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

Measurements of  $^{186}\text{Re}$  production cross section

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### Keywords:

Rhenium-186, cross section, stacked-foil, deuteron, technecium-99m, ARRONAX

### 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  $\beta$ - and  $\alpha$  targeted radiotherapy using protons or  $\alpha$  particles. Since the accelerator is also able to deliver deuteron beams up to 35 MeV, we have reconsidered the possibility to use them to produce medical isotopes. Indeed, in some cases, the use of deuterons allows higher production yield than protons.

#### Methods:

$^{186}\text{Re}$  is a  $\beta$ - emitter which has chemical properties close to the widely used  $^{99m}\text{Tc}$  and has been used in clinical trials for palliation of painful bone metastases resulting from prostate and breast cancer.  $^{186}\text{Re}$  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  $^{nat}\text{W}$  target foil in order to record efficiently the deuteron incident flux and energies all over the stack relying on the International Atomic Energy

30 Agency (IAEA) recommended cross section of the  $^{nat}\text{Ti}(d,x)^{48}\text{V}$  reaction. Since a good optimization process is  
31 supposed to find the best compromise between production yield and purity of the final product, isotope of  
32 interest and contaminants created during irradiation are measured using gamma spectrometry.

33 Results:

34 Our new sets of data are presented and compared with the existing ones and with results given by the  
35 TALYS code calculations. The thick target yield (TTY) has been calculated after the fit of our experimental values  
36 and compared with the IAEA recommended ones.

37 Conclusions:

38 Presented values are in good agreement with existing data. The deuteron production route is clearly  
39 the best choice with a TTY of 7.8 MB/ $\mu\text{Ah}$  at 30 MeV compared to 2.4 MBq/ $\mu\text{Ah}$  for proton as projectile at the  
40 same energy. The TALYS code gives satisfactory results for  $^{183,186}\text{Re}$  isotopes.

41 **Paper:**

42 1 Introduction

43 The targeted radionuclide therapy is one modality to treat cancer which consists in binding a  
44 radioactive isotope to a vector in order to target and then to destroy tumour cells. The choice of the isotope to  
45 be used depends on the characteristics of the targeted tumour:  $\alpha$  emitters will be well suited for microscopic  
46 disease whereas  $\beta^-$  emitter will be used for millimetric tumour. Many isotopes are considered for such  
47 application. Among them,  $^{186}\text{Re}$  could be advantageously produced using deuteron as projectile.  $^{186}\text{Re}$  ( $T_{1/2} =$   
48 3.7 days), is a  $\beta^-$  emitter which has been used in clinical trials for palliation of painful bone metastases resulting  
49 from prostate and breast cancer [1].

50  $^{186}\text{Re}$  is mainly produced in nuclear reactor using an  $^{185}\text{Re}$  enriched target [2, 3, 4] but also aluminum  
51 perrhenate target [5]. This technique leads to a low specific activity of  $^{186}\text{Re}$ . An alternative is to use other  
52 projectiles than neutrons, other targets and optimize the production. That can be done with cyclotrons able to  
53 accelerate protons or deuterons and with tungsten target.

54 Several cross section measurements have been made, since 1966, using proton or deuteron as  
55 projectile on a tungsten target [6]. Previous data show that deuterons as projectile are advantageous  
56 compared to protons, since it gives cross section values five times higher. As there are some disagreements  
57 between the existing series, this study aims to get additional data to better constrain the experimental trend.

58 In both cases, our new sets determined via the stacked-foil technique [7] are compared with the existing  
59 experimental data and with TALYS [8] code calculations made using default parameters.

## 60 2 Set-up and data measurements

61 Several stacks of natural tungsten have been irradiated at the ARRONAX cyclotron [9], in the AX hall  
62 devoted to experiments in physics, radiolysis and radiobiology. The stacks were placed in air, on an irradiation  
63 station called Nice-III. The beam line is closed using a 75  $\mu\text{m}$  thick kapton foil and the stack was located about 7  
64 cm downstream. For the determination of  $^{186}\text{Re}$  cross section, which energy threshold is low (3.6 MeV),  
65 deuteron beams at 16.4 MeV have been used. One high energy data point, around 22 MeV, has been obtained  
66 by putting a natural tungsten foil at the end of a  $^{232}\text{Th}$  stack (devoted to  $^{230}\text{Pa}$  cross section measurements)  
67 irradiated at 30 MeV. All foils were purchased from Goodfellow<sup>®</sup> (France) with high purity (more than 99.6%).  
68 Each thin foil has been weighed before irradiation using an accurate scale ( $\pm 10^{-5}$  g) and scanned, to precisely  
69 determine the thickness. Titanium monitor foils have been placed behind each target foil, to record the particle  
70 flux all along the stack through the  $^{\text{nat}}\text{Ti}(d,x)^{48}\text{V}$  reaction, as suggested by the IAEA [10].

