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OPTIMIZATION OF THE COPPER-64 PRODUCTION FROM NATURAL NICKEL TARGET AT A CYCLOTRON

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The paper is devoted to the problem of the copper-64 isotope production engineering that is important for application in the nuclear medicine. The production is carried out by proton irradiation of a nickel target (a natural mixture of isotopes). For this purpose, the energy dependence of the protons-nickel target interaction cross-sections, protons with initial kinetic energies of 10–15 MeV in this case, has been analyzed. Besides, the half-lives of the resulting isotopes were considered. Based on the analysis, the optimal conditions (the proton beam energy and the waiting time after irradiation) for obtaining the ^{64}Cu isotope from natural nickel were found. It was established that under conditions close to ideal, it could be expected that ^{64}Cu radionuclide purity would be very high and reach at least 99 %. Ideal conditions mean complete separation of nickel and cobalt isotopes from the required copper one.

Keywords: copper-64 isotope, cyclotron irradiation, nickel target, radionuclide purity, yield calculation

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ОПТИМИЗАЦИЯ ПОЛУЧЕНИЯ ИЗОТОПА МЕДИ-64 ИЗ ПРИРОДНОГО НИКЕЛЯ НА ЦИКЛОТРОНЕ

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Статья посвящена проблеме разработки технологии получения изотопа ^{64}Cu, важного для применения в ядерной медицине, путем циклотронного облучения протонами мишени из природного никеля. С этой целью проанализирована энергетическая зависимость сечений взаимодействия протонов, обладающих начальной кинетической энергией 10 – 15 МэВ, с мишенью из никеля (природная смесь изотопов). Кроме того, рассмотрены величины периодов полураспада образующихся изотопов. На основе проведенного анализа определены оптимальные условия получения изотопа ^{64}Cu (энергия пучка протонов и время выдержки после облучения) из природного никеля. Установлено, что в условиях, близких к идеальным (случай полного отделения изотопов никеля и кобальта от требуемого изотопа меди) можно ожидать, что радионуклидная чистота изотопа ^{64}Cu будет очень высокой и достигать не менее 99 %.

Ключевые слова: изотоп меди-64, циклотронное облучение, никелевая мишень, радионуклидная чистота, расчет выхода

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Introduction

It is well known that the ^{64}Cu isotope undergoes radioactive transformation as a result of three processes: positron and electron decay, and electron capture. This isotope emits β^+, β^- particles (their energies are 0.65 and 0.57 MeV, respectively, and the yields are 17.6 and 38.5%) with a half-life of 12.7 h. It plays an important role among bifunctional radioisotopes for both positron emission tomography (PET) and radionuclide therapy. The half-life of ^{64}Cu allows producing this isotope at regional or national cyclotron facilities, distributing it to local nuclear medicine departments with a loss of no more than one (approximately) half-life [1, 2].

Furthermore, the half-life of the ^{64}Cu isotope is compatible with the time scales required for administering a radiopharmaceutical (containing a molecular carrier: peptides, antibodies, nanoparticles, etc.), so that it is subsequently distributed and accumulated over the patient's body.

The ^{64}Cu isotope is better suited for high-resolution PET imaging than for therapy due to its low average energy of β^+ particles (278 keV) and very low intensity of accompanying gamma radiation (1345.77 keV, with the yield of 0.475%). At the same time, its average energy of β^- particles is convenient for radionuclide therapy of small tumors [1, 2].

The ^{64}Cu isotope has numerous advantages over such PET isotopes as ^{18}F (its half-life is $t_{1/2}$ = = 109.8 min) and ^{11}C ($t_{1/2}$ = 20.4 min) currently used in clinical practice. Since the half-lives of both ^{18}F and ^{11}C are relatively short, these isotopes are usually prepared at cyclotrons located near clinics.

The ^{64}Cu isotope can be produced in the reactor by either the thermal neutron capture reaction ^{63}Cu(n,γ)^{64}Cu or the fast neutron reaction 64Zn(n,γ)^{64}Cu. However, the yields of the reactions producing ^{64}Cu in a nuclear reactor are low [3].

It should be noted that two cyclotron methods are currently used to produce ^{64}Cu isotope. One of them uses the ^{64}Ni isotope as a target, and the other the ^{68}Zn isotope.

Producing the ^{64}Cu isotope by the ^{68}Zn(p,αn)^{64}Cu reaction using protons provides certain benefits, since in this case it is possible to simultaneously produce the

$$^{67}\text{Ga}(^{68}\text{Zn}(p,2n)^{67}\text{Ga})$$

and

$$^{64}\text{Cu}(^{68}\text{Zn}(p,\alpha n)^{64}\text{Cu}),$$

isotopes used in medicine from the same target [5].

