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EVALUATION OF THE YIELDS OF Ga-67 PRODUCED ON CYCLOTRON C18

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Abstract - Theoretical calculations of the excitation functions for the reaction $^{67}Zn(p,n)^{67}Ga$ on natural and enriched zinc targets in the energy range from threshold up to 18 MeV are carried out using nuclear reaction models from program packages EMPIRE-3 and TALYS 1.4. The results are compared with those available from experimental data. The yields of gallium isotopes produced in the reaction $^{67}Zn(p,n)^{67}Ga$ on the cyclotron C18, IBA Belgium with proton energy 18 MeV are evaluated. Our calculations indicate that the yield of ^{67}Ga production in case of natural and enriched zinc targets are expected to be 23.6 MBq/μA·h and 139 MBq/μA·h, respectively.

1. Introduction

The study of nuclear reactions with intermediate or high energy protons is very important because it provides a basis for a wide range of technical applications. Particularly, using the reactions induced by intermediate energy protons, it is directly possible to produce radionuclides used in medicine and industry. In recent decades, the widespread use of diagnostic and therapeutic radioisotopes takes place. Depending of the type of radiation, the diagnostic isotopes are classified into two groups: β^+-emitters (^{13}N, ^{15}O, ^{18}F, ^{62}Cu, ^{68}Ga, etc.) used in Positron Emission Tomography (PET), and γ–emitters (^{67}Ga, ^{99m}Tc, ^{123}I, etc.) used in Single Photon Emission Computed Tomography (SPECT).

In the radioisotope production procedure, the nuclear reaction data are mainly needed for respective optimization of production rates. This process involves a selection of the projectile energy range that will maximize the yield of produced isotopes and minimize the radioactive impurities. The total cross section of such a production is also important in accelerator technology from the point of view of radiation protection safety. The calculations based on standard models for nuclear reactions can be helpful for determining the accuracy of various parameters for these models as well those evaluated from experimental measurements [1].

The ^{67}Ga isotope is one of the main radionuclides produced on cyclotrons and used in clinical oncology for diagnostics of benign and malignant tumors.

^{67}Ga commonly is used in SPECT investigations as a trivalent citrate compound for nuclear medicine imaging, and is a valuable agent in the detection and localization of certain neoplasms and inflammatory lesions. It is well known that when ^{67}Ga is in the citrate form it is concentrated in many types of tumours, as well as non-malignant lesions. Although it is not a tumour-specific agent, it is used extensively for the localization of a variety of human malignant tumours and, due to its widespread application as a diagnostic tool in nuclear medicine. ^{67}Ga is one of the most widely employed cyclotron-produced radiopharmaceuticals.

Ga-67 with the half-life $T_{1/2}$ = 78.3 h decays to stable ^{67}Zn by electron capture. The scheme of Ga-67 decay and its energetic characteristics are taken from Nudat 2.5 [2] and are presented in Fig. 1 and Table 1, respectively.

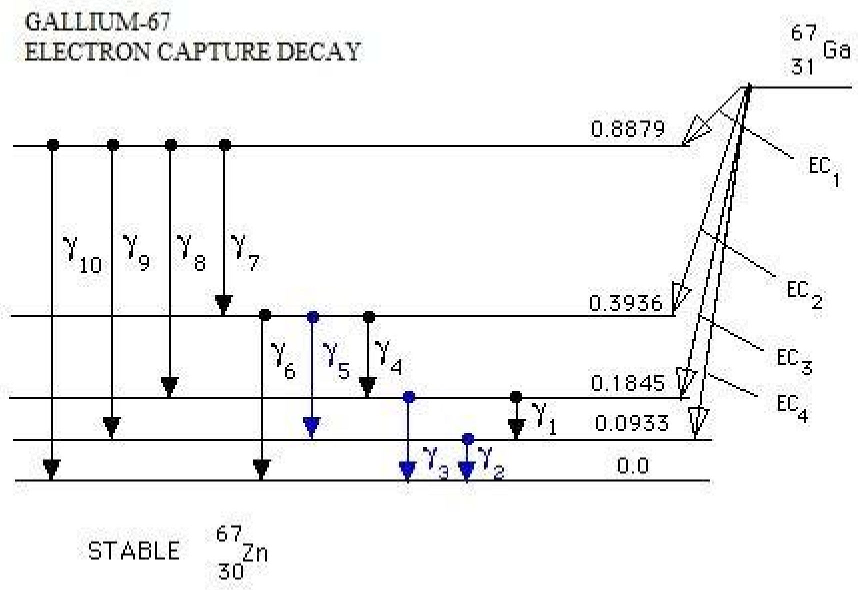

Fig. 1. The decay scheme of Ga-67.

