EPJ Web of Conferences **239**, 20005 (2020) https://doi.org/10.1051/epjconf/202023920005
ND2019

The activation of ^{nat}Zr by quasi-monoenergetic neutrons below 34 MeV.

Eva Šimečková[1,*], *Martin* Ansorge[1], *Pavel* Bém[1], *Mitja* Majerle[1], *Jaromir* Mrazek[1], *Jan* Novák[1], and *Milan* Štefánik[1]

[1]Nuclear Physics Institute of the Czech Academy of Sciences, 250 68 Řež, Czech Republic

Abstract. Good knowledge of cross section neutron induced reactions on Zr becomes of importance due to the use of zirconium as structural material in reactors, its applicability in neutron dosimetry and the theoretical model testing. Thin Zr foils (0.05 mm thickness, 99.2% purity) were irradiated in the quasi-monoenergetic p-Li neutron fields, the proton beams from NPI CAS variable-energy cyclotron U120M at proton energies 20.33, 22.44, 24.69, 27.64, 29.85, 32.31 and 35.11 MeV. Li target with carbon stopper was used for the generation of neutron flux. The reaction ^{7}Li (p,n) produces the high-energy quasi-monoenergetic neutrons with a tail to lower energies. The flux density and neutron spectra were evaluated by MCNPX code and validated with set of measurements including Time-Of-Flight and Proton recoil Telescope and additional activation monitors. The pneumatic tranfer system enables the investigation of short living isotopes. The foil activity determination was performed by the nuclear spectrometry method employing two calibrated HPGe detectors. The reaction rates for ^{nat}Zr(n,*)^{89m,89g,89}Zr, ^{87m,87,88,89m,90m,91m,92,93,94,95}Y and ^{87m,91,92}Sr were obtained and cross sections were extracted. The preliminary results are discussed.

1 Introduction

Zirconium is an important reactor material, but till recently its cross section database, especially for neutron treshold reactions, was rather weak. The available data mostly covered the energy region up to 15 MeV. Besides applications, the excitation functions of neutron threshold reactions are of the considerable interest for testing of nuclear models. The theoretical description of (n,p) and (n,α) reactions on Zr isotopes was recently done in Ref. [1]. The Fast Neutron Generator (FNG) facility at the NPI CAS variable-energy cyclotron U120M provides quasi-monoenergetic neutron fields in the region of 16 - 34 MeV (see [2]).

2 Experimental arrangement

A neutron irradiation experiment was performed at NPI CAS Řež. The standard ^{7}Li(p,n) reaction on a thin lithium target induced by 20-35 MeV proton beam from the variable-energy cyclotron U120M was used for the production of quasi-monoenergetic neutron field. A self-supporting Li target cooled by 5°C alcohol stream and a carbon stopper were utilized.

The stacks of foils (Zr,Co) to be irradiated were placed 86 mm from the production target. For the cross section determination of short-living isotopes (minutes to ten minutes region) the pneumatic transfer system was used. In that case the rabbits with irradiated foil were placed in the distance of 42 mm from the front of Li target. The samples were located in the axis of the neutron field.

The time profile of the neutron source strength during the irradiation was monitored by the proton beam current on the neutron-source target, recorded by a calibrated integrator on a PC time synchronized with the gamma spectroscopy device.

Table 1. Characteristics of separate runs

E_p(MeV)	Irr.time(s)	Q(μC)	I_{mean}(μA)
20.31(15)	31286	1.807E+05	5.78
22.44(15)	24031	1.446E+05	6.01
24.76(15)	29587	1.774E+05	6.00
27.51(15)	30371	1.818E+05	5.99
29.85(15)	32199	1.992E+05	6.19
32.31(15)	26489	1.876E+05	7.08
35.11(15)	29773	1.752E+05	5.89

3 Data acquisition and methods of results evaluation

The ^{nat}Zr disks (of 99.2 % purity, Goodfellow product) of 15 mm diameter and 0.05 mm thickness were irradiated in seven separated runs in various neutron fields (see Table 1). Futhermore, for the production cross section determination of short lived isotopes the samples were irradiated roughly 5 minutes and transported in front of HPGe detector during 30 - 40 s.

