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CHARGED AND NEUTRAL PARTICLES BEAMS APPLICATION FOR RESEARCH OF ACCUMULATION ROUTES OF THE MEDICAL RADIOISOTOPE ^{103}Pd

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The results of experimental data analysis about the accumulation routes of the important medical isotope ^{103}Pd induced by charged particle beams (protons, deuterons, helium-3, helium-4) and neutral particles (gamma and neutrons) are presented. The main generalized data about the nuclear reaction cross-section of ^{103}Pd production versus the irradiation techniques of natural and isotope-enriched targets and their content of unwanted accompanying radio nuclides were compared to theoretical predictions of too. The optimization procedure of the ^{103}Pd accumulation is discussed in order to develop more promising ones, taking into account the requirements for the production of medical radioisotopes.

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INTRODUCTION

The medical isotope ^{103}Pd is widely used in the treatment of oncological diseases, for example in brachytherapy. The basis of which is the radiation source placement directly in the affected tissue. Also, the radioisotope ^{103}Pd is widely used for the treatment of prostate cancer. Its application is more acceptable than the isotope ^{125}I for this due to its emission properties: $T_{1/2}$=16.99 d, 186 Auger electrons, 95 conversion electrons, about 80 X-rays per each 100 electron capture decays, absence of high-energy γ-rays. Also, ^{103}Pd is considered promising for Auger therapy. That's why new routes research of accumulating and optimizing existing ones is an important and complex task.

The reaction cross section is important for estimating the reaction yield [1]. In the case when the research radioisotope is produced in various nuclear reactions, including the daughter isotopes production by decay, the cross sections of all production reactions can be present like a sum (provided that $T_{1/2}$ is significantly less than that of the research isotope). In case of reactions on different isotopes of the same target chemical element, the cross sections can be added by multiplying by the relative content of each isotope. Thus, it is necessary to take into account the all cross-sections of isotopes production from the decay chain of the research isotope. Part of the left decay chain for ^{103}Pd [2] is shown in Fig. 1.

For medical application of ^{103}Pd, such main final product requirements are presented [3]: the radionuclide purity more than 99.95%, and the specific activity ≥50 Ci/g. In general, to increase the final nuclear reaction yield, it is necessary either to increase the charge particle beam current value, or to find another nuclear reaction with a larger value of the production cross-section. Accumulation of unwanted concomitant isotopes usually also has to be considered. For example, in our case ^{100}Pd ($T_{1/2}$ = 3.63 days), which difficult to separate from ^{103}Pd, which prevents its medical application due to intense gamma radiation.

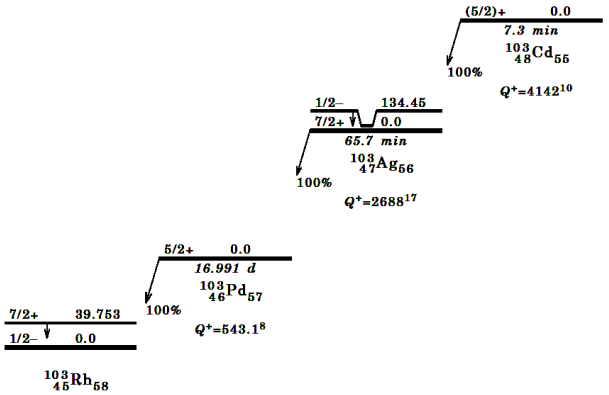


Fig. 1. Decay chain of ^{103}Pd radioisotope

Thus, the main requirements for optimizing the accumulation routes of radioisotopes should be taken into account:

- Selecting the isotopic composition of the target and the type of incident particle to increase the nuclear reaction yield;
- Approachability of incident particle beams application, with relevant energy and beam currents ranges for modern irradiation facilities;
- Inexpensive target material, simple chemical treatment of the target after irradiation with minimal losses;
- Reducing the nuclear reaction yields of unwanted isotopes for accordance of specific activity and purity requirements.

