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Measurements of ^{186}Re production cross section induced by deuterons on ^{nat}W target at ARRONAX facility

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ABSTRACT

Introduction: The ARRONAX cyclotron, acronym for "Accelerator for Research in Radiochemistry and Oncology at Nantes Atlantique" is a new facility installed in Nantes, France. A dedicated program has been launched on production of innovative radioisotopes for PET imaging and for β− and α targeted radiotherapy using protons or α particles. Since the accelerator is also able to deliver deuteron beams up to 35 MeV, we have reconsidered the possibility of using them to produce medical isotopes. Indeed, in some cases, the use of deuterons allows higher production yield than protons.
Methods: ^{186}Re is a β− emitter which has chemical properties close to the widely used ^{99m}Tc and has been used in clinical trials for palliation of painful bone metastases resulting from prostate and breast cancer. ^{186}Re production cross section has been measured between 9 and 23 MeV using the ARRONAX deuteron beam and the stacked-foil technique.
A novelty in our work is the use of a monitor foil behind each ^{nat}W target foil in order to record efficiently the deuteron incident flux and energies all over the stack relying on the International Atomic Energy Agency (IAEA) recommended cross section of the ^{nat}Ti(d,x)^{48}V reaction. Since a good optimization process is supposed to find the best compromise between production yield and purity of the final product, isotope of interest and contaminants created during irradiation are measured using gamma spectrometry.
Results: Our new sets of data are presented and compared with the existing ones and with results given by the TALYS code calculations. The thick target yield (TTY) has been calculated after the fit of our experimental values and compared with the IAEA recommended ones.
Conclusions: Presented values are in good agreement with existing data. The deuteron production route is clearly the best choice with a TTY of 7.8 MB/μAh at 30 MeV compared to 2.4 MBq/μAh for proton as projectile at the same energy. The TALYS code gives satisfactory results for ^{183,186}Re isotopes.

1. Introduction

The targeted radionuclide therapy is one modality to treat cancer which consists in binding a radioactive isotope to a vector in order to target and then to destroy tumour cells. The choice of the isotope to be used depends on the characteristics of the targeted tumour: α emitters will be well suited for microscopic disease whereas β− emitter will be used for millimetric tumour. Many isotopes are considered for such application. Among them, ^{186}Re could be advantageously produced using deuteron as projectile. ^{186}Re ($T_{1/2}$ = 3.7 days), is a β− emitter which has been used in clinical trials for palliation of painful bone metastases resulting from prostate and breast cancer [1].

^{186}Re is mainly produced in nuclear reactor using an ^{185}Re enriched target [2–4] but also aluminum perrhenate target [5]. This technique leads to a low specific activity of ^{186}Re. An alternative is to use other projectiles than neutrons, other targets and optimize the production. That can be done with cyclotrons able to accelerate protons or deuterons and with tungsten target.

Several cross section measurements have been made, since 1966, using proton or deuteron as projectile on a tungsten target [6]. Previous data show that deuterons as projectile are advantageous compared to protons, since it gives cross section values five times higher. As there are some disagreements between the existing series, this study aims to get additional data to better constrain the experimental trend. In both cases, our new sets determined via the stacked-foil technique [7] are compared with the existing experimental data and with TALYS [8] code calculations made using default parameters.

2. Set-up and data measurements

Several stacks of natural tungsten have been irradiated at the ARRONAX cyclotron [9], in the AX hall devoted to experiments in physics, radiolysis and radiobiology. The stacks were placed in air, on

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an irradiation station called Nice-III. The beam line is closed using a 75 μm thick kapton foil and the stack was located about 7 cm downstream. For the determination of ^{186}Re cross section, where energy threshold is low (3.6 MeV), deuteron beams at 16.4 MeV have been used. One high energy data point, around 22 MeV, has been obtained by putting a natural tungsten foil at the end of a ^{232}Th stack (devoted to ^{230}Pa cross section measurements) irradiated at 30 MeV. All foils were purchased from Goodfellow® (France) with high purity (more than 99.6%). Each thin foil has been weighed before irradiation using an accurate scale ($\pm 10^{-5}$ g) and scanned, to precisely determine the thickness. Titanium monitor foils have been placed behind each target foil, to record the particle flux all along the stack through the ^{nat}Ti(d,x)^{48}V reaction, as suggested by the IAEA [10].

