# ESTIMATION OF PALLADIUM-103 RADIONUCLIDE PRODUCIBILITY AT THE CYCLOTRON CV-28 FOR NUCLEAR MEDICINE

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Consideration is given to the possibility of $^{103}Pd$ radionuclide production in nuclear reactions on protons and deuterons at the cyclotron $CV-28$, for a subsequent use of the radionuclide in medicine, in particular, for brachytherapy. Activity estimates of $^{103}Pd$ produced on both the internal and external targets of the cyclotron $CV-28$ are given. The energy transferred by the proton/deuteron beam to the target under irradiation is determined. Possible ways of heat removal from the target substrate are considered.

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## 1. INTRODUCTION

Currently, there has been a considerable growth in the diagnostics of human diseases with the help of radioisotope injection into the human body (radionuclide diagnostics). Radionuclides are widely used in nuclear medicine for both the diagnostics and the treatment of different diseases [1, 2].

The prostate gland cancer is the most frequently occurring form of malignant tumor of men. The treatment may include a temporary implantation of radioactive source (radionuclide) for a certain base period, after which it is extracted from the body. As an alternative, the radioactive source may be implanted permanently into the patient's body, where it stays until it becomes weak for the pre-calculated time (brachytherapy). The use of temporary or permanent implantation is dependent on the type of the isotope chosen, the duration and intensity of required treatment. Compared to temporary radioactive sources, permanent implants for the treatment of prostate gland comprise radioisotopes that have a relatively short half-life and a low energy radiation. These are, for example, iodine-125 (half-life period $T_{1/2} = 60\,days$) or palladium-103 ($T_{1/2} = 17\,days$). The radioisotopes are usually placed inside a sealed container made from a biocompatible material (e.g., titanium) in the shape of a "grain", which is then implanted. Generally, about 50 to 120 such grains are introduced into the patient's body. They are arranged as a three-dimensional array formed by pricking numerously with a needle, which introduces the in-line grains. The radioactive source positioned in this way in the vicinity of the body's zone to be treated. The advantage of this technique is that high radiation doses can be delivered to the site of treatment at one time with relatively low radiation doses for the surrounding or intermediate healthy tissues.

Up to now, there has been no definite decision as to which of the radionuclides ($^{125}I$ or $^{103}Pd$) should be preferred in the treatment of prostate or pancreas.

In recent years, there has been an appreciable increase in the number of patients who threw off the prostate disease by the method of permanent brachytherapy (over 40 000 people in 1998 in the USA, and more than a 50% increase of recovered in 2006). Palladium-103 was proposed for interstitial implantation in 1958 [3]. And only in 1987 it became commercially possible to encapsulate palladium-103 sources, with 10 accelerators designed for their production. Regular communications on the satisfactory outcome of palladium-103 application in brachytherapy are published by the Brachytherapy Society of America [4].

## 2. PRODUCTION OF PALLADIUM-103 AT THE CYCLOTRON CV-28

Palladium-103 radionuclide is produced at the accelerators at rather low energies. Generally, palladium-103 can be produced in the reaction $^{102}Pd(n,\gamma)^{103}Pd$, which is based on natural palladium containing 1% of $^{102}Pd$, and shows rather high neutron capture cross sections. However, due to a low content of $^{102}Pd$, this technique does not yield short-lived palladium-103 isotope in sufficient quantities that are required for its regular use in medical 2- or 3-week cycles. Therefore, for the production of palladium-103 use was made of the reaction $^{103}Rh(p,n)$ $^{103}Pd$ with a low threshold energy

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$E_{thr} = 1.35\,MeV$. The method is based on irradiation of metallic rhodium targets with protons. The yield of palladium-103 isotope is strongly dependent on the proton energy, reaching its maximum at the highest attainable energies at the cyclotron CV-28 (21 to 24 $MeV$). This procedure is followed by a labor-intensive chemical separation of the radionuclide from an expensive metallic rhodium target. An increase in the incident proton energy above 18 $MeV$ in the production of palladium-103 has no sense for several reasons. Over this energy, an additional reaction yield is negligibly small (a gain in yield is less than 4% within the 18...20 $MeV$ energy range). However, at these energies the contamination with $^{101}Pd$, $^{101m,\,101g,\,102m,\,102g}Rh$ nuclides becomes appreciable [3].

