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Study of heat transfer parameters on rhodium target for ^{103}Pd production

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Abstract. The efficiency of cooling and the particle beam characteristics are important when high beam current irradiations are intended for production of radionuclides. The efficiency of cooling is determined by both the target carrier geometry and the flow rate of coolant, while the beam characteristics deal with the current density distribution on the irradiated surface area. Heat transfer on rhodium target to produce ^{103}Pd via the $^{103}Rh(p,n)^{103}Pd$ reaction was investigated and the beam current was obtained more than 500 μA.

Key words: heat transfer • ^{103}Pd • production • rhodium • target

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Introduction

The suitability of a given radionuclide for brachytherapy is determined by its half-life and by the type, energy and abundance of the emitted radiation. Due to the ^{103}Pd half-life (16.97 days) and its electron capture decay resulting in the abundant emission of Auger electrons and low-energy X-rays [EC, K_α = 20.1 keV (64.7%), K_β = 22.7 keV (12.3%)], ^{103}Pd can be used for the preparation of seeds as permanent interstitial implants for the treatment of rapidly proliferating tumours. Along with ^{125}I, the radionuclide has been used in the treatment of various cancers such as eye, brain, neck, uterus, colon, but it is now primarily used for prostate tumours. Over the last decade, it has proven to be very effective in treating this cancer with minimum side effects [9–11].

For cyclotron production of ^{103}Pd, the most suitable is the reaction $^{103}Rh(p,n)^{103}Pd$ [2, 6, 12]. One of the most important parameters in producing radionuclides by target bombardment with energetic charged particles is the heat transfer factor. When the accelerated particles hit atoms at the target, there is an interaction that leads to excitation, ionization and Bremsstrahlung. Some of the kinetic energy converts into heat that produces undesirable effects on the production conditions (such as melting of the target and peeling of the electroplating layer). In this research, the heat transfer factor on rhodium target for producing ^{103}Pd has been studied.

Methods

The production of ^{103}Pd is carried out via the $^{103}Rh(p,n)^{103}Pd$ reaction which is well suited to low-energy cyclotrons [2, 6, 12]. Cyclone-30 (IBA, Belgium,

I_{max} = 350 μA, $E_{P_{max}}$ = 30 MeV) at the Agricultural, Medical and Industrial Research School was employed. The solid targetry system in this cyclotron is made up of a pure copper backing on which the target materials are electrodeposited (surface area, s = 11.69 cm^2).

To take full benefit of the excitation function for the ^{103}Rh(p,n)^{103}Pd reaction and to avoid the formation of the radionuclide ^{101}Pd impurity, the proton entrance energy should be E_{P1} = 18 MeV [2, 6, 12]. The physical thickness of the rhodium layer is chosen in such a way that for a given beam/target angle geometry the particle exit energy should be E_{P2} = 5 MeV. According to SRIM code [15], the thickness has to be 475 μm for 90° geometry. To minimize the thickness of the rhodium layer a 6° geometry is preferred, in which case a 48 μm layer is recommended.

The target that undergoes bombardments by the proton beam at the cyclotron production consists of three layers, namely: a) rhodium layer, b) copper layer and c) copper layer without induced proton beam (see Fig. 1).

This target is surrounded by the accelerator vacuum on one side and by the coolant fluid on the other side. The rhodium target was irradiated with a proton beam at an incidence angle of 6°, and some of the proton energy converts into heat. If the irradiation is considered uniform on the rhodium layer, it means a current density (μA·mm^{-2}) on this layer and the relationship between the heat production rate Q_{Rh} (W) and the heat production rate per volume unit Q'''_{Rh} (W·cm^{-3}) will be:

(1) $$Q_{Rh} = Q'''_{Rh} \times a \times s$$

In this relation, s is the target surface area in cm^2. The exiting protons from the rhodium layer (a layer in cm) are stopped completely in the second layer (cuprous layer, b in cm). The relation between heat production rate in this layer Q_{Cu} and the heat production rate per volume unit Q'''_{Cu} is as follows:

(2) $$Q_{Cu} = Q'''_{Cu} \times b \times s$$

The third layer (c) serves just as a mechanic retentive device. The heat produced in the first and second layers, transfers to the coolant fluid through this last layer in the form of conduction.

(3) $$Q_t = Q_{Rh} + Q_{Cu}$$

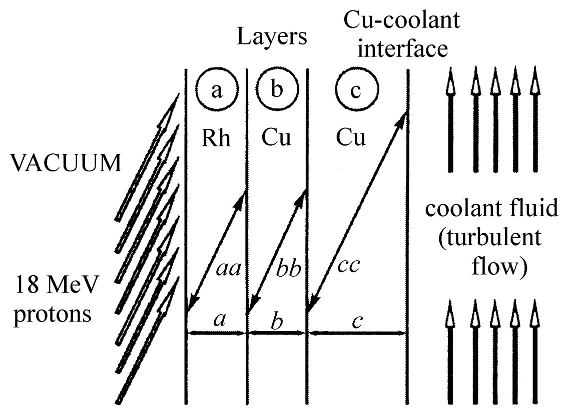

Fig. 1. Schematic of the three layers of Rh target.