In each foil, the ^{48}V activity value has been determined after the complete decay of ^{48}Sc ($T_{1/2} = 43.67$ h). Indeed, ^{48}Sc is also produced in the titanium targets and emits two same gamma lines than ^{48}V. Nuclear data [6] associated to ^{48}V are summarized in Table 1. In addition to monitor foils, a Faraday cup was placed after the stack to collect charges and control the intensity during the irradiation. The incident beam energy was fixed by the setting parameters of the cyclotron. The energy through each thin foil was determined in the middle of the foil using the SRIM software [11]. Energy losses in the kapton foil and air were taken into account. Typical irradiations were carried out with a beam intensity of about 100 nA during 30 minutes. Each target foil was separated from the stack and, after some cooling time, counting measurements were performed using a high purity germanium detector with low-background lead and copper shield from Canberra (France). Gamma spectra were recorded in a suitable geometry previously calibrated with standard ^{57,60}Co and ^{152}Eu gamma sources. The full widths at half maximum were 1.04 keV at 122 keV (^{57}Co ray) and 1.97 keV at 1332 keV (^{60}Co ray). The activity values of the produced radioisotopes were derived from the spectra and the nuclear decay data [12] given in Table 2, using the Fitzpeak spectroscopy software [13]. The dead time during the counting was always kept below 10% in order to reduce the effect of sum peaks.

Production cross section values can be determined from the activation formula (1) with the appropriate projectile flux.

$$\sigma = \frac{\chi.Act.A}{\Phi.N_a.\rho.ef\left(1-e^{-\lambda.t}\right)} \tag{1}$$

In this equation, the production cross section σ of a radioisotope depends on its measured activity (Act), its decay constant (λ), the target thickness (e_f), its purity (χ), its atomic number (A), its density (ρ), the irradiation duration (t) and the projectile flux (Φ). In our experiment, each target foil received the same projectile flux as the following monitor foil. It is then easier to use the relative Eq. (2) in which the knowledge of the projectile flux is no longer necessary. In this equation, the prime parameters are associated to ^{48}V monitor while the others relate to the rhenium isotopes.

$$\sigma = \sigma' \frac{\chi.Act.A.\rho'.e'_f.\left(1-e^{-\lambda'.t}\right)}{\chi' Act'.A'.\rho.e_f\left(1-e^{-\lambda.t}\right)} \tag{2}$$

To determine the activity associated to each radioisotope of interest, all the target and monitor foils were counted twice with an interval of 2 weeks and during more than 24 hours. The cross section uncertainty is estimated with a propagation error calculation. Since all the parameters of Eq. (2) are independent, the total error is expressed as a quadratic sum (3). The main error sources come from the recommended cross section (around 12%), ^{186,183}Re activities (up to 10%), ^{48}V activity (less than 2%) and thickness of foil (around 1%). The contribution of the irradiation time uncertainty is not significant and has been neglected.

Table 1
Vanadium-48 half-life and main γ rays.

Radioisotope	$T_{1/2}$ (days)	Eγ (keV)	Iγ (%)
^{48}V	15.9735 (25)	944.104	7.870 (7)
		983.517	99.98 (4)
		1312.096	98.2 (3)

Table 2
Properties of two rhenium radioisotopes produced from natural tungsten target.

Radioisotope	$T_{1/2}$ (days)	Eγ (keV)	Iγ (%)	Contributing reactions	$E_{threshold}$ (MeV)
^{186}Re	3.7183 (11)	137.157	9.42 (6)	^{186}W(d,2n)	3.626
^{183}Re	70.0 (11)	162.3219	23.3 (4)	^{182}W(d,n)	0.0
		208.8057	2.95 (5)	^{183}W(d,2n)	3.602
		292.7238	3.05 (16)	^{184}W(d,3n)	11.095
				^{186}W(d,5n)	24.180

$$\frac{\Delta\sigma}{\sigma} = \sqrt{\left(\frac{\Delta\sigma'}{\sigma'}\right)^2 + \left(\frac{\Delta Act'}{Act}\right)^2 + \left(\frac{\Delta Act'}{Act'}\right)^2 + \left(\frac{\Delta e}{e}\right)^2 + \left(\frac{\Delta e'}{e'}\right)^2} \tag{3}$$

3. Results

In this section, we present the ^{186}Re production cross section induced by deuteron on a natural tungsten target. All the contaminants produced during this experiment have been measured. ^{183}Re production cross section is also presented. Indeed, due to its long half-life ($T_{1/2} = 70$ d), ^{183}Re strongly affects the specific activity of the final product.

3.1. Production of ^{186}Re

The ^{186}Re radioisotope has a half-life of $T_{1/2} = 3.7183$ days and decreases at 92.53% by β− to ^{186}Os (stable) and at 7.47% by EC to ^{186}W (stable). Its gamma line, $E_\gamma = 137.157$ keV ($I_\gamma = 9.47\%$), coming from the β− decay, is used to measure the activity. In ^{nat}W, ^{186}Re can only come from the ^{186}W, the second most abundant isotope (28.6%). Our new data set is presented in Fig. 1 as full circles.