Table 1. Decay data of Ga-67.

Nuclide	Half-life	Decay mode, (%)	E_γ, keV	I_γ, %
^{67}Ga	78.3 h	EC (100)	93.31 184.58 300.2 393.5	38.81 21.41 16.64 4.56

2. The nuclear reaction cross-section calculations

One of the main parameters at production of radioisotopes under proton beam is the specific activity which is the isotopes yield normalized to unit of beam current and unit of irradiation time – Bq/μA·h or Ci/μA·h.

In the case of thick target at calculation of specific activity one needs to consider the decrease of protons' energy as they pass through the target, and therefore integrate the cross sections over the entire energy range.

The saturated yield Y per unit current (Bq/μA) can be calculated using the following formula [3]:

$$Y = 6.24 \, x \, 10^{12} \, x \frac{N_A}{M} \int_{Eout}^{Ein} \frac{\sigma(E)}{S(E)} dE \tag{1}$$

where 6.24×10^{12} is the number of protons per second per μA, N_A is the Avogadro number, M is the target atomic mass, $\sigma(E)$ is the reaction cross section (excitation function) as a function of energy expressed in mb and $S(E)$ is the target material stopping power expressed in units MeV cm^2 g^{-1}.

The criterion in choosing of a nuclear reaction for the production of isotopes for medicine is a high specific activity. Therefore, we consider the production of Ga-67 isotope on targets of natural and enriched zinc from the point of view of the specific activity

Ga-67 is commonly produced by using enriched ^{68}Zn target through the nuclear reaction ^{68}Zn(p,2n)^{67}Ga in the proton energy range E_p = 20-40 MeV [4-8].

We consider the possibility of Ga-67 isotope production through the reaction ^{67}Zn(p,n)^{67}Ga at proton energy 18 MeV of the cyclotron C18. For this purpose a study of the excitation function and the yield of the reaction ^{67}Zn(p,n)^{67}Ga on the targets made from natural and enriched zinc in the energy range of protons from the reaction threshold up to 18 MeV was performed.

In radioisotope production programmer, nuclear reaction data mainly needed for optimization of production routes. This process involves a selection of the projectile energy range that will maximize the yield of the product and minimize that of the radioactive impurities.

The calculation of the yields of Ga-67 isotope in the reaction $^{67}Zn(p,n)^{67}Ga$ allows to evaluate the efficiency of the production for both the natural and enriched targets.

Naturally occurring zinc is composed of 5 stable isotopes ^{64}Zn, ^{66}Zn, ^{67}Zn, ^{68}Zn, and ^{70}Zn.

In Table 2 the natural abundance of each isotope of Zn, all channels of reactions on each isotope of Zn and the thresholds for these reactions are listed. As seen from Table 2, due to the reactions threshold, the isotope Ga-67 could be produced on the proton beam of cyclotron C18 only on the isotopes ^{67}Zn and ^{68}Zn.

In addition is also can be seen from Table 2, that accompanying unwanted products of the contributing reactions have short lifetimes compared with the lifetime of the Ga-67. Consequently, their impurity in the final product is insignificant.

Table 2. Nuclear data of contributing reactions at natural zinc target.

Natural target	Abundance, %	Contributing reactions	Threshold, MeV	Half-life
^{64}Zn	48.63	$^{64}Zn(p,n)^{64}Ga$ $^{64}Zn(p,2n)^{63}Ga$	8. 18.6	2.9 min 32.4 s
^{66}Zn	27.90	$^{66}Zn(p,n)^{66}Ga$ $^{66}Zn(p,2n)^{65}Ga$	6. 15.3	9.49 h 15.2 min
^{67}Zn	4.10	$^{67}Zn(p,n)\,^{67}Ga$ $^{67}Zn(p,2n)\,^{66}Ga$	1.88 13.	78.3 h 9. 49 h
^{68}Zn	18.75	$^{68}Zn(p,n)^{68}Ga$ $^{68}Zn(p,2n)^{67}Ga$	3.75 12.15	67.71 min 78.3 h
^{70}Zn	0.62	$^{70}Zn(p,n)^{70}Ga$ $^{70}Zn(p,2n)^{69}Ga$ $^{70}Zn(p,3n)^{67}Ga$	1.45 9.2 33.9	21.14 min Stable 78.3 h

The calculation of the cross section for the reaction $^{nat}Zn(p,xn)^{67}Ga$ was performed by program TALYS 1.4 [9] and EMPIRE-3.1 [10]. In Fig. 2 the results of our calculation in which the natural abundance of each isotope and cross section of the each channel of the reaction was taken into account together with the experimental data from Refs. [11] and [12] are shown. There is good agreement between the results of calculations for both models TALYS 1.4 and EMPIRE-3.1, and also with the given experimental data.