3.1 Neutron fields

The reaction ^{7}Li(p,n) produces the high-energy quasi-monoenergetic neutron spectra with a monoenergetic peak at the energy of the incident protons (corrected for Q value and broadened because of the proton ionization losses in

*e-mail: simeckova@ujf.cas.cz

Table 2. Inventory of the observed radioactive nuclides

Nuclide	$T_{1/2}$	E_γ(keV)	I_γ(%)
^{87m}Y	13.37 h	380.8	78
^{87}Y	79.8 h	484.8	89.7
		388.5	82
^{87}Sr	2.803 h	388.5	81.9
^{91m}Y	49.71 min	555.6	95
^{91}Sr	9.63 h	1024.3	33
		749.8	23.61
^{93}Y	10.18 h	266.9	7.3
		947.1	2.09
^{95}Y	10.3 min	954.0	16

the lithium target) and the continuum spectra at lower energies (Figure 1).The neutron spectra and fluxes were calculated by MCNPX code using ^{7}Li(p,n) cross section data from libraries LA150H and JENDL-4.0/HE. The MCNPX calculated spectra were corrected according to the experimental measurements of the peak neutrons using the Time-Of-Flight method and the HPGe measurements of the producted ^{7}Be activity in the lithium target. Details are given in [3].

3.2 Gamma spectroscopy measurement

The gamma-rays from irradiated foils were measured repeatedly by two calibrated HPGe detectors of 50% efficiency and of FWHM 1.8 keV at 1.3 MeV. The inventory of the observed radioactive isotopes was taken from the Lund/LBNL Nuclear data Search [4] and is presented in Table 2.

3.3 Cross section evaluation

Experimental reaction rates were calculated from the specific activities at the end of irradiation corrected to the decay during the irradiation using total charges and foil characteristics as well.

The reaction rate RR is defined as the number of produced residual nuclei per atom of the sample target and per number of the incident protons (of neutron source). It corresponds to the integral over energy of a product of the neutron spectral fluency ϕ at the position of the sample and the cross section σ.

In the determination of cross section in the energy interval of neutron peak, the neutron spectra of each i-run were split into the parts corresponding to the the energy interval of separate monoenergetic peak j and the rest part corresponding to the low energy tail of the spectrum (Figure 1). In this procedure, the following set of equations

$$RR_i = \sum_{j}^{k} \sigma_j \phi_{ji} \triangle E_j + \sum_{l_{Eth}}^{l_{Emin=15MeV}} \sigma_l \phi_{li} \triangle E_l \qquad (1)$$

was solved using minimalization procedures with respect to cross section σ_j in the j-energy interval. Here k is total number of runs (k=7), l is number of the energy interval corresponding to the energy bin (0.25 MeV) in evaluated

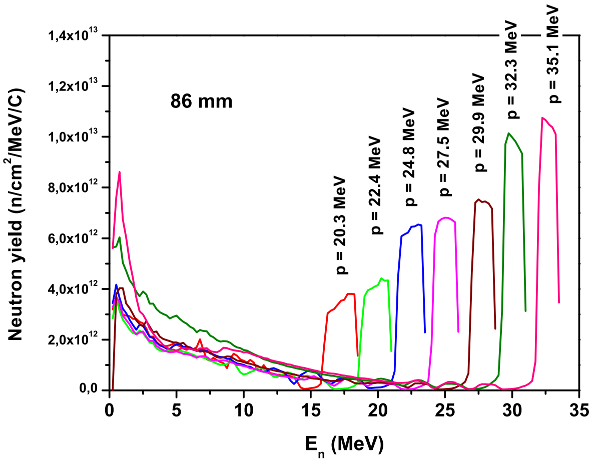

Figure 1. Spectral neutron flux of the ^{7}Li+p reaction at sample position x = 86 mm. Proton incident energies are shown.

spectral neutron flux, E_{th} - theshold of the reaction and E_{min} = 15 MeV (corresponds to the lowest energy which we are able to determine cross section using only our measured data).

No knowledge of the activation cross section curve was needed for the reactions having the threshold energy above the first quasi-monoenergetic peak in the fist run j = 1 (of about15 MeV). In other cases, experimental cross sections from EXFOR database (σ_l) were utilized in the calculation (1).