The nuclear data on cross sections for this reaction, on the yield from a thick target and on possible impurities occurring during the use of the reaction on rhodium are few in number, especially at energies higher than 12 $MeV$.

The mentioned reactions were investigated from threshold energies up to 23 $MeV$ (see ref. [5]).

The production of palladium-103 radioisotope in the reaction $^{103}Rh(d,\,2n)^{103}Pd$ is an alternative and more promising method [6]. For optimization of palladium-103 production from rhodium-103 the cross sections of two main reactions $^{103}Rh(p,n)^{103}Pd$ and $^{103}Rh(d,2n)^{103}Pd$ are of importance. The yields from thick targets, obtained from the corresponding cross sections at a deuteron energy of 20 $MeV$ are nearly two times higher than the yields at exposure to protons [6]. For palladium production, an increase in the deuteron energy beyond 20 $MeV$ is unbeneficial for two reasons: an additional yield at higher energies is insignificant (only a 10% increase at energies between 20 and 22 $MeV$). However, similarly to the case of proton irradiation, the $^{101m,\,101g,\,102m,\,102g}Rh$ impurities become increasingly important, because the cross sections of their production quickly grow at energies higher than 20 $MeV$. At the stage of rhodium target recovery, these impurities separate from palladium, however, the accumulation of these long-lived contaminating radionuclides can be observed with further recovery of rhodium targets.

At present, palladium-103 radionuclide is produced almost without exception during irradiation of the rhodium target with protons. Experimental data on the reaction cross sections have been collected and published elsewhere [3].

In the nearest future, the NSC KIPT team intends to put into operation the isochronous cyclotron CV-28 with an adjustable energy of light ions $p$, $d$, $^3He$, $^4He$. Below, Table gives the characteristics of this cyclotron [7].

| Accelerated particles | Accelerated particle energies, $MeV$ | External target current, $\mu A$ | Internal target current, $\mu A$ |
|---|---|---|---|
| $p$ | 2-24 | 40-60 | 200 |
| $d$ | 4-14 | 50-100 | 300 |
| $^3He$ | 6-36 | 5-50 | 135 |
| $^4He$ | 8-28 | 6-40 | 90 |

In connection with this it appears of interest to estimate the possibility of palladium-103 radionuclide production at this cyclotron for a subsequent use in medicine, in particular for brachytherapy.

From the above-given analysis it is evident that the palladium production at the cyclotron CV-28 can be realized from both the reaction $^{103}Rh(p,n)^{103}Pd$ and the reaction $^{103}Rh(d,2n)^{103}Pd$ on the internal and external targets of the cyclotron. As the energy increases over 18 $MeV$ for protons and over 20MeV for deuterons, then, as mentioned above, deeper nuclear reaction channels begin to open. Apart from the particle energy, the beam particle current and the irradiation duration are also the essential factors for radionuclide production.

Here we consider the possibility of palladium-103 production at the KIPT cyclotron CV-28 from both the reaction $^{103}Rh(p,n)^{103}Pa$ and the reaction $^{103}Rh(d,\,2n)^{103}Pd$ on the internal and external targets of the cyclotron.

Since the activity yield, which is determined as the activity produced under irradiation for 1 hour at 1 $\mu A$ current, and which is a convenient parameter that characterizes the rate of radionuclide production, we have calculated the activities of palladium-103 produced for the both cases of radionuclide production in proton and deuteron reactions for the internal and external targets of the cyclotron. The energy transferred by the proton/deuteron beam to the rhodium target under irradiation has also been calculated for the both cases.