However, this method comes with several drawbacks:

firstly, a cyclotron with a higher energy of 30 MeV is required;

secondly, complex radiochemical separation is necessary;

thirdly, production of the isotopes generates highly contaminated waste from several radionuclide impurities;

fourthly, the yield of ^{64}Cu isotopes is small, since the reaction cross section is low (about 20 mb at a proton energy of 30 MeV) [5, 6].

As noted above, ^{64}Cu isotope can be obtained by the ^{64}Ni(p,n)^{64}Cu reaction with both natural and nickel targets enriched with the ^{64}Ni isotope, using protons with a relatively low energy, 10 MeV (this is considerably below 30 MeV). The drawback of using a ^{64}Ni-enriched target is that the ^{64}Ni isotope is very expensive [4].

Relatively cheap targets made of natural nickel seem to be more attractive, but the disadvantage in this case is that the content of the ^{64}Ni isotope in the target is low, and, consequently, the production efficiency is insufficiently high; moreover, large amounts of other impurities are generated during irradiation, so complex chemical procedures are required for separating these impurities and isolating ^{64}Cu [5].

However, these difficulties can be largely overcome by selecting the correct initial proton energy corresponding to the maximum cross section for the ^{64}Ni(p,n)^{64}Cu reaction (647 mb at an energy of 10.5 MeV [11]) and optimizing the waiting time after irradiation.

The goal of this study consists in analysis and optimization of producing the ^{64}Cu isotope from natural nickel using protons with energies of

10 – 15 MeV at the MGC-20 cyclotron at Peter the Great St. Petersburg Polytechnic University.

Method for analyzing the yield of isotopes in a nickel target (natural mixture) irradiated with a proton beam at energies of 10–15 MeV

As noted above, a proton beam with energies of 10 – 15 MeV can be used to obtain the ^{64}Cu isotope. The target is a natural mixture of nickel isotopes: ^{58}Ni (68%), ^{60}Ni (26%), ^{61}Ni (1.14%), ^{62}Ni (3.71%) и ^{64}Ni (0.926%) [7].

Protons with energies of 10–15 MeV can induce various nuclear reactions in the target on different isotopes of nickel, generating different isotopes as by-products that can interfere with the process of separating the ^{64}Cu isotope from the resulting isotope mixture, and determining the amount of ^{64}Cu produced by spectroscopic methods.

It follows from the above that it is useful to know the total yield of each isotope produced in different reactions (see Table 1) and compare it with the yield of the ^{64}Cu isotope. The isotopes produced in the target through irradiating a natural mixture of nickel isotopes with protons are given in Table 1 [8 – 12].

Isotope yields for a natural mixture of nickel isotopes irradiated by protons can be determined accounting for the energy losses to excitation and ionization for protons passing through the target material [13]:

$$\left\langle -\frac{dE}{dx}\right\rangle = \\ = \frac{4\pi}{m_e c^2}\frac{nz^2}{\beta^2}\left(\frac{e^2}{4\pi\varepsilon_0}\right)^2\left[\ln\left(\frac{2m_e c^2\beta^2}{I}\right)\right], \quad (1)$$

where $-dE/dx$, MeV/cm, are specific ionization losses (x is the proton penetration depth); z is the charge number of the bombarding particle; m_e, g, is the electron mass; e, Cl, is the electron charge; c, cm/s, is the speed of light; β is the ratio of the speed of the bombarding particle to the speed of light ($\beta = v/c$); I, eV, is the mean ionization potential; ε_0, F/m, is the electrical constant; n, cm^{-3}, is the electron concentration of the target,

$$n = \frac{N_A Z\rho}{AM_u};$$

N_A, 1/mol, is the Avogadro constant; ρ, g/ m^3, is the target density; Z is the charge number of the target; A is the atomic mass; M_u, g/mol, is the molar mass.

The mean ionization potential of nickel is, like the mean ionization potential for other elements, $I = 328 \pm 10$ eV [14].

Eq. (1) is simplified in the nonrelativistic case $\beta^2 << 1$, where the proton is a bombarding particle ($z = 1$):

$$\left\langle -\frac{dE}{dx}\right\rangle = \\ = \left(\frac{144\rho Z z^2}{AE}\right)\ln\left[\frac{2179E}{I}\right]. \quad (2)$$

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

The production of all isotopes at different depths in the target is found by the following formula [15]:

$$\frac{dN_i}{dx} = \\ = \left(\frac{Jn_f}{\lambda e}\right)\left(1-\exp\left(-t_{rad}\lambda\right)\right)\sigma(x), \quad (3)$$

where N_i, cm^{-3}, is the number of atoms of type i of the radioisotope produced; J, A, is the cyclotron current; n_f is the concentration of nickel isotope nuclei in natural nickel; λ, s^{-1}, is the decay constant of the radionuclide produced; t_{rad}, s, is the target irradiation time.

