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COPPER-64 ISOTOPE PRODUCTION THROUGH THE CYCLOTRON PROTON IRRADIATION OF THE NATURAL-NICKEL TARGET

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A method of calculation has been developed as well numerical simulation have been performed for a production process of ^{64}Cu isotope by the ^{64}Ni (p, n)^{64}Cu nuclear reaction. The required radionuclide applicable in the nuclear medicine is produced through irradiating a cyclotron target of natural nickel with a proton beam. The process conditions were dictated by the capabilities of the cyclotron; the initial kinetic energy of 17 MeV (at a current of 10 μA) was fed into computation. As a result, dependencies of the ^{64}Cu isotope production on the target thickness and on the irradiation time were obtained. The target depth of proton penetration was investigated, and it was established where the peak of radionuclide concentration was produced. An analysis of the obtained data allowed us to find the optimal thickness of the nickel target, which is 0.54 mm.

Keywords: copper-64, natural nickel, yield calculation, target thickness, proton beam

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ПРОИЗВОДСТВО ИЗОТОПА МЕДИ-64 ПУТЕМ ОБЛУЧЕНИЯ ЦИКЛОТРОННОЙ МИШЕНИ ИЗ ПРИРОДНОГО НИКЕЛЯ ПУЧКОМ ПРОТОНОВ

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Разработана методика расчета и выполнено численное моделирование процесса получения изотопа меди-64 по ядерной реакции ^{64}Ni (p, n)^{64}Cu. Требуемый радионуклид, применяемый в ядерной медицине, производится путем облучения мишени из природного никеля пучком протонов, получаемым на циклотроне. Условия проведения процесса диктовались возможностями циклотрона. В расчеты закладывалась начальная кинетическая энергия протонов 17 МэВ (ток равен 10 мкА). В результате получены зависимости наработки изотопа меди-64 от толщины мишени и от времени облучения, изучена глубина проникновения протонов в мишень, установлено, где концентрация ядер наработанного радионуклида максимальна. Анализ полученных данных позволил определить оптимальную толщину никелевой мишени, она составила 0,54 мм.

Ключевые слова: медь-64, природный никель, расчет выхода, толщина мишени, пучок протонов

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Introduction

As known, radiology and radiation therapy are based on the use of various radiopharmaceuticals containing radioactive isotopes. Among the radionuclides belonging to different elements, Cu-64 isotope is unique due it's capability to emit β^+-, β^- particles (their energies equal 0.65 and 0.57 MeV, and their yield values are 17.5 and 38.5%, respectively) and Auger electrons as a result of its radioactive decay. Therefore, this isotope is applicable in both positron emission tomography (PET), as well as in theranostics (theranostics focuses on producing pharmaceuticals which can be simultaneously applied as a means of early diagnosis and as a therapeutic agent) [1, 2]. The isotope's considerable advantages over others lie not only in its chemical properties, but also in its long half-life (12.7 h) allowing simplification of production, transportation and application of Cu-64 labeled radiopharmaceuticals in comparison with those in common use nowadays.

The possibility of reduction of cupric Cu^{2+} to cuprous Cu^+ is used in molecular imaging and targeted radiotherapy of hypoxic tissues, including tumors [3, 4].

Clinical trials of ^{64}Cu based pharmaceuticals showed copper uptake only in hypoxic heart and brain cells. Peptides and antibodies radiolabeled with this isotope can also be applied in medical radiology [5–8].

A comparison of ^{64}Cu labeled radiopharmaceuticals and those based on ^{111}In-Octreoscan which is currently in use showed the advantage of the former as a positron-emitter for it allowed imaging even for some unpredicted metastatic growth [9]. Clinical testing of the ^{64}Cu-TETA-mab1A3 conjugate (applied in colorectal cancer detection) demonstrated the advantage of PET with ^{64}Cu over the similarly ^{111}In labeled conjugate [10–12].

The ^{64}Cu radionuclide can be produced in reactors by means of capturing either $^{63}\mathrm{Cu}(n, \gamma)^{64}\mathrm{Cu}$ thermal neutrons or $^{64}\mathrm{Zn}\ (n, p)^{64}\mathrm{Cu}$ fast neutrons (n, p are neutrons and protons, γ are gamma rays). The isotope can be produced in a cyclotron by the $^{64}\mathrm{Ni}\ (p, n)^{64}\mathrm{Cu}$ nuclear reaction [13].

However, the ^{64}Cu production yield in reactors is low, while their radionuclidic purity is often unsatisfactory [13]. Therefore, the use of cyclotrons in the ^{64}Cu production becomes necessary since they support charged particles induced reactions. Both natural and enriched (by 99.6%) nickel can serve as a target for the ^{64}Cu production in cyclotrons.

