HAL
open science

New excitation functions measurement of nuclear reactions induced by deuteron beams on yttrium with particular reference to the production of ^{89}Zr

Simone Manenti, Ferid Haddad, Flavia Groppi

▶ **To cite this version:**

Simone Manenti, Ferid Haddad, Flavia Groppi. New excitation functions measurement of nuclear reactions induced by deuteron beams on yttrium with particular reference to the production of ^{89}Zr. Nucl.Instrum.Meth.B, 2019, 458, pp.57-60. 10.1016/j.nimb.2019.08.002 . hal-02268463

HAL Id: hal-02268463

https://hal.science/hal-02268463v1

Submitted on 19 Mar 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

New excitation functions measurement of nuclear reactions induced by deuteron beams on yttrium with particular reference to the production of ^{89}Zr

Simone Manenti[a,b,*], Ferid Haddad[c,d], Flavia Groppi[a,b]

[a] Department of Physics, Università degli Studi di Milano, via Celoria 16, I-20133 Milano, Italy
[b] LASA, Department of Physics, Università degli Studi di Milano and INFN-Milano, via F.lli Cervi 201, I-20090 Segrate MI, Italy
[c] SUBATECH, Ecole des Mines de Nantes, Université de Nantes, CNRS/IN2P3, Nantes, France
[d] GIP Arronax, 1 rue Aronnax, 44817 Saint-Herblain, France

We investigated ^{89}Zr production induced by deuteron beams on yttrium targets at energies up to E_d = 32 MeV using the stacked-foil activation technique. Cross sections of the following nuclear reactions ^{89}Y(d,2n)^{89}Zr, ^{89}Y(d,3n)^{88}Zr and ^{89}Y(d,x)^{88}Y have also been measured. Based on the measured values, we determined the thick target yields for ^{89}Zr and ^{88}Zr which is the main contaminant associated to the production of ^{89}Zr.

1. Introduction

Thanks to its nuclear characteristics ($t_{1/2}$ = 78.41 h, 22.3% positron emission with a maximum decay energy of 900 keV), zirconium-89 (^{89}Zr) is a very promising radionuclide for immuno-PET (positron emission tomography using an antibody to target the cells of interest) [1–5]. It can also be used for bio distribution studies of labelled monoclonal antibodies [6]; furthermore, in literature it was reported a method to prepare pharmaceuticals for simultaneous magnetic resonance imaging and PET [7].

Our work presents and discusses the experimental determination of the cross-sections of the ^{89}Y(d,2n)^{89}Zr reaction in the 6–32 MeV energy range.

The earlier results for nuclear reactions induced by deuteron beams on Y target were published by Baron and Cohen [8], La Gamma and Nassif [9], Bissem et al. [10], Degering et al. [11], West et al. [12], Uddin et al. [13], Tárkányi et al. [14] and Lebeda et al. [15] and are rather scattered. These data will be compared with ours results, which contribute with a new set of experimental values.

In parallel, we evaluated the excitation functions for the co-produced zirconium-88 (^{88}Zr) and yttrium-88 (^{88}Y).

^{88}Zr, with $t_{1/2}$ = 83.4 d and single gamma-ray emission (392.87 keV), is the only radioisotopic impurity in ^{89}Zr production by deuterons on ^{89}Y and it has an impact on the specific activity of the final production.

^{88}Y has an half-life of 106.65 d and it may have an impact on waste management during the production process.

2. Experimental

We determined the excitation functions using stacks of high purity aluminium (as degrader, monitor and catcher foil, Goodfellow Cambridge Ltd., purity 99.999%, 4.30 mg·cm^{-2}), yttrium (as target foil) and titanium (as monitor foil, Goodfellow Cambridge Ltd., purity 99.6 + %, 8.80 mg·cm^{-2}): each stack was composed of four couples of Y and Al foils, by one couple of Ti and Al foils after each Y/Al foils and by some added aluminium foils as degraders.