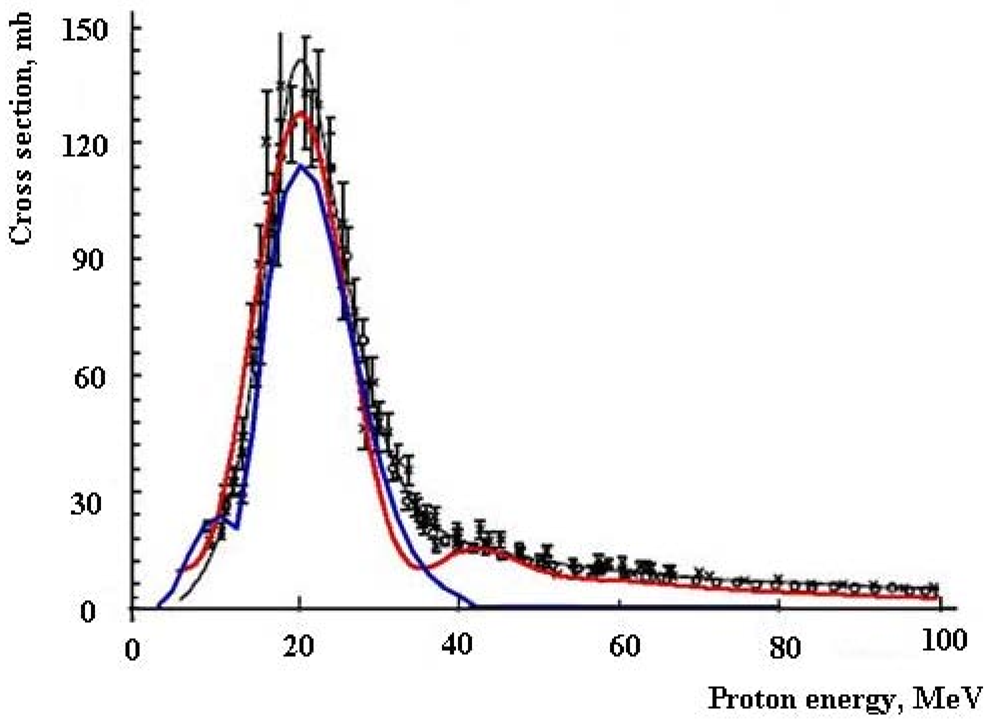

Fig. 2. The cross section of the reaction $^{nat}Zn(p,xn)^{67}Ga$ calculated by TALYS 1.4 [9] (red curve) and EMPIRE-3.1 [10] (blue curve). The open circles are the data of Ref. [11] and the crosses are the data of Ref. [12]. The black curve is a polynomial fit to the data.

From Fig. 2 is seen that in case of target from natural zinc the best energy range for production of isotope Ga-67 on the cyclotron C18 is E_p = 13–17 MeV and maximum cross section in this range is about 100 mb.

For this energy range, the numerical calculations of the yields of Ga-67 production were performed by formula (1). The value $\sigma(E)$ of the cross sections was taken from the program TALYS 1.4 [9] and the value of stopping power $S(E)$ was obtained from SRIM [13]. The results of these calculations are shown in Fig. 3.