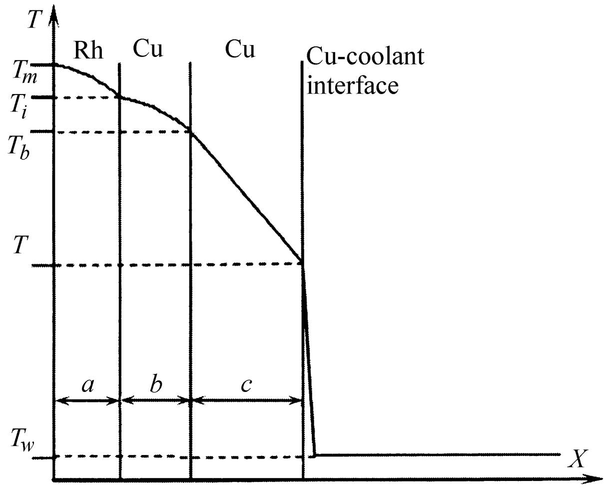

Fig. 2. Profile of temperature in the Rh target.

Under stationary conditions, heat transfer appears as a temperature profile (Fig. 2) in which T_m is the temperature at the vacuum-rhodium interface, T_i is the rhodium and second layer interface temperature, and T_b is the temperature of the second and third layer interface. Finally, T_c is the temperature of the Cu-coolant interface. T_w is the temperature of the coolant fluid.

The temperature profile can be obtained solving the Poisson, Laplace and Newton's equations in the three layers and the coolant. Poisson's equation in the first and second layer is $d^2T/dx^2 = -Q'''/k$ where k (W·cm^{-1}·K^{-1}) is the metal thermal conductivity. The Laplace's equation in the third layer is $d^2T/dx^2 = 0$ and Newton's equation for temperature profile in the third layer (c) and the coolant interface is $Q_t = h{\cdot}s{\cdot}\Delta T$ [1, 7, 8, 14]. In the latter formula, h is the heat transfer coefficient in convection method (W·cm^{-2}·K^{-1}).

Integration of Poisson, Laplace's equations and appropriate boundary conditions will result in a temperature profile relation which gives Q_{Rh}, Q_{Cu}, Q_t for beam current (I) and proton energy (ΔE) in the related layers.

The mentioned temperature profile relation is:

(4) $$T_m - T_w = \left(\frac{Q_{Rh}}{k_{Rh}}\cdot\frac{a}{s}\right) + \left(\frac{Q_{Rh}}{k_{Cu}}\cdot\frac{b}{s} + \frac{Q_{Cu}}{2k_{Cu}}\cdot\frac{b}{s}\right) + \left(\frac{Q_T}{k_{Cu}}\cdot\frac{c}{s}\right) + \left(\frac{Q_T}{h\cdot s}\right)$$

k_{Rh} and k_{Cu} (W·cm^{-1}·K^{-1}) are the rhodium and cupper thermal conductivity, respectively. The heat produced in the rhodium layer can be obtained by considering the mean energy of the proton and the total number of protons that enter in this layer, in units of time:

(5) $$Q_{Rh} = N \cdot \Delta E_P \text{ (W)}$$

ΔE_P is the beam energy variation at the desired layer and N is the total number of protons entering the layer per second, and it is obtained from the following equation:

(6) $$N = \frac{I \times 10^{-6}(C/s)}{q_p(C)} = 6.25 \times 10^{12} \times I(s^{-1})$$

where: $I(A)$ and $q_p(C)$ are the beam current and the proton charge, respectively.

After the protons exit from the rhodium layer, they will stop in the second layer (b layer). In this layer, the heat production rate (Q_{Cu}) can be calculated as well. By assuming turbulent flow regime, it means no boiling will happens in the interface between the third layer and the coolant, the difference between T_c and T_w can be obtained from the following equation:

$$Q_t = h \cdot s \cdot (T_c - T_w) \tag{7}$$

Dittus and Boelter proposed the following equation to predict heat transfer coefficient for turbulent flow of fluid [3–5, 13]:

$$h = C \times \frac{k_c}{D_e} \times R_e^m \times P_r^n \tag{8}$$

where: n, m, C are constants, R_e is Reynolds number dimensionless, P_r is Prandtl number, D_e is the equivalent diameter of the coolant channel (cm) and finally k_c is the fluid thermal conductivity ($W \cdot cm^{-1} \cdot K^{-1}$).