IAEA tabulated monitor reactions [16] – ^{nat}Ti(p,x)^{48}V and ^{27}Al(d,x)^{24}Na reactions – were used for the determination of beam intensity and energy. ^{89}Y targets (Goodfellow Cambridge Ltd., purity 99.6 + %) had a nominal thickness of 25 μm (i.e. 11.17 mg·cm^{-2}). We verified the homogeneity of the target with an analog thickness gauge (resolution 0.001 mm) and we measured accurately by weighing the value of target thickness: 11.13 mg·cm^{-2} (used in the 28.1–32.3 MeV and 18.2–21.0 MeV energy ranges) and 12.37 mg·cm^{-2} (in all the other energy ranges) with a relative uncertainty of ± 2%.

* Corresponding author at: LASA, Department of Physics, Università degli Studi di Milano and INFN-Milano, via F.lli Cervi 201, I-20090 Segrate MI, Italy.
E-mail address: simone.manenti@mi.infn.it (S. Manenti).

The irradiations were performed with the cyclotron IBA C70 of the ARRONAX center (Saint-Herblain, France) [17]: the mean beam intensity was about 180 nA and irradiations duration was 1 h.

We irradiated six stacks with different incident energies in order to limit the energy straggling and the energy spread to the minimum in the energy interval from 6 MeV up to 32 MeV. The stacks were irradiated in air with an external beam line; the line was closed by a kapton foil with a thickness of 75 µm and the distance between the beam line window and the first foil in the stacks was 82 mm.

SRIM 2013 [18] was used to compute the mean deuteron beam energy in each foil. The uncertainty of the mean energy includes the energy straggling of the beam through the target foils, the uncertainties related to the mean areal density and the energy uncertainty of the extracted deuteron beam.

Decay radiations associated to each radionuclide were measured without any chemical processing at the LASA laboratory (Segrate, INFN and Physics Dept. of University of Milan), by a calibrated HPGe (high purity germanium, 15% relative efficiency) detector. We measured the samples periodically for six months starting the first measurement within 48 h after end of bombardment (EOB).

To calculate the overall uncertainty related to the experimental cross sections, the several error sources reported in [19] are taken into account: this overall uncertainty has a relative error of 6–15%.

Relative uncertainties of the half-lives and the gamma emission intensities are very small (usually ≪0.1%) and, therefore, are neglected in the overall uncertainty calculations.

The decay characteristics for the radionuclides investigated are taken from [20,21] and are summarized in Table 1.

3. Results

We measured the thin foils by gamma spectrometry and calculated the cross-sections $\sigma(E)$ (mb) for each target as described in [19]: our data are presented in Table 2.

The measured excitation functions are compared with the literature data in Figs. 1, 3 and 5. Theoretical values were also extracted from TENDL-2017 [22] and EMPIRE-3.2.2 [23] and are also presented on Figs. 1, 3 and 5.

In order to bring more quantitative considerations, we calculated the Thick Target Yield (TTY) [24] for the production of ^{89}Zr and ^{88}Zr. Figs. 2 and 4 show the resulting TTY compared with experimental TTY available in literature (Dmitriev et al. [25] and Zweit et al. [26]) and a proton one [27].

3.1. ^{89}Y(d,2n)^{89}Zr

^{89}Zr has a half-life of 78.41 h and can be produced through the (d,2n) reaction. We assessed the activity through the 908.96 keV gamma line (I_γ = 99.87%). ^{89}Zr has a short-lived isomer ^{89m}Zr ($t_{1/2}$ = 4.18 min, isomeric transition probability equal to 93.77%) that was not measured: we measured a cumulative production of $^{89,m+g}$Zr.

Fig. 1 reports the measured experimental cross-sections together with curves from theoretical calculations with EMPIRE and TALYS codes (TENDL-2017) and the data of the earlier studies. Our cross-sections are in good agreement with the results of West et al. [12], Uddin et al. [27] and Lebeda et al. [15]. All the experimental data are lower than the prediction of TENDL-2017 while they are underestimated by the prediction of EMPIRE 3.2.2 in the energy range 7–21 MeV.