These results are very close to the reference [14] values in the range 7–12 MeV and follow the reference [15] trend up to 17 MeV. Only references [15] and [16] have contributed with higher energy beams and our result at 22 MeV is in agreement with their values. In

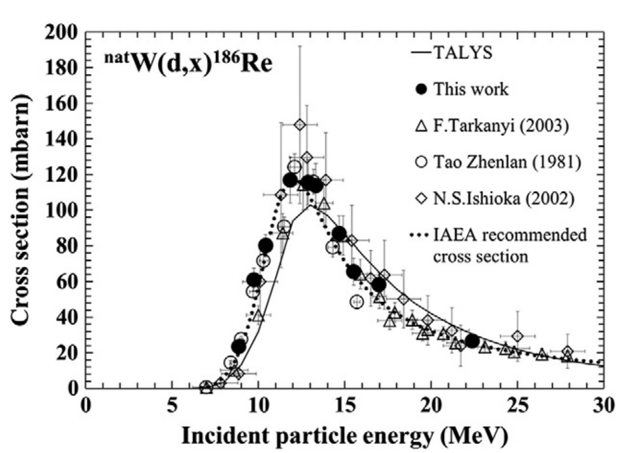

Fig. 1. Production cross section of ^{nat}W(d,xn)^{186}Re.

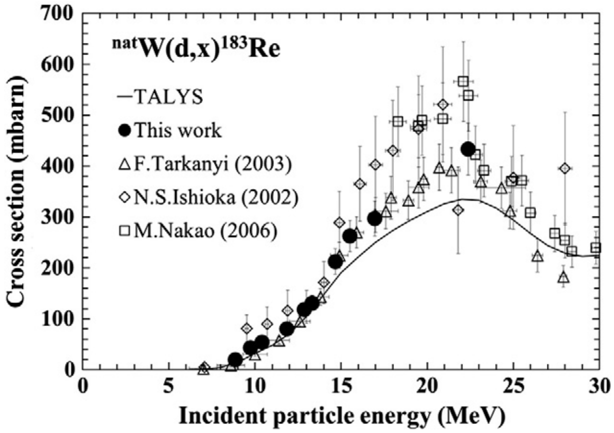

Fig. 2. Production cross section of $^{nat}W(d,xn)^{183}Re$.

this case, the TALYS code gives satisfactory results, even if the shape is slightly smaller below 12 MeV.

3.2. Production of ^{183}Re

The decay of the ^{183}Re contaminant is followed by three main gamma radiations presented in Table 2. It can be produced by four of the tungsten isotopes constituting the natural target. Our results are plotted in Fig. 2 with three other data sets.

These data show that the maximum cross section is around 20 MeV but with a different magnitude depending on the series. Our new values are coherent with the trend of reference [15] up to 17 MeV and around 22 MeV. New results above 17 MeV are needed to better discriminate between the different data sets previously published [15–17]. TALYS code gives good results below 15 MeV and above 25 MeV. Between these two energies, the different experiments give values 16%–35% higher than TALYS.

4. Conclusions

In this work, new data sets concerning $^{183,186}Re$ production cross sections induced by deuterons have been obtained. Presented values are in good agreement with the few existing data.

With a TTY (Fig. 3) of 7.8 MBq/μAh at 30 MeV, the deuteron production route is clearly the best choice. Indeed, with protons [10,18], the recommended TTY [10] is equal to 2.4 MBq/μAh at the same energy. Looking at the recommended TTY using deuterons plotted in Fig. 3, we can see that our experimental yield is slightly higher. This is mainly linked with the IAEA recommended cross section fit (plotted as a dot line in Fig. 1) showing a peak slightly thinner and lower than our points.

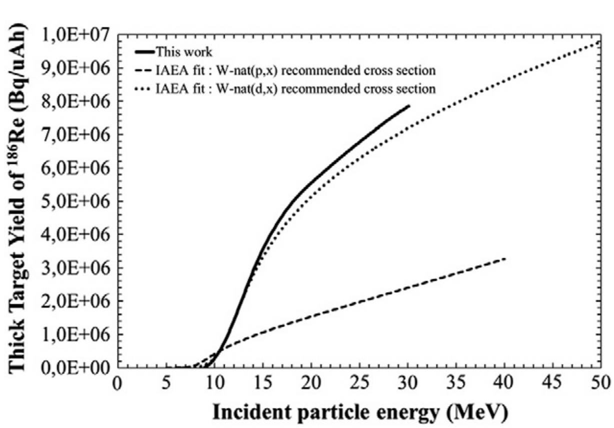

Fig. 3. ^{186}Re thick target yield for proton and deuteron production routes.

All the contaminants created during irradiation were measured since a good optimization process is supposed to find the best compromise between production yield and purity of the final product. An easy way to avoid the production of contaminants is to use an enriched ^{186}W target irradiated with deuteron beams. When using this tungsten isotope as target, only the ^{186}Re and $^{184m,g}Re$ could be produced. With a deuteron energy just below the $^{186}W(d,4n)^{184m,g}Re$ reaction threshold, 17.6 MeV, the ^{186}Re specific activity will be the greatest possible with a TTY of 16.8 MBq/μAh to be compared with the IAEA recommended value of 15.4 MBq/μAh with deuterons and 4.6 MBq/μAh with a proton beam at the same energy.

Comparisons with the TALYS code have been performed using the default parameters. For these two rhenium isotopes the TALYS code gives satisfactory results.

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