For the study of ^{103}Pd experimental yields from different reactions, their cross section values comparison is important and informative. Among the experimental techniques of nuclear reaction yields measurements depending the primary beam energy the stack thin foil technique is highlighted. This approach greatly simplifies the operational modes of charged particle accelerators, but it is characterized by the presence of primary beam losses during irradiation process, as well as difficulties in measuring activity due to X-ray self-absorption and the very weak intensity of ^{103}Pd X-rays. Therefore, it was previously decided to

apply the TALYS [4] version 1.96 computer code predictions to research for possible routes of ^{103}Pd accumulation. In practice the theoretical predictions of the dependence of the cross section (accumulation of the studied isotope) depending the primary particle beam energy for default mode, and compare it with available experimental or another theoretical model predictions of research nuclear reaction cross-section.

1. CHARGED PARTICLE BEAMS APPLICATIONS

For medical isotope accumulation, cyclotron particle accelerators are generally widely used, which are usually optimized for this purpose and provided stability and long-term operation during irradiation by requested accelerated ion beam current and energy range.

Currently, the general route of accumulating [5] ^{103}Pd is the irradiation of a rhodium target by 18 MeV proton beam of cyclotron irradiation facilities (in this case maximum cross-section value is 600 mb). Such facilities are widely used for the production of isotopes for Positron Emission Tomography. The route advantages are the high availability of such irradiation devices in the world, high specific activity, and the absence of unwanted radionuclide production. Disadvantages include the presented not enough high accelerated ion beam energy range which, accordingly, does not allow increasing the activation yield (since the thickness of the rhodium samples is ~0.05 cm as usual). It should also be taken into account that rhodium is an expensive target material.

An alternative route of ^{103}Pd accumulation is the accelerated helium ion beam application with energies up to 30 MeV [6]. A comparison of the experimental cross sections [5–7] induced by protons and helium with the theoretical prediction is presented in Fig. 2.

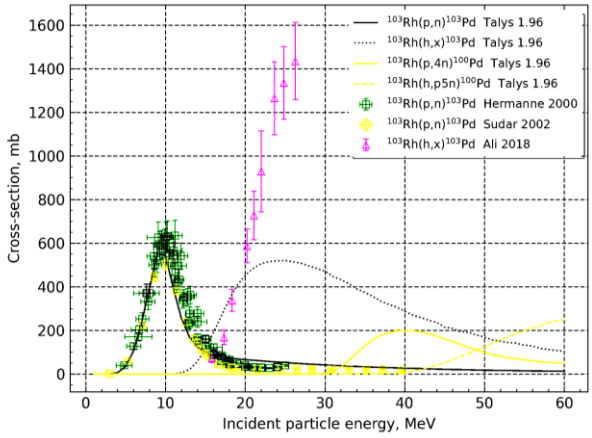


Fig. 2. Cumulative cross sections of ^{103}Pd and ^{100}Pd production for nuclear reactions of protons and heliums interactions with ^{103}Rh

It should be noted that there are a number of discrepancies both between experimental data and between experimental and calculated data. The calculated activation yield for a thick ^{103}Rh target for 18 MeV proton beam energy is 8.02 MBq/(μA·h). Instead, the experimental activation yield values are 10.24 MBq/(μA·h) measured by Hermanne et al. [5], and Sudár et al. [7] – 8.1 MBq/(μA·h). There is also a discrepancy between the experimental and calculated activation yields for helium beam application. The existing discrepancy requires a more detailed experimental study of the accumulation of ^{103}Pd since the most promising route is helium beam application with an energy of up to 30 MeV.

Similar inconsistencies are also observed in the analysis of experimental data for deuteron beam application [8–10] and *Rh* target. The experimental data do not have significant deviations, but have a large discrepancy with the calculated data (Fig. 3). The theoretical thick target activation yield for 40 MeV incident deuteron beam energy equals 20.3 MBq/(μA·h), compared with 31.4 MBq/(μA·h) measured by Hermanne et al. [8]. By the way, also found differences between the results of calculated data of Talys 1.2 and Talys 1.96 versions applications.