Fig. 2 shows the calculated TTY, two experimental TTY available in literature (Dmitriev al. [25] and Zweit et al. [26]) and a proton one (dash-dot line, Uddin et al. [27]).

A very good agreement between the curves calculated from data of the present work and the experimental data points can be seen. A comparison between a curve for the proton production and our curve for the deuteron production of ^{89}Zr shows that the proton one is higher up to 26.5 MeV leading to higher production yield. For higher particle energies the achievable TTY with protons is lower than with deuterons.

Table 1
Zr and Y radionuclides decay data [20] and contributing reactions. The E_{th} is evaluated on the base of the mass defects in [21].

Nuclide	$t_{1/2}$	Contributing reactions	E_{th} (MeV)	E_γ (keV)	I_γ (%)
^{89}Zr	78.41 h	^{89}Y(d,2n)^{89}Zr	5.97	908.96	99.87
^{88}Zr	83.4 d	^{89}Y(d,3n)^{88}Zr	15.50	392.87	97.31
^{88}Y	106.65 d	^{89}Y(d,t)^{88}Y	5.34	898.04	93.7
		^{89}Y(d,dn)^{88}Y	11.74		
		^{89}Y(d,p2n)^{88}Y	14.02		

Table 2
Experimental cross-sections (one standard deviation) of the ^{89}Y(d,xn)^{89,88}Zr and ^{89}Y(d,x)^{88}Y reactions.

Energy (MeV)	^{89}Zr (mb)	^{88}Zr (mb)	^{88}Y (mb)
6.0 ± 0.3	8.62 ± 0.70		
9.6 ± 0.3	446 ± 36		0.255 ± 0.023
12.3 ± 0.3	728 ± 59		3.77 ± 0.22
14.1 ± 0.3	857 ± 70		7.63 ± 0.44
14.7 ± 0.3	851 ± 69		8.56 ± 0.50
15.4 ± 0.3	897 ± 73	0.164 ± 0.018	9.66 ± 0.79
15.6 ± 0.3	866 ± 70	0.106 ± 0.016	9.89 ± 0.57
17.1 ± 0.3	895 ± 73	6.54 ± 0.53	14.84 ± 0.85
17.6 ± 0.3	928 ± 75	9.74 ± 0.79	13.2 ± 1.6
17.9 ± 0.3	901 ± 73	20.8 ± 1.7	18.0 ± 1.6
18.2 ± 0.3	946 ± 77	25.1 ± 2.0	18.0 ± 1.5
19.6 ± 0.3	907 ± 74	94.9 ± 7.7	41.5 ± 3.4
19.6 ± 0.3	815 ± 66	68.9 ± 5.6	30.3 ± 2.9
21.0 ± 0.3	730 ± 59	192 ± 16	65.9 ± 5.4
21.5 ± 0.3	685 ± 56	210 ± 17	67.8 ± 5.9
23.1 ± 0.3	597 ± 48	392 ± 32	109.9 ± 9.3
24.8 ± 0.3	449 ± 36	498 ± 40	144 ± 12
26.4 ± 0.3	370 ± 30	601 ± 49	207 ± 17
27.9 ± 0.3	292 ± 24	629 ± 51	240 ± 20
28.1 ± 0.4	312 ± 25	653 ± 53	254 ± 21
29.6 ± 0.4	252 ± 20	646 ± 52	292 ± 24
31.0 ± 0.3	220 ± 18	651 ± 53	319 ± 27
32.3 ± 0.3	197 ± 16	634 ± 52	348 ± 28