4 Results and discussion

Natural zirconium consists of five stable isotopes. The neutron cross sections were studied previously mainly only up to 15 MeV. We determined neutron cross sections for the ^{89m,89g,89}Zr, ^{87m,87,88,89m,90m,91m,92,93,94,95}Zr and ^{87m,91,92}Sr for the first time. In the next figures typical examples of our resulting cross section in the energy region 15 - 34 MeV are shown together with the data of the other authors from EXFOR databases and compared to the TENDL 2017 neutron library [5]. All data are renormalized to the natural abundance of Zr. The statistical errors of the activity determination were 1-2%, the beam charge uncertaity was 5%. Sensitivity analysis [3] using the data libraries (LA150H, JENDL-4.0/HE) resulted in the estimated neutron flux uncertainties from 3% up to 7.5%.

The cross section data for the ^{nat}Zr(n,x)^{87m}Y reaction (see Figure 2a)) and for the ^{nat}Zr(n,x)^{87}Y reaction (see Firure 2 b)) are determined for the first time. Our data are in agreement with TENDL 2017 library.

In the case of ^{87m}Sr production (see Figure 2 c)) we had to take into acount the more recent data of other authors studying ^{90}Zr(n,α) reaction up to 15 MeV, Ref [6], [7], [8] and [9]. No data were observed for the ^{nat}Zr(n,x)^{87m}Sr reaction in energy region above 15 MeV. Our data from long-time irradiation and short-time one are