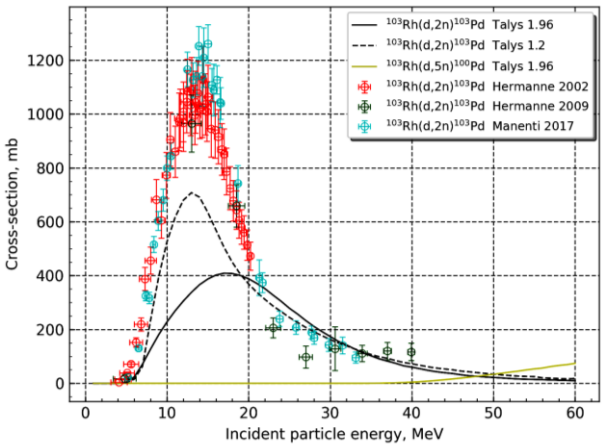


Fig. 3. Cumulative cross sections of ^{103}Pd and ^{100}Pd production for the nuclear reactions of deuteron interactions with ^{103}Rh

Another promising route of ^{103}Pd accumulation is natural or enriched silver target application for high energy proton beam interaction. The natural target is a relatively cheap and consists only of two stable isotopes: ^{107}Ag (51.8 %) and ^{109}Ag (48.2 %). The corresponding dependencies of ^{103}Pd, ^{103}Ag and ^{100}Pd production cross-sections [11–13] versus proton beam energy are presented in Fig. 4.

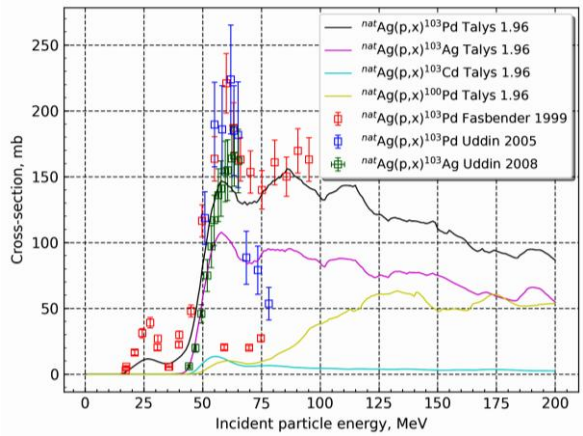


Fig. 4. Cumulative cross sections of ^{103}Pd, ^{103}Ag and ^{100}Pd production for the nuclear reactions of proton interactions with ^{nat}Ag target

The prediction of theoretical thick target activated yield for protons with an energy of 78 MeV is equal to 37.26 MBq/(μA·h), but Uddin et al. [11] experimental data for the same energy range is equal to 46.51 MBq/(μA·h).

Skakun and Qaim [14] measured and analyzed the thick target activation yields of helions and α-particles interactions with isotopically enriched Ru targets. Experimental data of cross-sections of ^{103}Pd production are shown in Fig. 5. The thick target activation yield is equaled to 0.96 MBq/(μA·h) for the ^{100}Ru(α,n)^{103}Pd reaction at 25 MeV incident beam energy (theoretical prediction value – 0.57 MBq/(μA·h)) and 1.05 MBq/(μA·h) for ^{101}Ru(a,2n)^{103}Pd (0.76 MBq/μA·h) at 23 MeV incident beam energy respectively. Corresponding theoretical cross-section values are low than experimental as usual. Proposed route applications have strong limitations because the target material is expensive.

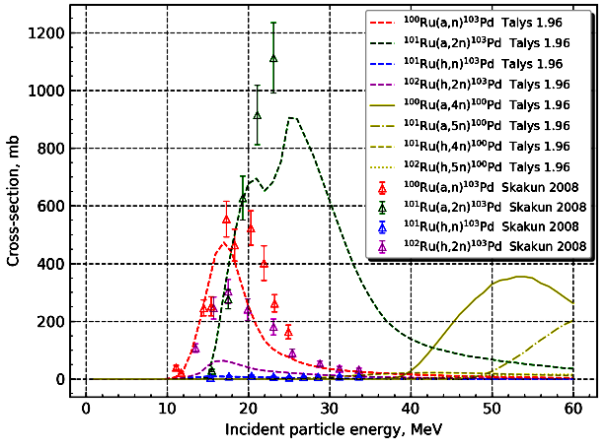


Fig. 5. Cumulative cross sections of ^{103}Pd and ^{100}Pd production for the nuclear reactions of helicon and α-particles interactions with enriched Ru targets