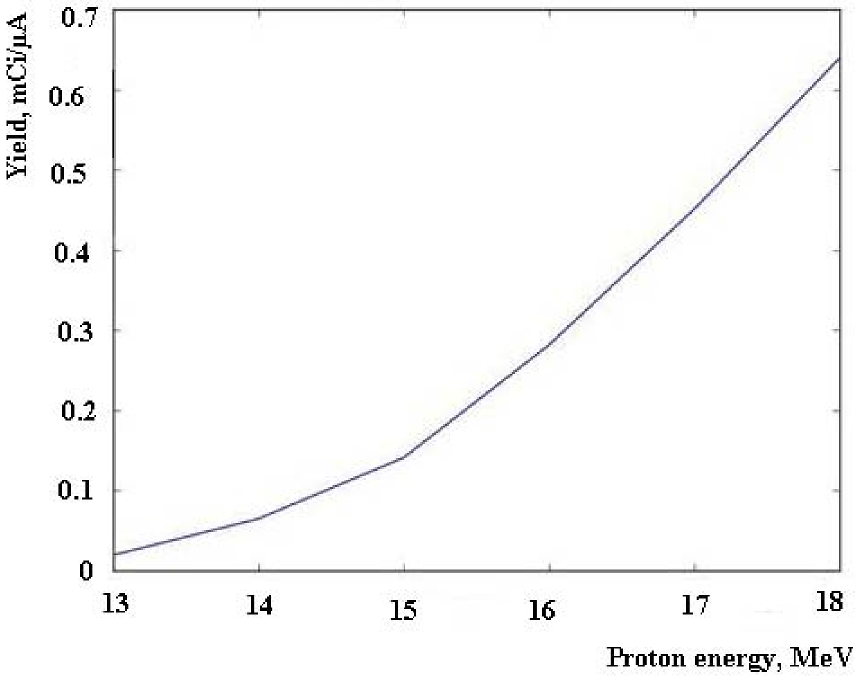

Fig. 3. The dependence of Ga-67 production yield vs. proton energy on the natural zinc target.

The calculations showed that the specific activity in case of target from natural zinc is 0.638 mCi/µA*h (23.6 MBq/µA*h). At irradiation during 2 hours and proton beam with energy 18 MeV and current 30 µA the yield is 38 mCi. Since the dose for one patient survey is 2-5 mCi the obtained yield is enough for inspection of about 8 patients.

From a commercial point of view, this method of production of Ga-67 isotope from natural target from Zn is ineffective. Therefore, the reaction for the production of ^{67}Ga isotope on enriched target of ^{67}Zn was studied.

The excitation function of the reaction ^{67}Zn(p,n)^{67}Ga for enriched target was calculated by means of programs TALYS 1.4 [9] and EMPIRE-3.1 [10].

The results of the calculations together with set of experimental data from IAEA Technical Reports [14] are shown in Fig. 4. It is seen from Fig. 4 that in the range of low energy the results of calculations by programs TALYS 1.4 [9] and EMPIRE-3.1 [10] are in good agreement with the set of experimental data [14]. TALYS 1.4 and EMPIRE-3.1 codes predicted the maximum cross-section to be respectively 587.3 mb and 575.6 mb at the same energy 10 MeV. The effective energy range for Ga-67 production on the enriched zinc target is E_p = 5-14 MeV.