R_e defined by $R_e = D_e \cdot \nu \cdot \rho / \mu$, where ν is the average linear velocity of coolant fluid ($cm \cdot s^{-1}$), ρ is the coolant density and $\mu = 10^{-2}$ is the kinematic viscosity of the coolant fluid in $cm^2 \cdot s^{-1}$. The quantity D_e is obtained in the following equation:

$$D_e = 4 \times O/L \tag{9}$$

where O is the cross-sectional area of the coolant channel and L is its perimeter. Figure 3 shows the cross-sectional area of the coolant channel. By choosing the right parameters in order to have $R_e > 10{,}000$ condition, turbulent flow condition will be satisfy. The magnitude of h depends on P_r too, which is shown below, as an equation, k_{H_2O} ($W \cdot cm^{-1} \cdot K^{-1}$) and C_p ($J \cdot cm^{-3} \cdot K^{-1}$) are the thermal conductivity and specific heat of the coolant fluid, respectively.

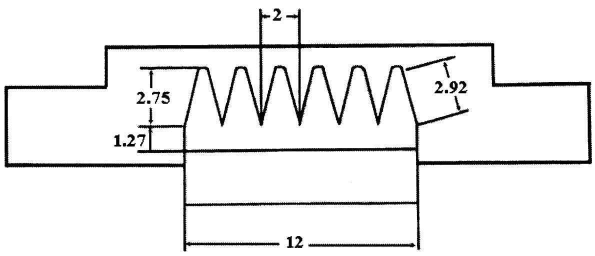

Fig. 3. Cross-section of the Rh target.

$$P_\gamma = C_p \times \mu / k_{H_2O} \tag{10}$$

Results and discussion

In Table 1 are shown all the data of target and parameters that were considered for the present work. The thermal shock tests involved the heating of the target up to 500°C (the temperature that the Rh layer can experience during a high current irradiation) for 1 h followed by submersion of the hot target in cold water

Table 1. Nomenclature and values of parameters

Quantity	Symbol	Value	Dimension
Input data			
Thickness of the third layer	c	0.1	cm
Reynolds number	R_e	67251	dimensionless
Empirical constant	m	0.8	dimensionless
Empirical constant	n	0.4	dimensionless
Empirical constant	C	0.023	dimensionless
Coolant flow rate	f	833	$cm^3 \cdot s^{-1}$
Prandtl number	P_r	7.0	dimensionless
Rhodium thermal conductivity	k_{Rh}	1.50	($W \cdot cm^{-1} \cdot K^{-1}$)
Cupper thermal conductivity	k_{Cu}	4.00	($W \cdot cm^{-1} \cdot K^{-1}$)
Thermal conductivity of the coolant fluid	k_{H_2O}	0.597 E-2	($W \cdot cm^{-1} \cdot K^{-1}$)
Specific heat of the coolant fluid	C_p	4.18	($J \cdot cm^{-3} \cdot K^{-1}$)
Average linear velocity of coolant	ν	2627	$cm \cdot s^{-1}$
Kinematic viscosity of the coolant fluid	μ	1 E-2	$cm^2 \cdot s^{-1}$
Temperature at the vacuum-rhodium interface	T_m	500	°C
Temperature of the coolant fluid	T_w	20	°C
Calculated values			
Quantity	Symbol	Value	Dimension
Thickness of the Rh layer	a	48 E-4	cm
Thickness of the second layer	b	8 E-4	cm
Cross-sectional area of the coolant channel	O	0.317	cm^2
Perimeter of the coolant channel	L	4.95	cm
Hydraulic equivalent diameter	D_e	0.256	cm
Heat transfer coefficient	h	8.5	($W \cdot cm^{-2} \cdot K^{-1}$)
Heat production rate in the Rh layer	Q_{Rh}	13 I	(W)
Heat production rate in the Cu layer	Q_{Cu}	5 I	(W)
Heat production rate	Q_t	18 I	(W)

(20°C). Observation of neither crack formation nor peeling off the rhodium layers indicated a good adhesion for the purpose.