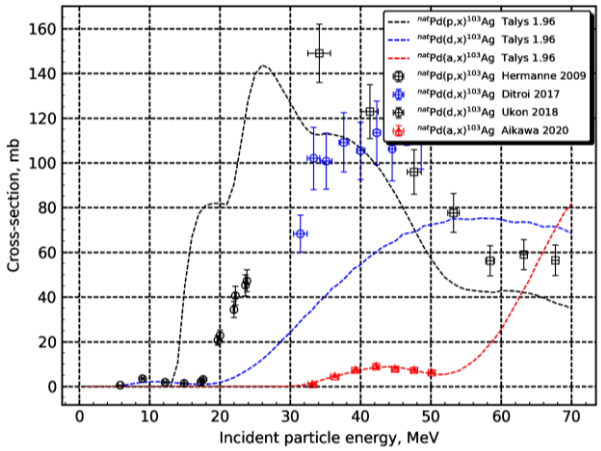


Fig. 6. Cumulative cross sections of ^{103}Ag production for the nuclear reactions of proton, helicon and α-particles interactions with ^{nat}Pd target

Hermann et al. [8], Ditroi et al. [15], Ukon et al. [16], and Aikawa et al. [17] studied nuclear reactions induced by protons, deuterons, and α-particles with ^{nat}Pd targets for ^{103}Ag production ($T_{1/2}$=65.7 min). The main idea is possibility to chemical extraction application of ^{103}Ag after accumulation process until it decays into ^{103}Pd. Corresponding cross-sections are presented in Fig. 6 and Talys 1.96 prediction data for comparison too.

In addition, TALYS 1.96 applying, the values of the tritons interaction cross sections with natural silver and rhodium targets (Fig. 7) for the accumulation of ^{103}Pd were calculated. As results for 40 MeV tritons ^{103}Rh thick target yield is 31 MBq/(μA·h). And triton energy range to 25 MeV is more acceptable in comparison the case ^{107}Ag application. There are no experimental data about the discovered cross-section data. The main problem is fast neutron yield increasing during long-term experimental target irradiation for high-energy tritium beam application.

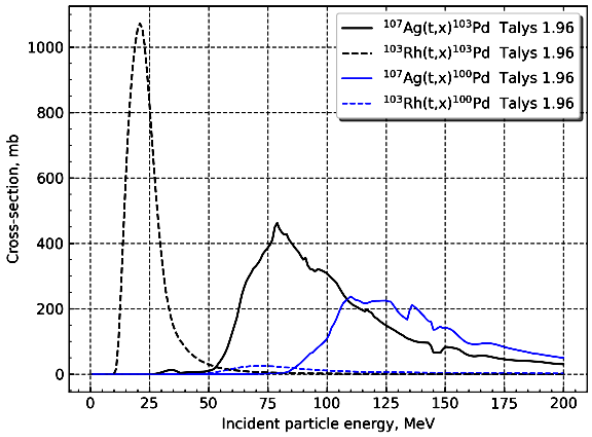


Fig. 7. Cumulative cross sections of ^{103}Pd and ^{100}Pd production for the nuclear reactions of tritons interactions with ^{107}Ag and ^{103}Rh targets

2. NEUTRON ACTIVATION

For a long period, the leading position in the medical isotopes production was occupied by nuclear reactors [18]. Their application provide the 10^{14}…10^{15} n/(s·cm^2) thermal neutron flux irradiation of experimental samples with significant volumes. Potentially, other sources can also be used for the production of isotopes, for example [19] controlled by ion accelerators with standard targets application of light elements (such beryllium, lithium…). Nuclear reactions (p,n), (d,n), (α,n) are usually used, but they have a low efficiency (10^{-2} n/particle [20]), respectively, a relatively low neutron flux of 10^{11}…10^{12} n/s, which is limited by accelerating ion current and difficulties with target cooling.

The most compact neutron sources are also DT generators, which are deuteron accelerators up to energy ~ 300 keV, and neutron energy ~ 14 MeV. Of cause, they are not suitable for medical isotope production due to the low neutron intensity $\geq 10^{10}$ n/s for 4π distributions. And also the strong limitation of their operation (neutron production target lifetime) is present too and close to several thousand hours depending on the operation mode (direct, pulse…).

Intense electron accelerators of irradiation facilities are also used as neutron sources, the bremsstrahlung radiation of which is used to generate photoneutron fluxes [21]. But it is well known that spallation sources produce the highest neutron intensity among all types of neutron generation facilities. For example, for SNS [22] 1 GeV accelerated proton beam with a current of

1.5 mA interacts with a liquid mercury target. As a result, the neutron production intensity is 10^{17} n/s. Of cause, high neutron production intensity is very important for the medical isotope accumulation technique providing. Production of high-energy neutron flux by spallation source opens a new possibility for medical isotope accumulation development.