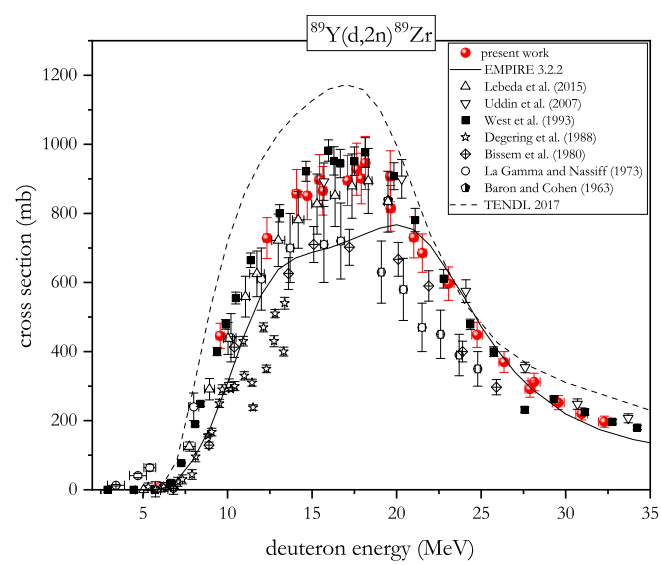

Fig. 1. Excitation functions for ^{89}Y(d,2n)^{89}Zr nuclear reactions.

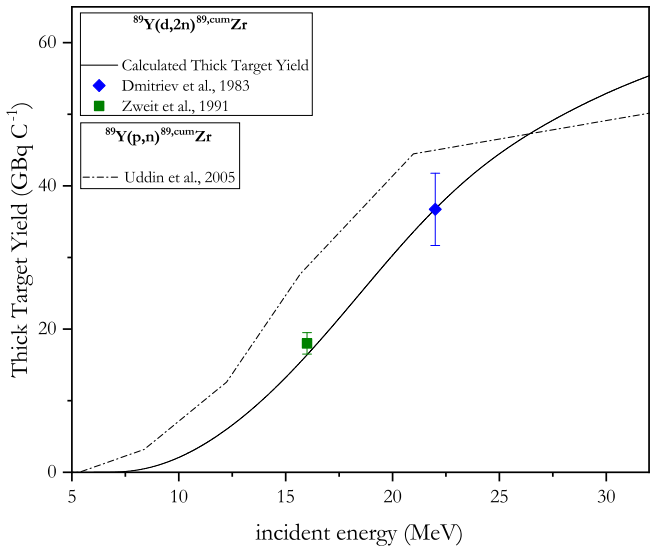

Fig. 2. Thick Target Yield for ^{89}Y(d,2n)^{89}Zr nuclear reactions compared with TTY obtained from (p,n) reaction [27].

3.2. $^{89}Y(d,3n)^{88}Zr$

^{88}Zr has a half-life of 83.4 d and can be produced through the (d,3n) reaction. The activity was assessed through the 392.87 keV gamma line (I_γ = 97.31%).

Our cross-sections (Fig. 3) are in good agreement with the two previous results available in the same energy range [12,15]. The experimental data from the work of La Gamma and Nassif [9] are higher than ours and these data are probably wrong, while data from Tárkányi et al. [14] are lower. Still in this case, the prediction of TENDL-2017 is higher than all the experimental data. However, EMPIRE 3.2.2 gives a good description of the cross-sections.

Also in this case, Fig. 4 shows the resulting TTY in comparison with one experimental TTY available in literature (Dmitriev al. [25]) and the proton one (dash-dot line, Uddin et al. [27]).

For all particle energies the achievable TTY with protons is higher than with deuterons. This indicates that more contaminants are produced using proton beams than using deuteron beams. We can then expect a better purity of the ^{89}Zr produced by deuteron induced reaction on ^{89}Y.