The purpose of this article is to develop a method of calculation, and the corresponding algorithm and program, as well as to conduct a numerical simulation of the ^{64}Cu production by means of irradiating a natural nickel target with a proton beam.

Numerical parameters of the model were determined by the characteristics of the MGC-20 cyclotron located in Peter the Great St. Petersburg Polytechnic University: proton energy of 17 MeV at a current of 10 μA. The numerical simulation allowed us to determine the optimal target thickness for the maximum yield of the desired isotope.

Method of calculation

As it was noted before, for the ^{64}Cu production, we used a proton beam with the initial energy of 17 MeV (current of 10 μA). A natural ^{64}Ni isotopic mixture (the percentage of ^{64}Ni isotope in the natural nickel amounts to 0.926%) serves as the target.

The computations allow for proton energy losses due to excitation and ionization resulting from the passage through of the target substance [14]:

$$\left\langle -\frac{dE}{dx} \right\rangle = \frac{Kz^2 Z\rho}{A\beta^2}\left[\frac{1}{2}\ln\left(\frac{2m_e c^2\beta^2\gamma^2 W_{\max}}{I^2}\right) - \beta^2 - \frac{\delta(\beta_\gamma)}{2}\right], \quad (1)$$

where $-dE/dx$, MeV/cm, are specific ionization losses (x is the depth of proton penetration); z, Z are charge numbers of the projectile particle and the target respectively; A, g /mol, is atomic mass; ρ, g/cm^3,is the target density; m_e, g, electron mass; c, cm/s, – the speed of light; β is a ratio of the projectile particle velocity to the speed of light ($\beta = v/c$); γ is the Lorentz factor; $W_{\max}$, MeV, is maximum energy transfer in a unit collision; I, eV, is mean ionization potential; δ(βγ) is a correction factor allowing for the influence of the target density on the ionization potential.

The K coefficient is calculated by the formula

$$K = 4\pi N_A r_e^2 m_e c^2 = 0.307\,\text{MeV/mol},$$

where N_A is the Avogadro constant; r_e, cm, is the classical electron radius.

The maximum energy transfer is expressed as

$$W_{\max} = \frac{2m_e c^2\beta^2\gamma^2}{\left[1 + \frac{2\gamma m_e}{M} + \left(\frac{m_e}{M}\right)^2\right]},$$

where M, g, is the mass of the projectile particle.

The Ni mean ionization potential is equal to (just as for other elements) $I = 311 \pm 10$ eV [15].

We should note that in a nonrelativistic case $\beta^2 << 1$; furthermore, a proton ($z = 1$, $M >> m_e$) is the projectile particle, the energy loss occurs at all Ni isotope electron shells in natural nickel, i.e., $\rho = \rho_{\text{Ni}} = 8.908$ g/cm^3; $A = \langle A \rangle = 58.6934$ g/mol. Then, Eq. (1) is simplified:

$$\left\langle -\frac{dE}{dx} \right\rangle = \left(\frac{KZ\rho}{A\beta^2}\right) \ln\left[\frac{2m_e c^2\beta^2}{I}\right]. \qquad (2)$$

The solution to Eq. (2) gives a dependence $E(x)$ of the mean proton energy on the depth of proton penetration.

The Cu^{64} isotope is produced by means of the Ni^{64} (p, n) Cu^{64} reaction. Energy dependence of this reaction cross section $\sigma(E)$ was measured in many experiments. This article used the results of the combined data presented in paper [16].

Using the solution to Eq. (2), let us determine the dependence of the reaction cross section on the depth of proton penetration $\sigma(x)$. The Cu^{64} production at different depths of target penetration is then determined by the formula:

$$\frac{dN_{\text{Cu64}}}{dx} = \left(\frac{Jn_{\text{Ni64}}}{\lambda e}\right) \times \\ \times\left(1 - \exp\left(-t\lambda_{rad}\right)\right)\sigma(x), \qquad (3)$$

where N_{Cu64} is the number of ^{64}Cu isotope nuclides; J, A, is the cyclotron current; n_{Ni64} is ^{64}Ni nuclides concentration in the natural nickel; λ is the ^{64}Cu decay constant; e, C, is the electron charge; t_{rad}, s, is the time of the target irradiation.

By integrating Eq. (3) from zero to the target thickness τ, we obtain a distribution of the ^{64}Cu production with respect to the isotope penetration:

$$N_{\text{Cu64}}\left(\tau, t_{rad}\right) = \int_0^\tau dx \left\{\frac{dN_{\text{Cu64}}}{dx}\right\}, \qquad (4)$$

In this case, we can disregard a decrease in intensity of the proton beam with the increasing of the target depth, as well other processes resulting in proton loss. The obtained formula does not consider the cooling time of the target which can be allowed for by multiplying it by an exponential factor of the $\exp(-\lambda t_{cool})$ form. It can be seen from Eq. (3) that at $t_{rad} \approx 3/\lambda$, there is no point in any further irradiation as the accumulation curve reaches a plateau.