By taking into account all the concepts in previous section, Eq. (4) can be rewritten as follows:

$$Q_{\mathrm{Rh}} = (E_{P1} - E_{P2}) \cdot I = 13\,I, \quad Q_{\mathrm{Cu}} = E_{P2} \cdot I = 5\,I, \quad Q_t = E_{P1} \cdot I = 18\,I$$

$$T_m - T_w = \left(\frac{(E_{P1}-E_{P2})\cdot I}{2k_{\mathrm{Rh}}}\cdot\frac{a}{s}\right) + \left(\frac{(E_{P1}-E_{P2})\cdot I}{k_{\mathrm{Cu}}}\cdot\frac{b}{s} + \frac{E_{P2}\cdot I}{2k_{\mathrm{Cu}}}\cdot\frac{b}{s}\right) + \left(\frac{E_{P1}\cdot I}{k_{\mathrm{Cu}}}\cdot\frac{c}{s}\right) + \left(\frac{E_{P1}\cdot I}{h\cdot s}\right) \tag{11}$$

where E_{P1} and E_{P2} are the entrance and outlet proton energy in the first layer, respectively,

$$T_m - T_w = (T_m - T_i) + (T_i - T_b) + (T_b - T_c) + (T_c - T_w) \tag{12}$$

Replacing the parameters from Table 1 in Eq. (11), results in 2100 μA for a maximum proton current at 500°C temperature, which is the temperature of thermal shock test. Different quantities of temperature differences in layers have been obtained from the above equation and according to Table 2. The maximum tolerated current density for target equals:

$$i_m = \frac{I_m}{s} = \frac{2100}{1169} = 1.796\ \mu\mathrm{A/mm}^2 \tag{13}$$

Table 2. Calculated temperature decrements over Rh target

I (μA)	$T_m - T_i$	$T_i - T_b$	$T_b - T_c$	$T_c - T_w$	$T_m - T_w$
100	0.18	0.03	3.85	18.12	22.17
200	0.36	0.05	7.70	36.23	44.34
300	0.53	0.08	11.55	54.35	66.51
400	0.71	0.11	15.40	72.46	88.68
500	0.89	0.13	19.25	90.58	110.84
600	1.07	0.16	23.10	108.69	133.01
700	1.25	0.19	26.95	126.81	155.18
800	1.42	0.21	30.80	144.92	177.35
900	1.60	0.24	34.64	163.04	199.52
1000	1.78	0.27	38.49	181.15	221.69
1100	1.96	0.29	42.34	199.27	243.86
1200	2.14	0.32	46.19	217.38	266.03
1300	2.31	0.34	50.04	235.50	288.20
1400	2.49	0.37	53.89	253.61	310.36
1500	2.67	0.40	57.74	271.73	332.53
1600	2.85	0.42	61.59	289.84	354.70
1700	3.02	0.45	65.44	307.96	376.87
1800	3.20	0.48	69.29	326.07	399.04
1900	3.38	0.50	73.14	344.19	421.21
2000	3.56	0.53	76.99	362.30	443.38
2100	3.74	0.56	80.84	380.42	465.55
2200	3.914	0.58	84.69	398.53	487.72

The effective parameters on maximum current target

Effect of rhodium carrier material

In a previous section, copper with 4.00 ($\mathrm{W \cdot cm^{-1} \cdot K^{-1}}$) thermal conductivity was considered as rhodium carrier. If silver is used instead of copper, the maximum allowable target current will increase a little to k_{Ag} = 4.30 ($\mathrm{W \cdot cm^{-1} \cdot K^{-1}}$), but because of its high price, there is no advantage from the economical point of view.

Effect of coolant channel shape

Calculating heat transfer coefficient was done by the convection method in the case of coolant channel with fins. The heat transfer coefficients obtained were 5.14 and 8.04 $\mathrm{W \cdot cm^{-2} \cdot K^{-1}}$ by eliminating fins and decreasing the effective depth of channel to 2.5 mm with no fins, respectively. Comparison of the recent amount of h (= 8.5) with quantities of h for carrier with no fins, results in these cases, h decreases by 40% and 5%, respectively. Decreasing the heat transfer coefficient allows decreasing the beam current.

Effect of coolant flow rate

If the flow rate is increased (e.g. f = 1666 $\mathrm{cm^3 \cdot s^{-1}}$), for the carrier with no fins:

$$\nu = \frac{f}{O} = \frac{1666}{0.300} = 5553\ \mathrm{cm/s} \tag{14}$$

$$R_e = \frac{D_e \cdot \nu \cdot \rho}{\mu} = \frac{0.413 \times 5553 \times 1}{1 \times 10^{-2}} = 229{,}398 \tag{15}$$

and Prandtl number is equal to 7, then the heat transfer coefficient will be 14 $\mathrm{W \cdot cm^{-2} \cdot K^{-1}}$. So, the beam current can be improved.

Conclusion

High beam current (I > 500 μA) irradiations can be used to produce the ^{103}Pd radionuclide from a Rh target. The relation Eq. (11) shows that decreasing the thickness of layer c has a positive effect on a proton current increase. The effect of channel shape in the case of with or without fins, influences the parameter h; increasing the heat transfer coefficient allows increasing the current. Also increasing the coolant flow rate has a positive effect on the parameter h.

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