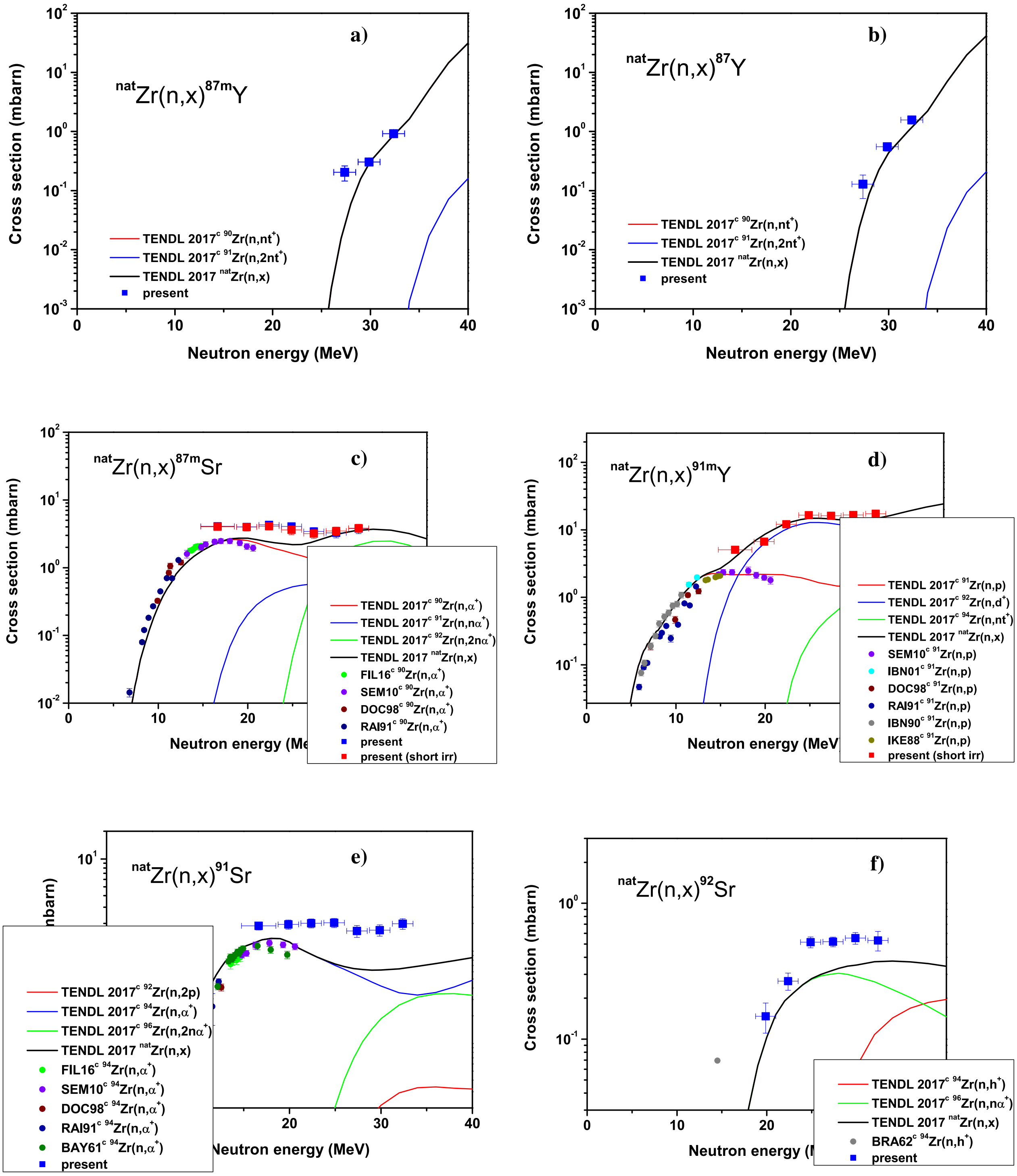

Figure 2. Production cross sections of the ^{nat}Zr(n,x)^{87m}Y (a), ^{nat}Zr(n,x)^{87}Y (b), ^{nat}Zr(n,x)^{87m}Sr (c), ^{nat}Zr(n,x)^{91m}Y (d), ^{nat}Zr(n,x)^{91}Sr (e) and ^{nat}Zr(n,x)^{92}Sr (f) reactions. Literature data and TENDL 2017 curves are renormalized to the natural abundances of Zr. The blue points corresponds to the long time irradiation, red ones the the short irradiation.

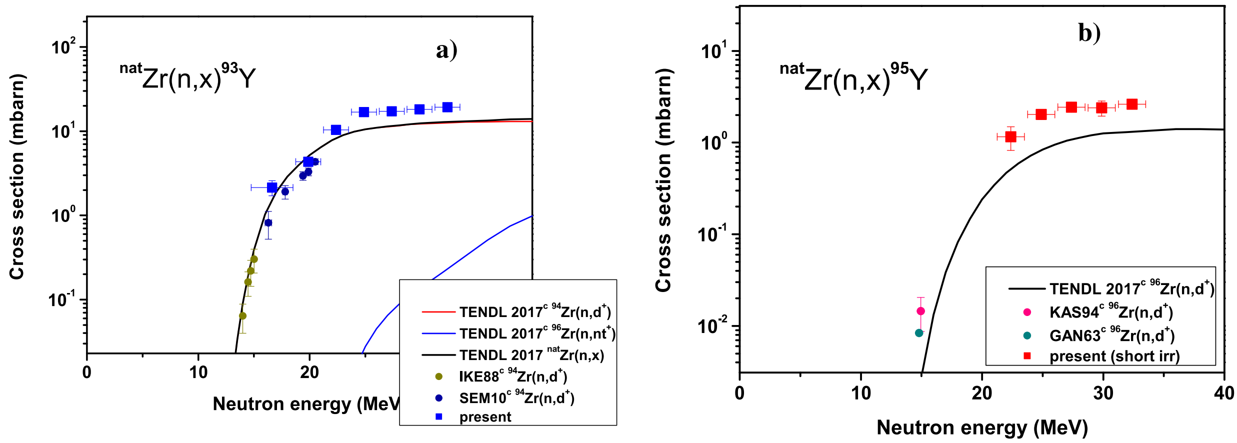

Figure 3. Production cross sections of ^{nat}Zr(n,x)^{93}Y a) and ^{nat}Zr(n,x)^{95}Y b) reactions.

in a mutual agreement. They are in a rough agreement and with TENDL 2017.

The ^{nat}Zr(n,x)^{91m}Y reaction cross sections shown in Figure 2 d) are evaluated on the base of our experimental data and data from Refs. [7], [10], [8], [9], [11] and [12]. Our data are in full agreement with the TENDL 2017 library.