## 3. ESTIMATION OF ACTIVITY FOR PALLADIUM-103 PRODUCED IN PROTON AND DEUTERON REACTIONS AT THE CYCLOTRON CV-28

The $^{103}Rh(p,\,n)^{103}Pd$ reaction cross sections have been measured in ref. [3] in the proton energy range from 14.7 to 29.4 $MeV$. The excitation function of the reaction was determined in the energy range from $(4.8 \pm 1.0)$ to $(25 \pm 0.4)$ $MeV$ by fitting to the experimental data newly obtained in [3] and those existed previously [11, 12]. On the strength of the mentioned data the radionuclide yields from a thick target were calculated. We have used the data to estimate the activity of radionuclides produced at the cyclotron CV-28. Thus, according to our estimates, for the proton energy chosen to be $E_p = 18\,MeV$ and currents $I_p = 200\,\mu A$ and $I_p = 60\,\mu A$ on internal and external targets of the cyclotron, respectively, the activities $2.05\,GBq$ ($55.35\,mCi$) and $0.61\,GBq$ ($16.6\,mCi$) can be, correspondingly, attained for 1 hour of irradiation.

For the chosen proton energy and rhodium target, the specific activity yield, i.e., activity per unit target mass, was calculated in the assumption of uniform activity distribution in the radius and thickness of the target. At target diameter $d = 1\,cm$ and the target thickness equal to the proton range in rhodium at energy $E = 18\,MeV$ the specific activity in the internal and external targets exposed to radiation will make up $B = 0.1\,mCi/mg$ and $B = 0.03\,mCi/mg$, respectively. In this case, the target thickness should be somewhat greater than the proton range in rhodium equal to $0.7003\,g \cdot cm^{-2}$ or $564.3\,\mu m$ [3].

On this basis, with production of palladium-103 radionuclide in the deuteron reaction $^{103}Rh(d,2n)\,103$ we have chosen the deuteron energy to be $14\,MeV$. The reaction cross sections were measured in work [6] at deuteron energies of $15.3\,MeV$ and $20.5\,MeV$. The excitation function was determined by fitting to already available theoretical calculations [13] and to the experimental data recently measured in [6] in the energy range from $(4.1 \pm 1.0)$ to $(20.2 \pm 0.2)\,MeV$. As in the case of proton irradiation, the radionuclide yields from thick targets, calculated from the excitation function [5], were used to estimate the palladium radionuclide activities, which might be produced at the cyclotron CV-28 at the deuteron energy $E_d = 14\,MeV$ and deuteron currents $I_d = 300\,\mu A$ and $I_d = 100\,\mu A$ in the internal and external targets, respectively. We have obtained that for 1 hour of irradiation, activities 3.09 and $1.03\,GBq$ (or 83.5 and $27.8\,mCi$) can be attained on internal and external targets of the cyclotron, respectively.

The yield of specific activity of palladium-103 radionuclide produced from the deuteron reaction was calculated in a similar way. In this case, the deuteron range in rhodium is calculated from the proton range by the relation given in paper [10] as

$$R_d = \frac{m_d}{m_p} R_p \left( \frac{m_p}{m_d} E \right),$$

where $R_d$ and $R_p$ are the ranges, while $m_d$ and $m_p$ are the masses of the deuteron and the proton, respectively.

At deuteron energy $E_d = 14\,MeV$ the range Rd will be equal to $0.2926\,g/cm^2$ or $235.8\,\mu m$.

The specific activity of palladium-103 at the beam diameter d=1 cm will be $B = 0.37\,mCi/mg$ and $B = 0.12\,mCi/mg$ on internal and external targets of the cyclotron, respectively.

The target thickness must be somewhat greater than the deuteron range in rhodium.