For the accumulation of ^{103}Pd in the ^{102}Pd(n,γ)^{103}Pd reaction, the application of thermal neutrons was considered as more effective. Natural palladium consists of 6 isotopes: ^{102}Pd (1.02%), ^{104}Pd (11.14%), ^{105}Pd (22.33%), ^{106}Pd (27.33%), ^{108}Pd (26.46%), ^{110}Pd (11.72%). Provided the experimental target is enriched with ^{102}Pd, a sufficient specific activity of 500 Ci/g [23] produces during irradiation in the neutron reactor irradiation facility with a relatively low content of unwanted accompanying isotopes.

The average energy of fast neutrons in the reactor is approximately 1…2 MeV. For the spallation neutron source irradiation facility high energy (more than 45 MeV) neutron interactions with an enriched or natural silver target could be discuss as a perspective ^{103}Pd accumulation route. Theoretical predictions of according to the nuclear reaction cross-section are presented in Fig. 8.

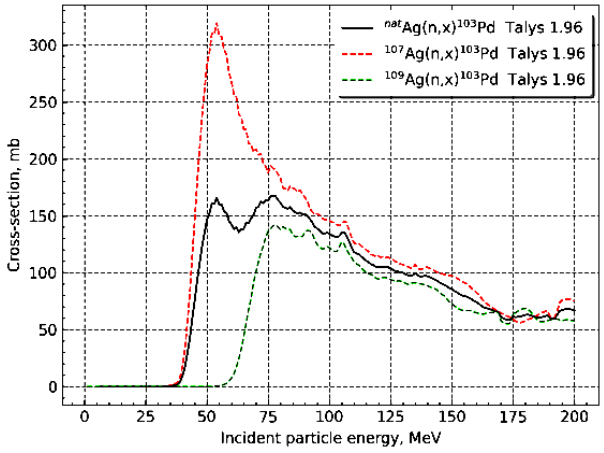


Fig. 8. Cumulative cross sections of ^{103}Pd production for the nuclear reactions of high energy neutron interactions with ^{107}Ag, ^{109}Ag and ^{nat}Ag targets

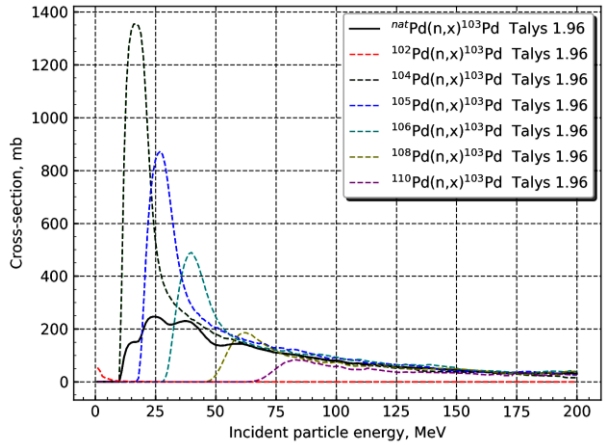


Fig. 9. Cumulative cross sections of ^{103}Pd production for the nuclear reactions of fast neutron interactions with stable palladium isotopes

For the case of ^{104}Pd enriched target application in the ^{103}Pd accumulation route, it is sufficient to apply fast neutrons with energy of up to 20 MeV. The calculated cross-section data are presented in Fig. 9. The presented data is characterized by the presence of a narrow maximum, which requires the fulfillment of special conditions during the irradiation and enrichment of experimental palladium samples.

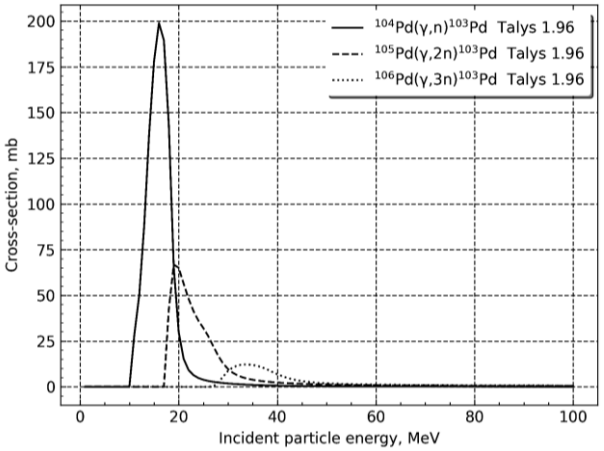
3. PHOTOACTIVATION

The generation of intense high-energy flux of γ-quanta occurs due to the application of electron accelerators, which, depending on the principle of their formation, differ only in the spectral distribution of energy (bremsstrahlung or Compton scattering).