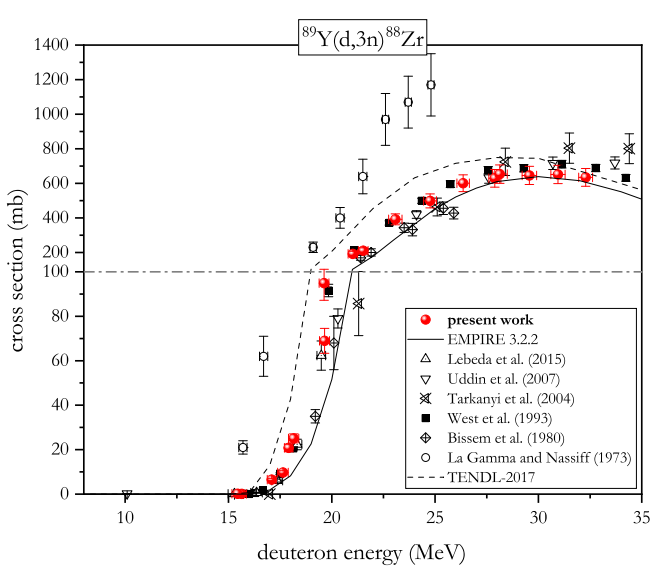

Fig. 3. Excitation functions for ^{89}Y(d,3n)^{88}Zr nuclear reactions.

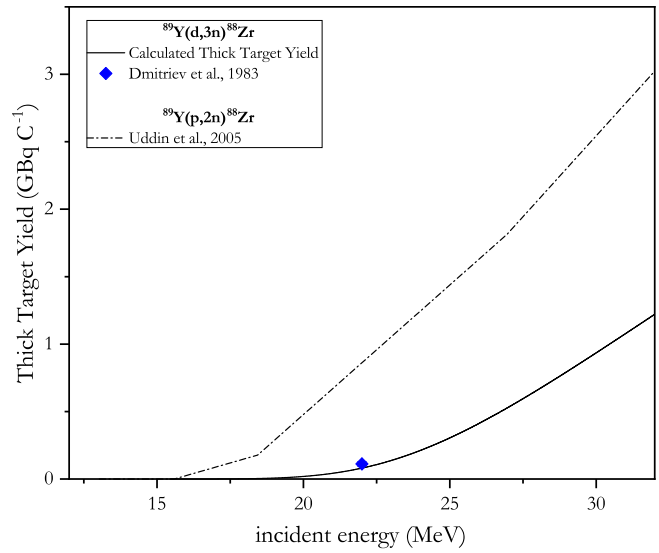

Fig. 4. Thick Target Yield for ^{89}Y(d,3n)^{88}Zr nuclear reactions compared with TTY obtained from (p,n) reaction [27].

3.3. $^{89}Y(d,x)^{88}Y$

^{88}Y has a half-life of 106.65 d and the activity was assessed through the 898.04 keV gamma line (I_γ = 93.7%); it can be produced through the (d,x) reactions.

Our cross-sections are, in general, in good agreement with the results of Lebeda et al. [15], Uddin et al. [13], Tárkányi et al. [14] and West et al. [12] (Fig. 5). The experimental data from the work of La Gamma and Nassif [9] are lower than ours. Also TENDL-2017 is in good agreement with our experimental points while EMPIRE 3.2.2 gives higher values above 20 MeV. For both nuclear code, low energy values are not correct. In this region, the (d,t) reaction is the only one in place which means that these nuclear code must improve this reaction mechanism.

4. Conclusions

^{89}Zr is extensively used in the research of new PET radiopharmaceuticals.

We presented the excitation functions of the reactions on ^{89}Y

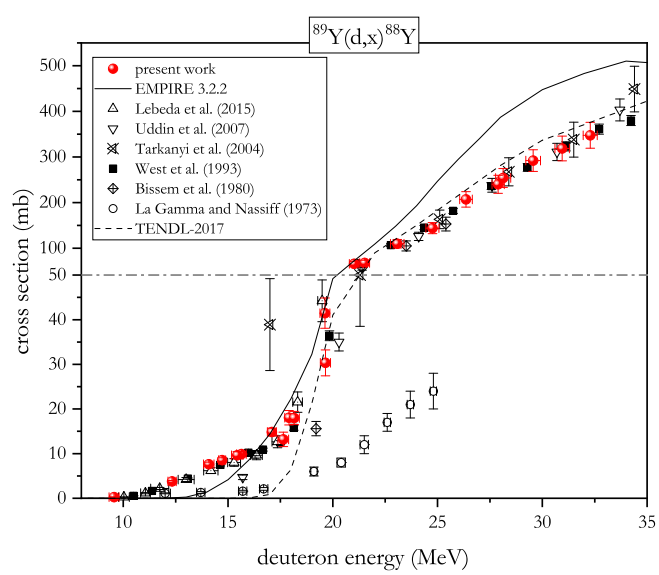

Fig. 5. Excitation functions for ^{89}Y(d,x)^{88}Y nuclear reactions.

