td>
<td>918.7±5</td>
<td>56.3±5</td>
</tr>
</tbody>
</table>

The uncertainties of the measured cross sections were obtained by combining individual uncertainties in quadrature according to the law of error propagation. The dominant contributions to the data uncertainties are from the counting statistics (\(^{90}\text{Zr}(n,α)^{87}\text{mSr}\): 2-4%, \(^{90}\text{Zr}(n,p)^{90}\text{mY}\): 1-2%, \(^{90}\text{Zr}(n,x)^{90}\text{mY}\): 1.5-3%, \(^{92}\text{Zr}(n,α)^{92}\text{mY}\): 2-5%, \(^{92}\text{Zr}(n,p)^{92}\text{mY}\): 5%, \(^{94}\text{Zr}(n,α)^{94}\text{mY}\): 2-3%), the detector efficiency: 2-3%, low energy correction: 0-12%, reference monitor cross section: 0.5-5%, and coincidence summing correction: 0-7%.
RESULTS AND DISCUSSION

The measured cross sections, TALYS-0.57 [1] and EMPIRE-II [8] model calculations, evaluated nuclear data files and experimental data measured by other authors are shown in Fig. 1.

The results from the TALYS calculations represent completely blind predictions. All nuclear structure parameters were independently prepared and all nuclear model parameters were taken from global systematic expressions. The EMPIRE results are default calculations and no parameter adjustment was

FIGURE 1. Excitation functions of neutron-induced light charged-particle production reactions on Zr isotopes. The present results are labeled as GEL.
made, except for the diffuseness of the alpha OMP that was reduced by 10% to improve the behavior of the \((n,\alpha)\) reaction under the barrier.

Most of the reactions are extensively studied in the 13–15 MeV region. Our results are in agreement with a large number of recent measurements around 14 MeV.

Results of cross section measurements up to 16.6 MeV have been reported in the work of Marchinkowski et al. from 1990. Our data are in agreement with those data in the case of \(^{90}\text{Zr}(n,p)^{89}\text{Y}\). For the \(^{90}\text{Zr}(n,\alpha)^{87}\text{Sr}\), \(^{91}\text{Zr}(n,p)^{90}\text{Y}\), \(^{92}\text{Zr}(n,p)^{90}\text{Y}\), \(^{96}\text{Zr}(n,\alpha)^{91}\text{Sr}\), and \(^{96}\text{Zr}(n,p)^{91}\text{Y}\) reaction cross sections the data of Marchinkowski et al. above 15 MeV are lower than the results from the present experiment.

For the \(^{91}\text{Zr}(n,x)^{90}\text{mY}\) and \(^{92}\text{Zr}(n,x)^{91}\text{mY}\) reaction cross sections our data are unique in the measured energy range. In both cases our results are consistent with the results of Ikeda at al. from 1988 [9].

Below 14 MeV the status of the data is reasonable, although certain discrepancies exist in the case of \(^{91}\text{Zr}(n,p)^{91}\text{mY}\), \(^{92}\text{Zr}(n,p)^{92}\text{Y}\), and \(^{94}\text{Zr}(n,p)^{91}\text{Y}\). So, we could consider the excitation curves as well determined by experimental data from threshold to 20 MeV.

The TALYS and EMPIRE model calculations are in reasonable agreement with the data up to 14 MeV; however above 15 MeV agreement with the data and between model calculations varies strongly, showing the relevance of experimental data for parameters, respectively, code improvement.

The new experimental data and improved model calculations could contribute to valuable new evaluations.

**SUMMARY AND CONCLUSIONS**

Activation cross sections for \(^{90}\text{Zr}(n,\alpha)^{87}\text{mSr}\), \(^{90}\text{Zr}(n,p)^{90}\text{mY}\), \(^{91}\text{Zr}(n,x)^{90}\text{mY}\), \(^{91}\text{Zr}(n,p)^{91}\text{mY}\), \(^{92}\text{Zr}(n,x)^{91}\text{mY}\), \(^{92}\text{Zr}(n,p)^{92}\text{Y}\), \(^{94}\text{Zr}(n,\alpha)^{91}\text{Sr}\), and \(^{94}\text{Zr}(n,p)^{94}\text{Y}\) reactions were measured for the first time in the energy range from 14.8 MeV to 21 MeV. The use of enriched samples allowed the separation of \((n,p)\) and \((n,p+pn+d)\) contributions to the same activity. The new experimental data improve the knowledge of the excitation functions of the investigated reactions substantially. The data were shown to be essential to benchmarking of model calculations.

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**REFERENCES**