For photoactivation technique as a route of accumulating medical isotopes, there are certain limitations associated with the mandatory isotopic enrichment of the experimental target, the selection of the optimal energy range of irradiation to ensure the radioactive purity of the accumulated isotope, and the influence of the background conditions associated with the accompanying photoneutrons.

A detailed investigation of the ^{103}Pd accumulation nuclear reactions in the interaction of γ-quanta with stable isotopes of palladium has not yet been carried out. Dikiy et al. [24] estimated the possibility of ^{103}Pd accumulation. Natural targets were irradiated by fluxes of bremsstrahlung and photoneutrons of 40 MeV Electron Linac NSC KIPT. Integral an activation yield of ^{103}Pd was experimentally determined. And it equals to 4 μCi/(μA·h·g).

Taking into account Talys 1.96 predictions (Fig. 10) of photonuclear reaction cross sections and experimental data [24], it follows that optimization of ^{103}Pd accumulation in this case depends on gamma-quanta energy range distributions, isotopic enrichment of experimental targets and the neutron background reduction during irradiation experiment. The application of bremsstrahlung flux should not be considered promising, but taking into account the regional features of the placement of charged particle accelerators for the development of medical isotopes, it is possible as an additional one.



Puc. 10. The cross-section of ^{103}Pd production for the nuclear reactions of gamma-quanta interactions with natural isotopes of palladium

CONCLUSIONS

Preliminary data analysis on experimental and theoretical values of nuclear reaction cross-sections is presented. All of them applied or considered possible for ^{103}Pd accumulation technology. The existing nuclear data base of experimental cross-section requires deeper analysis and comparison with theoretical predictions in order to optimize computer codes, which will allow more accurate prediction not only of nuclear reactions of desired isotopes, but also of accompanying ones.

The main results of presented data about ^{103}Pd accumulation routes are following that: isotopic enrichment of any experimental samples for example with ^{102}Pd (1.02% – natural distribution) or ^{104}Pd (11.14%) makes any of the proposed routes of accumulating ^{103}Pd more expensive.

It is an indisputable fact that, at present, ^{103}Rh application as a target has advantages over all other target materials. On the other hand, the accumulation of the desired isotopes definitely depends on the parameters of the beams and flux of primary particles, namely their number in the required energy range.

The rapid development of accelerator facilities for spallation neutron source provides to consider more complex ^{103}Pd accumulation routes not only due to the possibility of applying significant flux of fast neutrons. In particular, it is necessary to carry out a more detailed theoretical analysis taking into account fission reactions as a promising accumulation of ^{103}Pd, for example, nuclear reactions of ^{nat}Bi(n,f), ^{nat}Bi(γ,f) or ^{nat}Pb(n,f), ^{nat}Pb(γ,f). These elements are widely used in cooling systems neutron production targets of many types of spallation neutron sources.

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ЗАСТОСУВАННЯ ПУЧКІВ ЗАРЯДЖЕНИХ ТА НЕЙТРАЛЬНИХ ЧАСТИНОК ДЛЯ ДОСЛІДЖЕННЯ ШЛЯХІВ НАКОПИЧЕННЯ МЕДИЧНОГО РАДІОІЗОТОПУ ^{103}Pd

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Представлено результати аналізу експериментальних даних з накопичення важливого медичного ізотопу ^{103}Pd при використанні пучків заряджених частинок (протонів, дейтронів, гелію-3, гелію-4) та нейтральних частинок (гамма та нейтронів). Отримані узагальнені дані з активаційних виходів ^{103}Pd у залежності від методики опромінення натуральних та ізотопно-збагачених мішеней і вмісту в них небажаних супутніх радіонуклідів. Обговорюються шляхи оптимізації накопичення ^{103}Pd з метою пошуку більш перспективних з урахуванням вимог виробництва медичних радіоізотопів.