IL NUOVO CIMENTO **45 C** (2022) 99
DOI 10.1393/ncc/i2022-22099-2

COMMUNICATIONS: SIF Congress 2021

Advancements in nuclear reaction modeling for innovative medical radioisotope production

A. COLOMBI(1)(2)(*), F. BARBARO(1)(3), L. CANTON(3), M. P. CARANTE(1)(2) and A. FONTANA(2)

(1) *Dipartimento di Fisica, Università di Pavia - Pavia, Italy*
(2) *INFN, Sezione di Pavia - Pavia, Italy*
(3) *INFN, Sezione di Padova - Padova, Italy*

received 26 January 2022

Summary. — A method is presented to improve the agreement between theoretical excitation functions and the available experimental data, in the context of radioisotope production. In general, a better reproduction of the cross section results in more accurate estimations for the production of the interested medical radionuclide, within optimal irradiation conditions. Advancement is achieved in this work by tuning the nuclear level density parameters of the most recent microscopic models implemented in the TALYS code. The procedure for a better cross section reproduction is applied to the specific reaction $^{\mathrm{nat}}\mathrm{V}(p,x)$ to produce ^{47}Sc, which is an important theranostic radionuclide. The study of ^{47}Sc production is extended also to its principal contaminant, ^{46}Sc, which plays an important role in assessing the quality of the production route.

1. – Introduction

Nuclear reaction modeling is very important in the field of medical radionuclide production to provide estimations of yields as well as of purities [1]. Sometimes the theoretical excitation functions obtained with nuclear reaction codes do not reproduce well the trend of the available experimental data for a given reaction. The models may overestimate/underestimate the cross sections or they reproduce an energy shifted cross section peak. In the context of radiopharmaceutical production, the selection of the optimal energy window is crucial and therefore we illustrate here a method to obtain the agreement between the theoretical curve and the data.

In this work we apply the approach to the reaction $^{\mathrm{nat}}\mathrm{V}(p,x)^{47}\mathrm{Sc}$ using the nuclear reaction code TALYS [2]. The reaction of interest has been already studied in the INFN

(*) E-mail: `alessandro.colombi@pv.infn.it`

project PASTA (Production with Accelerators of Sc-47 for Theranostic Applications) and preliminary results have been presented in ref. [3]. The clear disagreement between the theoretical curves and the data is an actual problem for the production of ^{47}Sc ($T_{1/2} = 3.35\,\mathrm{d}$).

Scandium is a theranostic candidate which can be used to perform SPECT (Single Photon Emission Computed Tomography) imaging ($E_\gamma = 159\,\mathrm{keV}$, $I_\gamma = 68.4\%$) and to treat small size tumors thanks to its β^- decay ($E_{\beta^-} = 162\,\mathrm{keV}$). Furthermore, ^{47}Sc can be combined with positron emitters ^{43}Sc or ^{44g}Sc to produce chemically identical radiopharmaceuticals for PET (Positron Emission Tomography) imaging and treatment [4].

In this paper we focus the attention on the production of ^{47}Sc and its main contaminant ^{46}Sc ($T_{1/2} = 83.79\,\mathrm{d}$) while a complete description of all the radionuclides produced in the process is discussed in ref. [5].

2. – Methods

In the description of a nuclear reaction, the nuclear level density plays an important role in the Hauser-Feshbach formalism for the compound nucleus description. Different models have been developed for the nuclear level density, and in particular six models are available within the TALYS nuclear reaction code [6]. Among them, three are phenomenological models based on constant temperature Fermi gas, back-shifted Fermi gas and the generalized superfluid descriptions, while the others are microscopic models. The latter have been developed more recently and are based on the microscopic Hartree-Fock Method (HFM), such as HF in two different formulations, HF-Bardeen-Cooper-Schrieffer (HF-BCS) [7] and HF-Bogoliubov (HFB) [8, 9].