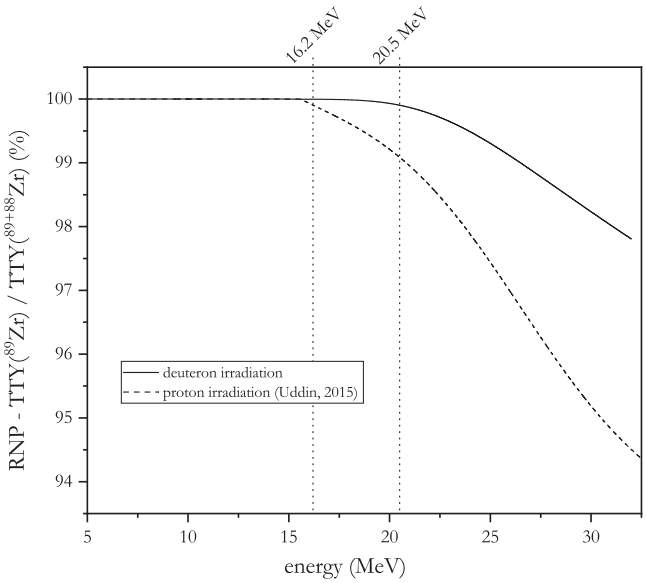

Fig. 6. Radionuclidic purity at the end of an instantaneous bombardment.

induced by deuteron beams up to 32 MeV: significant amounts of ^{89}Zr can be produced by accelerators using deuteron beams and yttrium as a target. A very low amount radionuclidic impurities is produced in the energy range considered in this study (Fig. 6).

From Fig. 6 we can see that a define radionuclidic purity of 99.9% corresponds to a higher beam energy for deuterons (20.5 MEV) than for protons (16.2 MeV). This turns out in a 9% higher production yield for deuteron.

So, the use of deuteron beams bring an advantage in term of activity production with respect to the use of proton beams and, at the same time, it requires a less amount of ^{89}Y to be involved in the radiochemical separation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was funded in the framework of the research project TECHN-OSP by Istituto Nazionale di Fisica Nucleare - INFN (Italian National Institute of Nuclear Physics, CSN5) and it was, in part, supported by grants from the French National Agency for Research called "Investissements d'Avenir", Equipex Arronax-Plus no ANR-11-EQPX-0004, Labex IRON no ANR-11-LABX-18-01 and ISITE NExT no ANR-16-IDEX-007.

References

[1] V. Tolmachev, S. Stone-Elander, Radiolabelled proteins for positron emission tomography: pros and cons of labelling methods, Biochim. Biophys. Acta – General Subjects 1800 (5) (2010) 487–510, https://doi.org/10.1016/j.bbagen.2010.02.002.

[2] S. Knowles, A. Wu, Advances in immuno-positron emission tomography: antibodies for molecular imaging in oncology, J. Clin. Oncol. 30 (31) (2012) 3884–3892, https://doi.org/10.1200/JCO.2012.42.4887.

[3] M. Deri, B. Zeglis, L. Francesconi, J. Lewis, PET imaging with ^{89}Zr: from radiochemistry to the clinic, Nucl. Med. Biol. 40 (1) (2013) 3–14, https://doi.org/10.1016/j.nucmedbio.2012.08.004.

[4] A. Bansal, M. Pandey, Y. Demirhan, J. Nesbitt, R. Crespo-Diaz, A. Terzic, A. Behfar, T. DeGrado, Novel ^{89}Zr cell labeling approach for PET-based cell trafficking studies, EJNMMI Research 5 (1).10.1186/s13550-015-0098-y.