On the other hand the data for ^{nat}Zr(n,x)^{91}Sr (mainly the ^{94}Zr(n,α)^{91}Sr in that energy region of interest) are higher than TENDL 2017 evaluated curve - see Figure 2 e). For cross section determination we used the experimental data of Ref. [6], [7], [8], [9] and [13].

Similar case is for the reaction ^{nat}Zr(n,x)^{92}Sr shown in Figure 2 f), where TENDL 2017 underestimated experimental data, mainly for (n,α) reaction. Besides our data there is only one old measurement [14].

In the Figure 3 a) the production cross sections for the ^{nat}Zr(n,x)^{93}Y are shown together with the data of authors [7] and [12]. All the data are in agreement up to about 22 MeV.

In Figure 3 b) the cross sections of the ^{nat}Zr(n,x)^{95}Y, where the only ^{94}Zr(n,d)^{95}Y reaction is posible, are shown together with the data of authors [15] and [16].

Some of preliminary data of ^{nat}Zr(n,x) reactions are presented. Data are in satisfactory agreement with the TENDL 2017 library. Discrepances are in the 20-30 MeV region. Data analysis will continue.

Acknowledgements

The authors are indebted to the operating crew of the U120M cyclotron and the project CANAM that supported the operation of the cyclotron. This publication was supported by MEYS, under the project SPIRAL2-CZ, CZ.02.1.01/0.0/0.0/16_013/0001679.

References

[1] V.Avrigeanu and M. Avrigeanu, Phys. Rev. C **96**, 044610 (2017)

[2] P.Bém, V.Burjan, J.Dobeš, U.Fischer, M.Gotz, M.Honusek, V.Kroha, J.Novák, S.P.Simakov and E.Šimečková, *Proc. Int. Conf. on Nuclear Data for Science and Technology (ND2007)* (Nice, France, 2007) 983

[3] M.Majerle, M.Ansorge, P.Bém, J.Novák, E.Šimečková and M.Štefánik, *INDC(NDS) Reports No. 0789*, 2019

[4] S.Y.F.Chu, L.P.Ekström and R.B.Firestone, The Lund/LBNL Nuclear Data Search, Version 2.0, 1999

[5] A.J.Konnong and D.Rochman, Nuclear data Sheets **113**, 2841 (2012)

[6] A.A.Filankov, *USSR reports to the INDC (NDS) No.0460*, 2016

[7] V.Semkova, E.Bauege, A.J.M.Plompen and D.L.Smith, Nucl. Phys. A **832**, 149 (2010)

[8] R.Dóczi, V.Semkova, A.Fenyvesi, N.Yamamuro, Cs.M.Buczkó and J.Csikai, Nucl. Science and Engineering **129**, 164 (1998)

[9] P.Raics, S.Nagy, S.Szegeli, N.V.Kornilov and A.B.Kagalenko, *Conf. on Nucl. Data for Sci. and Technol.* (Juelich, 1991) 660

[10] M.Ibn Majah, A.Chiadli, S.Sudár and S.M.Qaim, Appl. Radiation and Isotopes **54**, 655 (2001)

[11] M.Ibn Majah and S.M.Qaim, Nucl. Sci. and Engineering **104**, 271 (1990)

[12] Y.Ikeda, C.Konno, K.Oishi, T.Nakamura, H.Miyade, K.Kawade, H.Yamamoto and T.Katoh, *JAERI Reports No. 1312*, 1988

[13] M.IbnMajah, and S.M.Qaim, Nucl. Sci. and Engineering **104**,271 (1990)

[14] E.T.Bramlitt, R.W.Fink, D.G.Gardner and A.Poularikas, Phys. Rev. **125**, 297 (1962)

[15] Y.Kasugai, H.Yamamoto, K.Kawage, Y.Ikeda, Y.Uno and H.Maekawa, *Conf. on Nucl. Data for Sci. and Technol.* (Gatlinburg 2, 1994) 935

[16] Y.Ganguly and H.Bakhru, *Nucl. and Sol. State Physics Symp.* (Bambay, 1963) 240