The nuclear level density, obtained with any of the HFM approach, can be parameterized and rescaled with the following transformation:

$$\rho(E, J, \pi) = \exp(c\sqrt{E-p})\rho_{\mathrm{HFM}}(E-p, J, \pi), \tag{1}$$

where parameter c acts as a normalization factor, while parameter p acts as an energy shift caused by pairing or shell effects. Starting from the values of ρ_{HFM}, already tabulated inside the code, new level densities can be obtained by varying c and p, and this leads to new cross sections which are dependent from these two parameters.

Specifically, we have tuned these parameters considering the level density of the model based on the HF-BCS method (referred to as *ldmodel 4*). The ultimate goal was to find a good agreement between theoretical results and experimental data and this was achieved by a two-step procedure. Initially, a grid search on the values of c and p for all the nuclide of interest has allowed the selection of a set of values that has been subsequently used in the second step, represented by a global chi-square minimization using MINUIT [10], combining both gradient and simplex optimizations. The final set of values provided the optimized cross section curves which we refer to as "TALYS modified".

We have adopted the JLM (Jeukenne-Lejeune-Mahaux) semi-microscopic optical potential model [11], and the exciton model with transition rates calculated numerically for the preequilibrium processes (which is the default preequilibrium option in the TALYS code).

We cannot guarantee that the solution we found is unique. However, it provides a much better description of the cross sections consistent with the data.

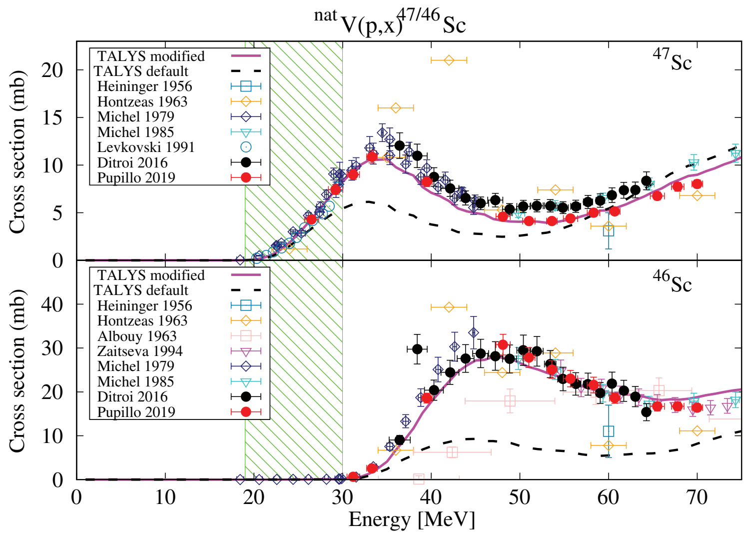

Fig. 1. – Comparison of the TALYS modified curve (solid lines), TALYS default (dashed lines) and the experimental data for the reactions ${}^{\mathrm{nat}}\mathrm{V}(p,x){}^{47}\mathrm{Sc}$ (top) and ${}^{\mathrm{nat}}\mathrm{V}(p,x){}^{46}\mathrm{Sc}$ (bottom). The shaded area represents the optimal energy window (19–30 MeV) for the radionuclide production. See [5] for a comprehensive discussion and for the references of the experimental data.

3. – Results

Figure 1 compares the production of ${}^{\mathrm{nat}}\mathrm{V}(p,x){}^{47/46}\mathrm{Sc}$ with TALYS modified, TALYS default (*i.e.*, the default result of the code) and the available experimental data, whose references can be found in ref. [5]. The shaded area corresponds to the optimal energy window (19–30 MeV) for the maximum production of ${}^{47}\mathrm{Sc}$ and minimum co-production of ${}^{46}\mathrm{Sc}$. The new theoretical cross sections adequately describe the data and reproduce well the trends both at low and high energies. This allows a more accurate calculation