[5] D. Brasse, A. Nonat, Radiometals: towards a new success story in nuclear imaging? Dalton Trans. 44 (11) (2015) 4845–4858, https://doi.org/10.1039/c4dt02911a.

[6] F. Bensch, M.M. Smeenk, S.C. van Es, J.R. de Jong, C.P. Schröder, S.F. Oosting, M.N. Lub-de Hooge, C.W. Menke-van der Houven van Oordt, A.H. Brouwers, R. Boellaard, E.G.E. de Vries, Comparative biodistribution analysis across four different ^{89}Zr-monoclonal antibody tracers-the first step towards an imaging warehouse, Theranostics 8 (30214621) (2018) 4295–4304, https://doi.org/10.7150/thno.26370.

[7] D. Abou, D. Thorek, N. Ramos, M. Pinkse, H. Wolterbeek, S. Carlin, B. Beattie, J. Lewis, ^{89}Zr-labeled paramagnetic octreotide-liposomes for PET-MR imaging of cancer, Pharm. Res. 30 (3) (2013) 878–888, https://doi.org/10.1007/s11095-012-0929-8.

[8] N. Baron, B.L. Cohen, Activation cross-section survey of deuteron-induced reactions, Phys. Rev. 129 (1963) 2636–2642, https://doi.org/10.1103/PhysRev. 129.2636.

[9] A. La Gamma, S. Nassiff, Excitation functions for deuteron-lnduced reactions on ^{89}Y, Radiochim. Acta 19 (4) (1973) 161–162, https://doi.org/10.1524/ract.1973.19.4.161.

[10] H.H. Bissem, R. Georgi, W. Scobel, J. Ernst, M. Kaba, J.R. Rao, H. Strohe, Entrance and exit channel phenomena in d- and ^{3}He-induced preequilibrium decay, Phys. Rev. C 22 (1980) 1468–1484, https://doi.org/10.1103/PhysRevC.22.1468.

[11] D. Degering, S. Unterricker, W. Stolz, Excitation function of the ^{89}Y(d,2n)^{89}Zr reaction, J. Radioanal. Nucl. Chem. 127 (1) (1988) 7–11, https://doi.org/10.1007/BF02165500.

[12] H.L.J. West, M.G. Mustafa, H. O'Brien, R.G. Lanier, Some Light-Ion Excitation-Function Measurements on Titanium, Yttrium, and Europium, and Associated Results, 1993, Ch. Some Excitation Functions of Proton and Deuteron Induced Reactions on ^{89}Y, pp. 4-1-4-15.https://doi.org/10.2172/10142277.

[13] M.S. Uddin, M. Baba, M. Hagiwara, F. Tárkányi, F. Ditrói, Experimental determination of deuteron-induced activation cross sections of yttrium, Radiochim. Acta 95 (4) (2007) 187–192, https://doi.org/10.1524/ract.2007.95.4.187.

[14] F. Tárkányi, F. Ditrói, S. Takács, J. Csikai, I. Mahunka, M.S. Uddin, M. Hagiwara, M. Baba, T. Ido, A. Hermanne, M. Sonck, Y. Shubin, A.I. Dityuk, Excitation functions for production of ^{88}Zr and ^{88}Y by proton and deuteron irradiation of Mo, Nb, Zr, and Y, AIP Conf. Proc. 769 (1) (2005) 1658–1661, https://doi.org/10.1063/1.1945326.

[15] O. Lebeda, J. Štursa, J. Ráliš, Experimental cross-sections of deuteron-induced reaction on ^{89}Y up to ^{20} MeV; comparison of ^{nat}Ti(d, x)^{48}V and ^{27}Al(d, x)^{24}Na monitor reactions, Nucl. Instrum. Methods Phys. Res., Sect. B 360 (2015) 118–128, https://doi.org/10.1016/j.nimb.2015.08.036.