The resulting total radionuclide activity is limited only by the heat power that can be dissipated on internal cyclotron targets. The energy transferred by the proton beam to the target under irradiation and converted mainly to heat will attain $1.08\,kW \cdot h$ and $3.6\,kW \cdot h$ on the external and internal targets, respectively. At irradiation with deuterons, the energy released on the external and internal targets will be $1.4\,kW \cdot h$ and $4.2\,kW \cdot h$, correspondingly. Though the melting temperature of rhodium is high ($1963°C$), it would be necessary, when working, to remove the heat from high heat conduction target backing by cooling them with a heat carrier (e.g., water).

The manufacture of a water-cooled rhodium target that permits the dissipation of high heat energy presents difficulties. So the heat removal from the target to be exposed to protons and deuterons at the cyclotron CV-28 is a rather complicated but solvable problem.

Thus in ref. [8] it was shown that the use of an external water-cooled rhodium target of certain design and its positioning at an angle to the main beam trajectory make it possible to dissipate up to $2\,kW$ of heat energy.

A more considerable heat removal from 6 up to $12\,kW$ (current $400\,\mu A$ and the proton irradiation energy $30\,MeV$) was realized with some substantial modifications in the design of a set of targets and with a special method of target cooling with water by means of a ducting system [9].

## 4. CONCLUSIONS

The present estimates of palladium-103 radionuclide activities point to a real possibility of production of the mentioned radionuclide on internal and external targets of the NSC KIPT cyclotron CV-28, using both proton and deuteron reactions.

If the irradiation run lasts 10 *hours*, then with the reaction on deuterons the total activity of the resulting palladium-103 makes 0.835 and $0.276\,Ci$ on the internal and external cyclotron targets, respectively, while in the case of the proton reaction it makes 0.553 and $0.166\,Ci$ on the internal and external targets, respectively.

Therefore, with the use of the deuteron reaction, the cyclotron CV-28 can annually produce about $200\,Ci$ per an. on the internal target and about $100\,Ci$ per an. on the external cyclotron target. If it is remembered that one run of brachytherapy consumes about 50 to $70\,mCi$, then it can be easily seen that the amount of the activity produced will be sufficient to carry out about 2000 brachytherapy runs for the patients.

At an average price of about 50 hryvnyas ($\sim$ 10 U.S. dollars) for $1\,mCi$ of palladium-103, the annual capacity of the complex under discussion may be worth of 1...2 million hryvnyas.

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## ОЦЕНКА ВОЗМОЖНОСТИ НАРАБОТКИ РАДИОНУКЛИДА ПАЛЛАДИЙ-103 НА ЦИКЛОТРОНЕ CV-28 ДЛЯ ЯДЕРНОЙ МЕДИЦИНЫ

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В работе рассматривается возможность получения радионуклида палладий-103 на циклотроне CV-28 в реакциях на протонах и дейтронах для использования его в медицине, для брахитерапии в частности. Приведены рассчитанные оценки активностей наработанного $^{103}Pd$ на внутренней и наружной мишенях циклотрона CV-28. Определена энергия, передаваемая пучком протонов и дейтронов мишени при облучении. Обсуждается возможность отвода тепла от подложки мишеней.

## ОЦІНКА МОЖЛИВОСТІ НАПРАЦЮВАННЯ РАДІОНУКЛІДА ПАЛАДІЙ-103 НА ЦИКЛОТРОНІ CV-28 ДЛЯ ЯДЕРНОЇ МЕДИЦИНИ

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У роботі розглядується можливість здобуття радіонукліда паладій-103 на циклотроні CV-28 в реакціях на протонах і дейтронах для використання його в медицині, для брахітерапії зокрема. Приведено розраховані оцінки активностей напрацьованого $^{103}Pd$ на внутрішній і зовнішній мішенях циклотрона CV-28. Визначена енергія, яка передається пучком протонів і дейтронів мішені при опроміненні. Обговорюється можливість відведення тепла від підкладки мішеней.