Integrating distribution (3) from zero to target thickness τ, we obtain the dependence for the radioisotopes produced in target on thickness τ:

$$N_i\left(\tau, t_{rad}\right) = \int_0^\tau dx\left\{\frac{dN_i}{dx}\right\}. \quad (4)$$

The decrease in proton flux with depth, as well as other processes removing protons from the

Table 1

Characteristics of isotopes produced in the target made of natural mixture of nickel isotopes irradiated with protons *p* as a result of nuclear reactions [8 – 12]

Isotope	Half-life	Nuclear reaction	E_{thr}, MeV	σ, mb, at	
				15 MeV	10 MeV
^{55}Co	17.5 h	^{58}Ni(p,α)	1.36	35.7	8.7
^{57}Co	271.74 days	^{58}Ni(p,2p)	8.3	149.3	4.9
		^{58}Ni(p, p + n)^{57}Ni→ ^{57}Co	12.3		
		^{60}Ni(p,α)	0.3		
		^{61}Ni(p, n + α)	8.2		
		^{62}Ni(p, 2n + α)	18.9		
		^{58}Ni(p,d)	10.0		
^{58}Co	70.86 days	^{61}Ni(p,α)	0.7	0.88	0.78
		^{62}Ni(p,α + n)	10.3		
^{57}Ni	35.6 h	^{58}Ni(p, p + n)	12.4	8.8	–
		^{58}Ni(p,d)	10.1		
^{60}Cu	23.7 min	^{64}Ni(p,n)	7.0	58.8	79.8
^{61}Cu	3.3 h	^{61}Ni(p,n)	3.1	186	472
		^{62}Ni(p,2n)	13.0		
^{62}Cu	9.67 min	^{62}Ni(p,n)	5.0	359.3	498.9
^{64}Cu	12.7 h	^{64}Ni(p,n)	2.5	206.0	647.0

N o t a t i o n s: E_{thr} is the threshold reaction energy, σ is the reaction cross section (for two values of the initial kinetic energy of the proton beam).

beam can be neglected in this case.

Eqs. (2), (4) and the values of the reaction cross section σ (see Table 1) were used to determine the activity of each isotope for protons with initial kinetic energies of 15 (Fig. 1, *a*,*b*) and 10 MeV (Fig. 1, *c*,*d*), cyclotron current of 2 μA, and a target made of natural nickel. Fig. 1 shows the computational results for the activity of the ^{55}Co and ^{64}Cu isotopes for targets of various thicknesses and various irradiation times.

Considering Fig. 1, we can see that a sufficient target thickness for an initial proton energy of 15 MeV is 400 μm: the dependence of the activity accumulated on the target thickness disappears at this value. This value is 200 μm for an initial energy of 10 MeV.

The activity of each isotope after the end of irradiation and after different waiting times was calculated by the equation

$$A_t = A_0 \exp(-\lambda t), \quad (5)$$

where A_0, s^{-1}, is the isotope activity at $t = 0$; A_t, s^{-1}, is the isotope activity after waiting time t, s; λ, s^{-1}, is the isotope decay constant.

The computed activities and ratios of the activities of the produced isotopes to the activities of the ^{64}Cu isotope produced for different waiting times after the end of irradiation are given in Table 2 (target irradiation time is 1.5 h, E = 15 and 10 MeV, cyclotron current is 2 μA).

After the nickel target (natural mixture of isotopes) was irradiated, the ^{64}Cu isotope had to be separated from the target. This is typically achieved by the well-known method of ion-exchange chromatography with a resin column (Dowex1-8X [1] or AG1-X8 [16]). Since some radioisotopes with a short half-life are produced in the target during irradiation (see Table 1), it is

Table 2

Comparison of the activity values of isotopes produced in the target made of a natural mixture of nickel isotopes irradiated with protons as a result of nuclear reactions