[16] A. Hermanne, A. Ignatyuk, R. Capote, B. Carlson, J. Engle, M. Kellett, T. Kibédi, G. Kim, F. Kondev, M. Hussain, O. Lebeda, A. Luca, Y. Nagai, H. Naik, A. Nichols, F. Nortier, S. Suryanarayana, S. Takács, F. Tárkányi, M. Verpelli, Reference cross sections for charged-particle monitor reactions, Nucl. Data Sheets 148 (2018) 338–382, https://doi.org/10.1016/j.nds.2018.02.009 special Issue on Nuclear Reaction Data.

[17] F. Haddad, L. Ferrer, A. Guertin, T. Carlier, N. Michel, J. Barbet, J.-F. Chatal, ARRONAX, a high-energy and high-intensity cyclotron for nuclear medicine, Eur. J. Nucl. Med. Mol. Imaging 35 (7) (2008) 1377–1387, https://doi.org/10.1007/s00259-008-0802-5.

[18] J.F. Ziegler, M.D. Ziegler, J.P. Biersack, SRIM – the stopping and range of ions in matter (2010), Nucl. Instrum. Methods Phys. Res., Section B 268 (11–12) (2010) 1818–1823, https://doi.org/10.1016/j.nimb.2010.02.091.

[19] S. Manenti, M.D.C. Alí Santoro, G. Cotogno, C. Duchemin, F. Haddad, U. Holzwarth, F. Groppi, Excitation function and yield for the ^{103}Rh(d,2n)^{103}Pd nuclear reaction: optimization of the production of palladium-103, Nucl. Med. Biol. 49 (2017) 30–37, https://doi.org/10.1016/j.nucmedbio.2017.02.005.

[20] R.B. Firestone, C.M. Baglin, S.Y.F. Chu, Table of Isotopes: 1999 Update with CD-ROM, 1999 Update, eighth ed., Wiley-Interscience, 1999.

[21] M. Wang, G. Audi, A.H. Wapstra, F.G. Kondev, M. MacCormick, X. Xu, B. Pfeiffer, The Ame 2012 atomic mass evaluation 36 (12) (2012) 1603–2014.https://doi.org/10.1088/1674-1137/36/12/003.

[22] A. Koning, D. Rochman, Modern nuclear data evaluation with the TALYS code system, Nucl. Data Sheets 113 (12) (2012) 2841–2934, https://doi.org/10.1016/j.nds.2012.11.002 special Issue on Nuclear Reaction Data.

[23] M. Herman, R. Capote, B. Carlson, P. Obložinský, M. Sin, A. Trkov, H. Wienke, V. Zerkin, EMPIRE: nuclear reaction model code system for data evaluation, Nucl. Data Sheets 108 (2007) 2655–2715, https://doi.org/10.1016/j.nds.2007.11.003.

[24] M. Bonardi, Proceedings of the IAEA Consultants' Meeting on Data Requirements for Medical Radioisotope Production, Tokyo, Japan, 20–24 April 1987, Vol. INDC(NDS)-195/GZ, IAEA, 1987, Ch. The contribution to nuclear data for biomedical radioisotope production from the Milan Cyclotron Laboratory, pp. 98–112.

[25] P.P. Dmitriev, N.N. Krasnov, G.A. Molin, Yields of radioactive nuclides formed by bombardment of a thick target with 22-MeV deuterons, Tech. Rep. INDC(CCP)-210/L, 1983.

[26] J. Zweit, S. Downey, H. Sharma, Production of no-carrier-added zirconium-89 for positron emission tomography, Int. J. Radiat. Appl. Instrum. Part A. Appl. Radiat. Isotopes 42 (2) (1991) 199–201, https://doi.org/10.1016/0883-2889(91)90074-B.

[27] M. Uddin, M. Hagiwara, M. Baba, F. Tárkányi, F. Ditrói, Experimental studies on excitation functions of the proton-induced activation reactions on yttrium, Appl. Radiat. Isot. 63 (3) (2005) 367–374, https://doi.org/10.1016/j.apradiso.2005.04.006.