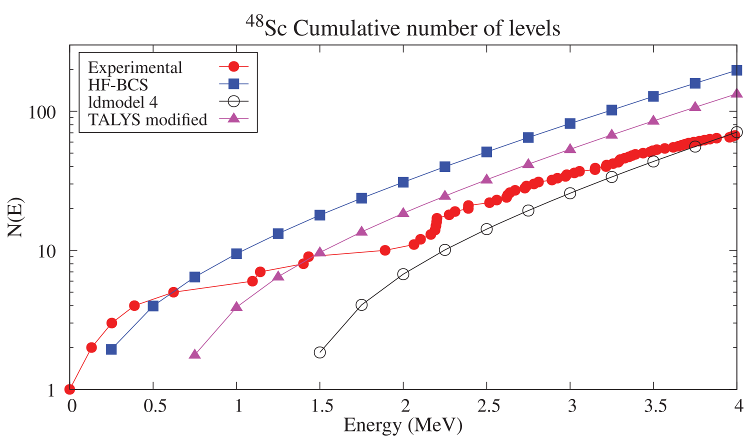

Fig. 2. – Comparison between the theoretical level cumulatives and the experimental one for the compound nucleus ${}^{48}\mathrm{Sc}$. A study involving an extensive set of produced nuclides can be found in ref. [5].

of the production yield for ^{47}Sc and ^{46}Sc, as shown in ref. [5], where two irradiation conditions have been assumed, one with 24 h and the other with 80 h irradiation time, both with 1 μA beam current.

Figure 2 compares, for ^{48}Sc, the level cumulatives derived from the current adjustment of the c and p parameters (TALYS modified), with the original HF-BCS model and with the so-called *ldmodel* 4, corresponding to a specific global tuning of the density levels, with the c and p parameters tabulated in the code. The comparison with the experimental cumulative, shown by the red points, suggests that our cumulative obtained with the parameter adjustment is not distant from the measurements, even if we started from cross section optimization, rather than from the improvement of the nuclear level densities.

4. – Conclusions

Radionuclide ^{47}Sc represents an emerging theranostic candidate and different production routes are under investigation to find the most suitable one. In this work we have focused on the reaction $^{\rm nat}$V$(p,x)^{47}$Sc and an advancement of the nuclear reaction modeling has been obtained. The approach is based on tuning the nuclear level density to better describe the cross sections for both ^{47}Sc and ^{46}Sc. This has important consequences in the evaluation of yield and purity for an optimal production of the relevant radionuclide, also in view of its possible medical applications. The procedure is general and flexible and can be implemented also for the production of other innovative medical radioisotopes.

* * *

This work represents an extension of the study conducted in the PASTA project in the framework of LARAMED (LAboratory of RAdioisotopes for MEDicine) program at INFN-Legnaro National Laboratories (INFN-LNL).

REFERENCES

[1] Canton L. *et al.*, *Eur. Phys. J. Plus*, **135** (2020) 770.
[2] Goriely S. *et al.*, *Astron. Astrophys.*, **487** (2008) 767.
[3] Pupillo G. *et al.*, *J. Radioanal. Nucl. Chem.*, **322** (2019) 1711.
[4] Müller C. *et al.*, *Br. J. Radiol.*, **91** (2018) 20180074.
[5] Barbaro F. *et al.*, *Phys. Rev. C*, **104** (2021) 044619.
[6] Koning A. J. *et al.*, *Proceedings of the International Conference on Nuclear Data for Science and Technology, April 22–27, 2007, Nice (France)*, edited by Bersillon O., Gunsing F., Bauge E., Jacqmin R. and Leray S. (EDP Sciences) 2008, pp. 211–214.
[7] Minato F., *J. Nucl. Sci. Technol.*, **49** (2011) 984.
[8] Hilaire S. *et al.*, *Nucl. Phys. A*, **779** (2006) 63.
[9] Hilaire S. *et al.*, *Phys. Rev. C*, **86** (2012) 064317.
[10] James F. and Roos M., *Comput. Phys. Commun.*, **10** (1975) 343.
[11] Jeukenne J. P. *et al.*, *Phys. Rev. C*, **15** (1977) 10.