Isotope	Activity A_t, s^{-1} / (Ratio A_t / A_t ^{64}Cu)						
	$t = 0$	t=9.67 min	t=23.7 min	t=3.3 h	t=12.7 h	t=17.5 h	t=50 h
Initial kinetic energy of proton beam: 15 MeV							
^{55}Co	$\frac{2.5\cdot10^7}{(2.3)}$	$\frac{2.4\cdot10^7}{(2.4)}$	$\frac{2.4\cdot10^7}{(2.5)}$	$\frac{2.1\cdot10^7}{(2.3)}$	$\frac{1.5\cdot10^7}{(2.8)}$	$\frac{1.2\cdot10^7}{(2.85)}$	$\frac{3.4\cdot10^6}{(4.8)}$
^{57}Co	$\frac{2.3\cdot10^5}{(0.02)}$	$\frac{229996}{(0.02)}$	$\frac{229990}{(0.02)}$	$\frac{229919}{(0.02)}$	$\frac{229698}{(0.04)}$	$\frac{229572}{(0.05)}$	$\frac{229781}{(0.32)}$
^{58}Co	$\frac{17500}{(0,002)}$	$\frac{17498}{(0.002)}$	$\frac{17497}{(0.002)}$	$\frac{17476}{(0.001)}$	$\frac{17409}{(0.003)}$	$\frac{17375}{(0.004)}$	$\frac{17147}{(0.02)}$
^{57}Ni	$\frac{4.15\cdot10^5}{(0.04)}$	$\frac{413699}{(0.04)}$	$\frac{413819}{(0.038)}$	$\frac{389166}{(0.04)}$	$\frac{324061}{(0.06)}$	$\frac{295139}{(0.07)}$	$\frac{156722}{(0.22)}$
^{60}Cu	$\frac{6.0\cdot10^8}{(54.5)}$	$\frac{4.5\cdot10^8}{(45.0)}$	$\frac{3.0\cdot10^8}{(28.0)}$	$\frac{1.8\cdot10^6}{(0.19)}$	–	–	–
^{61}Cu	$\frac{3.7\cdot10^7}{(3.4)}$	$\frac{3.5\cdot10^7}{(3.5)}$	$\frac{3.4\cdot10^7}{(3.2)}$	$\frac{1.8\cdot10^7}{(1.9)}$	$\frac{2.5\cdot10^6}{(0.5)}$	$\frac{9.3\cdot10^5}{(0.2)}$	$\frac{1024}{(0.001)}$
^{62}Cu	$\frac{5.7\cdot10^8}{(51.8)}$	$\frac{2.8\cdot10^8}{(28)}$	$\frac{1.0\cdot10^8}{(9.3)}$	–	–	–	–
^{64}Cu	$\frac{1.1\cdot10^7}{(1.0)}$	$\frac{1.09\cdot10^7}{(1.0)}$	$\frac{1.07\cdot10^7}{(1.0)}$	$\frac{9.1\cdot10^7}{(1.0)}$	$\frac{5.4\cdot10^6}{(1.0)}$	$\frac{4.2\cdot10^6}{(1.0)}$	$\frac{7.0\cdot10^5}{(1.0)}$
Initial kinetic energy of proton beam: 10 MeV							
^{55}Co	$\frac{2.4\cdot10^6}{(0.30)}$	$\frac{2.4\cdot10^6}{(0.32)}$	$\frac{2.3\cdot10^6}{(0.30)}$	$\frac{2.1\cdot10^6}{(0.30)}$	$\frac{1.5\cdot10^6}{(0.40)}$	$\frac{1.2\cdot10^5}{(0.40)}$	$\frac{3.3\cdot10^5}{(0.66)}$
^{57}Co	$\frac{12500}{(0.001)}$	$\frac{12499}{(0.001)}$	$\frac{12499}{(0.001)}$	$\frac{12495}{(0.001)}$	$\frac{12483}{(0.003)}$	$\frac{12476}{(0.004)}$	$\frac{12433}{(0.02)}$
^{58}Co	$\frac{5500}{(0.0007)}$	$\frac{5499}{(0.0007)}$	$\frac{5499}{(0.0007)}$	$\frac{5492}{(0.0008)}$	$\frac{5471}{(0.001)}$	$\frac{5460}{(0.001)}$	$\frac{5389}{(0.01)}$
^{60}Cu	$\frac{1.8\cdot10^8}{(24.0)}$	$\frac{1.3\cdot10^8}{(17.5)}$	$\frac{9.0\cdot10^7}{(12.3)}$	$\frac{5.5\cdot10^5}{(0.08)}$	–	–	–
^{61}Cu	$\frac{2.2\cdot10^7}{(2.90)}$	$\frac{2.1\cdot10^7}{(2.83)}$	$\frac{2.0\cdot10^7}{(2.74)}$	$\frac{1.1\cdot10^7}{(1.70)}$	$\frac{1.5\cdot10^6}{(0.4)}$	$\frac{5.5\cdot10^5}{(0.2)}$	$\frac{609}{(0.001)}$
^{62}Cu	$\frac{2.9\cdot10^8}{(38.6)}$	$\frac{1.4\cdot10^8}{(19.0)}$	$\frac{5.3\cdot10^7}{(7.3)}$	–	–	–	–
^{64}Cu	$\frac{7.5\cdot10^6}{(1.0)}$	$\frac{7.4\cdot10^6}{(1.0)}$	$\frac{7.3\cdot10^6}{(1.0)}$	$\frac{6.3\cdot10^6}{(1.0)}$	$\frac{3.7\cdot10^6}{(1.0)}$	$\frac{2.8\cdot10^6}{(1.0)}$	$\frac{0.5\cdot10^6}{(1.0)}$