Such a behavior of the dependence is due to a gradual appearance of commensurability of the isotope production and its decay rates. However, the value of 1/λ for ^{64}Cu equals to 18.3 h, which significantly exceeds the real time of irradiation. At $t_{rad} << 1/\lambda$ the factor

$$\left[1 - \exp\left(-\lambda t_{rad}\right)\right]/\lambda \approx t_{rad},$$

i.e., the production is proportional to time.

Results of the used method and discussion

The solution to Eq. (2) is presented in Fig. 1,*a* for the protons of the initial kinetic en-

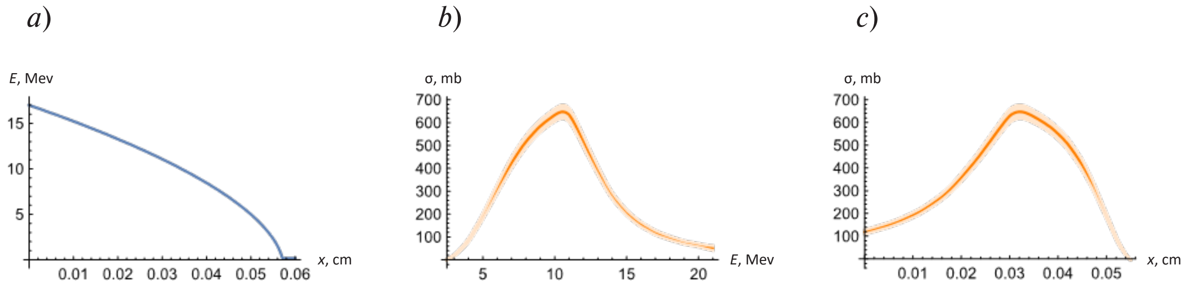

Fig. 1. Proton energy distribution throughout the depth of the natural nickel target (solution to Eq. (2)) (*a*) and dependence of the ^{64}Ni(p, n)^{64}Cu reaction cross section on the proton energy (*b*) and on the target depth (*c*). Initial kinetic proton energy is 17 MeV; in Fig. *b*) and *c*), the lines show the "cross section" curves behavior, while the bands correspond to the cross section error [16]

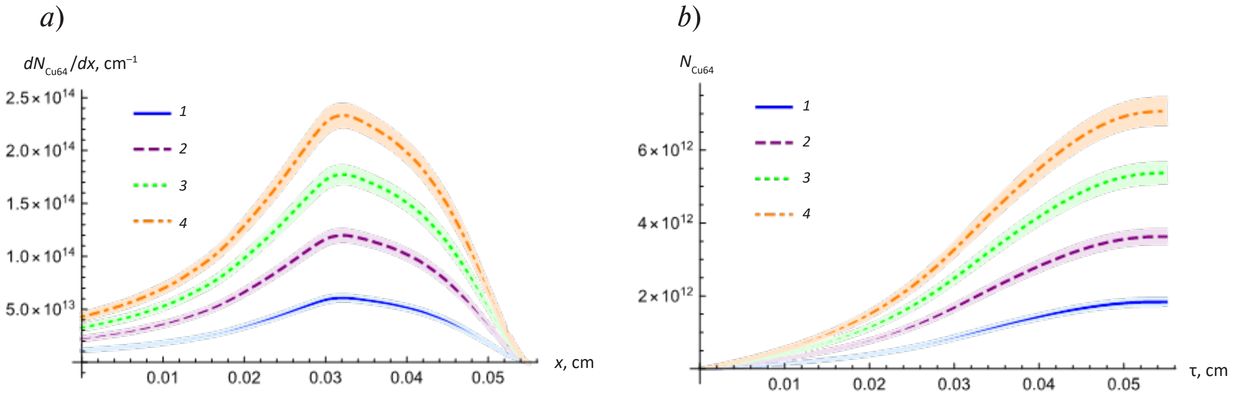

Fig. 2. Ionization loss distribution of ^{64}Cu nuclides throughout the depth of target penetration (*a*) and dependencies of the number of the produced nuclides on the target thickness (*b*) for different irradiation intervals, h: 0.5 (*1*), 1.0 (*2*), 1.5 (*3*), 2.0 (*4*). The lines show the dependencies curves, while the bands correspond to their error (connected with the reaction cross section errors)

ergy of 17 MeV and the natural nickel target. We can see that the protons lose all of their energy at the 0.56 mm target depth.