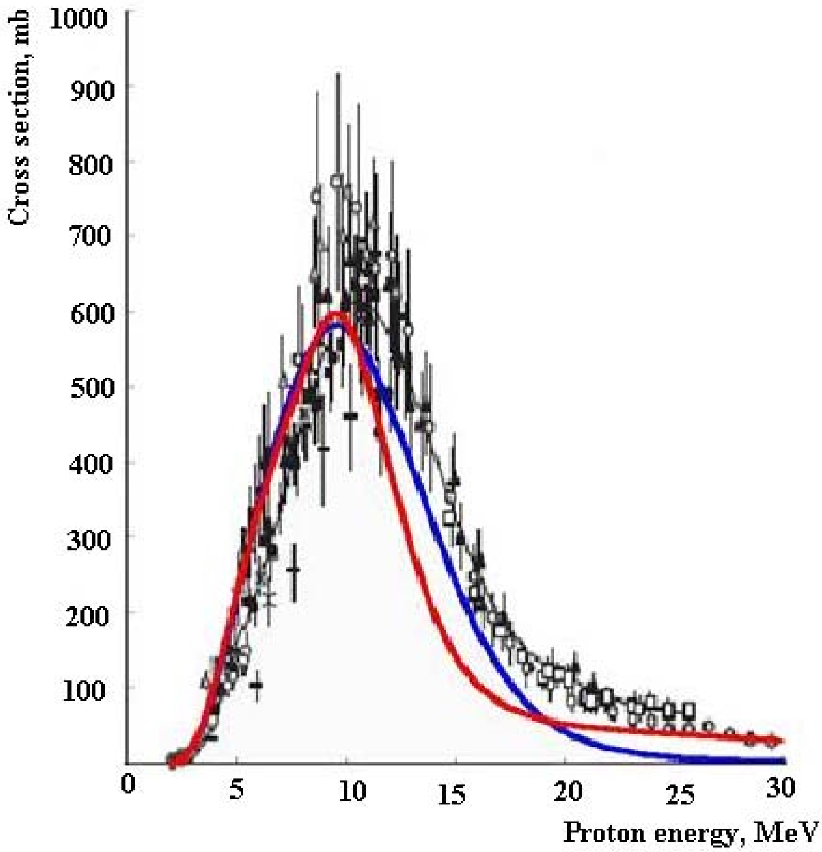

Fig. 4. The comparison of the cross sections of reaction $^{67}Zn(p,n)^{67}Ga$ calculated by programs TALYS 1.4 (red curve) and EMPIRE-3.1 (blue curve). The set of experimental data is given from Ref. [14].

The specific activity for Ga-67 production was calculated on the base of formula (1) using data of programs TALYS 1.4 [9] and SRIM [13]. In case of enriched zinc target the special activity is found to be 3.772 mCi/μA·h (139 MBq/μA·h), which agrees with the value 3 mCi/μA·h from Ref. [4].

The results of calculation of the yields for Ga-67 isotopes on enriched zinc target performed in the proton energy range with the maximum cross section are shown in Fig. 5.

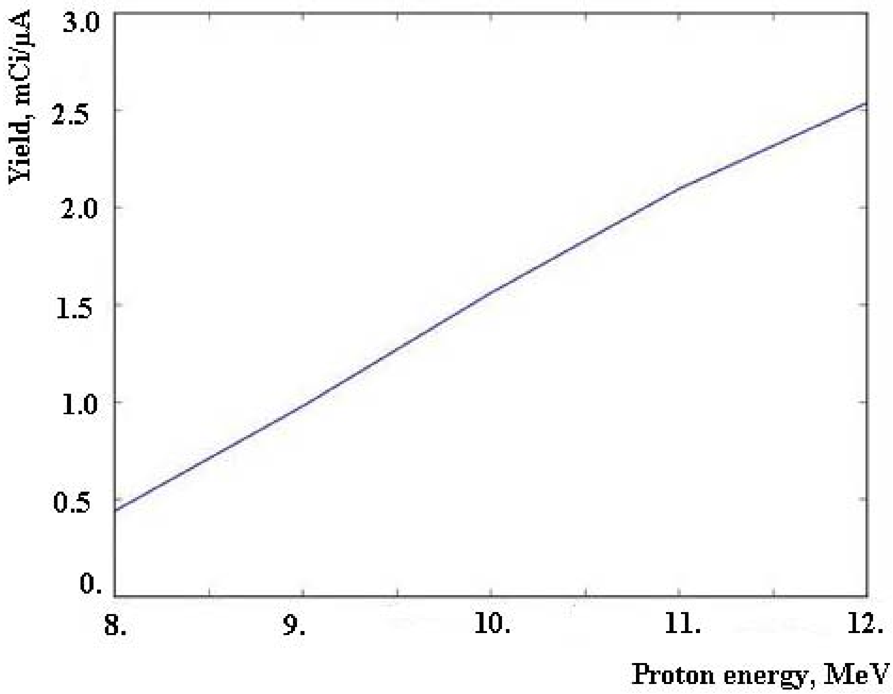

Fig. 5. The dependence of Ga-67 activity via incident proton energy on the enriched zinc target.

The yield of isotope Ga-67 in case of irradiation of the enriched zinc target during 2 h by the proton beam with current 30 μA is 226 mCi. This amount of Ga-67 isotope is sufficient for diagnostic survey of more than 45 patients.

Taking into account the long half-life (78.3 hours) and the amount of the obtained isotope Ga-67 could be considered its delivery in the clinics of the region.

3. Conclusions

New calculations on the excitation function of reactions $^{nat}Zn(p,xn)Ga^{67}$ and $^{67}Zn(p,n)Ga^{67}$ have been carried out using nuclear reaction models from TALYS 1.4 and EMPIRE-3.1. The yield of the reactions for the production of isotope Ga-67 on the natural and enriched targets of zinc was evaluated using program packages TALYS 1.4 and SRIM. It is shown that on the proton beam of the commercial cyclotron C18 with current 30 μA and proton energy 18 MeV using enriched target

Zn-67 it is possible to produce Ga-67 isotope in amounts sufficient to meet the needs of the clinics of the region.

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