N o t e s. 1. The values are given for the activities A_t of the isotopes produced, as well as the ratios of A_t to the corresponding activities of the ^{64}Cu isotope produced (in brackets)) for different waiting times t after the end of irradiation. 2. The target irradiation time was 1.5 h, the cyclotron current was 2 μA. 3. Dashes indicate that the activity of the isotope is below 1 decay per second.

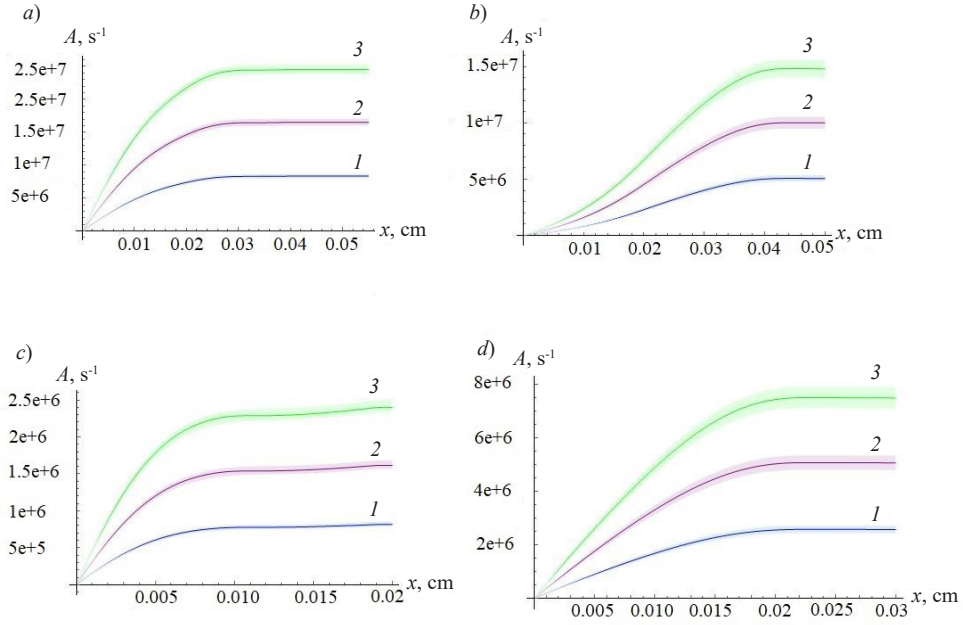

Fig. 1. Dependences of accumulated activity of ^{55}Co (*a*,*c*) and ^{64}Cu (*b*,*d*) radioisotopes on the thickness of natural nickel target for protons with an initial kinetic energy of 15 MeV (*a*,*b*) and 10 МэВ (*c*,*d*), for different irradiation times, h: 0.5 (*1*), 1.0 (*2*), 1.5 (*3*). The lines correspond to the dependence curves, and the bands to measurement uncertainty (associated with the errors in finding the reaction cross section)

preferable to wait 12–17 h before chromatography (as follows from the data in Table 2) in order to reduce the activity of the ^{60}Cu and ^{62}Cu isotopes. This gives a reduction of the activity by more than 1 million times. The chemical separation process takes about 20 h on average. As follows from the data in Table 2, a very high radionuclide purity of the ^{64}Cu isotope can be achieved, at least 99%, if it is completely separated (ideal case) from the nickel and cobalt isotopes).

Conclusion

The technology for producing the ^{64}Cu isotope has been analyzed, which has important applications in nuclear medicine, by cyclotron irradiation of a target made of natural nickel with protons (the initial kinetic energy of the proton beam is 10 and 15 MeV, the cyclotron current is 2 μA) for different irradiation times. It is established that the radionuclide purity of the ^{64}Cu isotope can be expected to be very high, reaching at least 99%, under conditions close to ideal (if nickel and cobalt isotopes are completely separated from the required copper isotope).

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