Fig. 1,*b* shows the result of interpolating the reaction cross section (3), obtained in paper [16]. By processing this dependence and using the solution of Eq. (2) we found a dependence of the cross section on the target depth (Fig. 1,*c*). It is clear that the cross section is maximal at the proton energy of approximately 10 MeV, which is reached at the depth of around 0.32 mm. At the same depth, we also observe the peak concentration of the produced ^{64}Cu radionuclides.

Fig. 2,*a* shows the computational results of ^{64}Cu number distribution throughout the depth for various irradiation intervals (we took a range from 0.5 to 2 h, see Eq. (3)). Fig. 2,*b* presents the results of integrating dependence (4) with respect to the target thickness τ for the same irradiation intervals.

Fig. 3 shows the results of computing the number of the produced ^{64}Cu nuclides depending on two irradiation intervals (5 and 50 h) for four values of the target thickness. We can see that for the 5 h irradiation interval the number of the produced nuclides grows in a linear manner and with a fair degree of accuracy, while for the longer period of time the growth levels off after the saturation point.

An analysis of the obtained data allows us to conclude that the peak ^{64}Cu production is achieved at the target thickness of 0.545 ± 0.006 mm. The error is primarily due to the equivocation arising

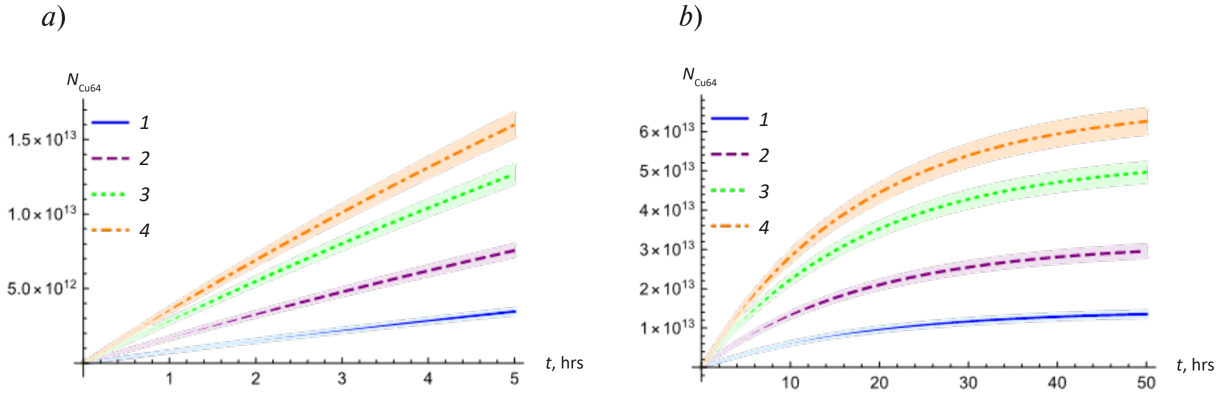

Fig. 3. Dynamics of the number of accumulated ^{64}Cu nuclides in 5 h (*a*) and 50 h (*b*) of irradiating the target of different thickness τ, mm: 0.2 (*1*), 0.3 (*2*), 0.4 (*3*), 0.5 (*4*). The lines show the dependencies curves, while the bands correspond to their error (connected with the reaction cross section errors)

from measuring the cross section $\sigma(E)$ [16] at the reaction energy threshold of 2.5 MeV (we allowed for the equivocation from using the simplified Eq. (2) instead of the complete Eq. (1)). Any further increase of the target thickness should not lead to an increase in the production as at greater thickness values the mean proton energy falls below the reaction energy threshold. The peak ^{64}Cu nuclide concentration lies at the target depth from 0.20 to 0.49 mm.

Thus, we can consider the identified target thickness value, i.e. 0.545 ± 0.006 mm, optimal for the maximum production number of the ^{64}Cu isotope.

Conclusion

This article developed a design procedure and numerical simulation of a production process for ^{64}Cu isotope important for application in the nuclear medicine. Th process of the ^{64}Cu isotope through irradiating a target of natural nickel with a proton beam obtained in a cyclotron. The initial kinetic energy of protons was 17 MeV at a current of 10 μA.

By means of computations we obtained dependencies of the ^{64}Cu isotope production on the target thickness and on the time of irradiation. We identified the depth of target penetration at which the nuclide concentration of the produced isotope reaches its peak.

As a result of the conducted simulation of the process, we found the optimal target thickness, which is 0.54 mm, to produce the maximum number of the ^{64}Cu isotope. The obtained result is essential for diagnostics and therapy of various diseases applied in nuclear medicine.

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