71 In each foil, the  $^{48}\text{V}$  activity value has been determined after the complete decay of  $^{48}\text{Sc}$  ( $T_{1/2} = 43.67$   
72 h). Indeed,  $^{48}\text{Sc}$  is also produced in the titanium targets and emits two same gamma lines than  $^{48}\text{V}$ . Nuclear data  
73 [6] associated to  $^{48}\text{V}$  are summarized in Table 1. In addition to monitor foils, a Faraday cup was placed after the  
74 stack to collect charges and control the intensity during the irradiation. The incident beam energy was fixed by  
75 the setting parameters of the cyclotron. The energy through each thin foil was determined in the middle of the  
76 foil using the SRIM software [11]. Energy losses in the kapton foil and air were taken into account. Typical  
77 irradiations were carried out with a beam intensity of about 100 nA during 30 minutes. Each target foil was  
78 separated from the stack and, after some cooling time, counting measurements were performed using a high  
79 purity germanium detector with low-background lead and copper shield from Canberra (France). Gamma  
80 spectra were recorded in a suitable geometry previously calibrated with standard  $^{57,60}\text{Co}$  and  $^{152}\text{Eu}$  gamma  
81 sources. The full widths at half maximum were 1.04 keV at 122 keV ( $^{57}\text{Co}$  ray) and 1.97 keV at 1332 keV ( $^{60}\text{Co}$   
82 ray). The activity values of the produced radioisotopes were derived from the spectra and the nuclear decay  
83 data [12] given in Table 2, using the Fitzpeak spectroscopy software [13]. The dead time during the counting  
84 was always kept below 10% in order to reduce the effect of sum peaks.

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

$$87 \quad \sigma = \frac{\chi \cdot Act \cdot A}{\Phi \cdot N_a \cdot \rho \cdot e_f (1 - e^{-\lambda \cdot t})} \quad (1)$$

88 In this equation, the production cross section  $\sigma$  of a radioisotope depends on its measured activity (Act), its  
89 decay constant ( $\lambda$ ), the target thickness ( $e_f$ ), its purity ( $\chi$ ), its atomic number (A), its density ( $\rho$ ), the irradiation  
90 duration (t) and the projectile flux ( $\Phi$ ). In our experiment, each target foil received the same projectile flux as  
91 the following monitor foil. It is then easier to use the relative equation (2) in which the knowledge of the  
92 projectile flux is no longer necessary. In this equation, the prime parameters are associated to  $^{48}\text{V}$  monitor  
93 while the others relate to the rhenium isotopes.

$$94 \quad \sigma = \sigma' \frac{\chi \cdot Act \cdot A \cdot \rho' \cdot e_f' \cdot (1 - e^{-\lambda' \cdot t})}{\chi' \cdot Act' \cdot A' \cdot \rho \cdot e_f (1 - e^{-\lambda \cdot t})} \quad (2)$$

95 To determine the activity associated to each radioisotope of interest, all the target and monitor foils  
96 were counted twice with an interval of 2 weeks and during more than 24 hours. The cross section uncertainty is  
97 estimated with a propagation error calculation. Since all the parameters of equation (2) are independent, the  
98 total error is expressed as a quadratic sum (3). The main error sources come from the recommended cross  
99 section (around 12%),  $^{186,183}\text{Re}$  activities (up to 10%),  $^{48}\text{V}$  activity (less than 2%) and thickness of foil (around  
100 1%). The contribution of the irradiation time uncertainty is not significant and has been neglected.

$$101 \quad \frac{\Delta\sigma}{\sigma} = \sqrt{\left(\frac{\Delta\sigma'}{\sigma'}\right)^2 + \left(\frac{\Delta Act}{Act}\right)^2 + \left(\frac{\Delta Act'}{Act'}\right)^2 + \left(\frac{\Delta e}{e}\right)^2 + \left(\frac{\Delta e_f}{e_f}\right)^2} \quad (3)$$

## 102 3 Results

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

107

### 108 3.1 Production of $^{186}\text{Re}$

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

113            These results are very close to the reference [14] values in the range 7 to 12 MeV and follow the  
114 reference [15] trend up to 17 MeV. Only references [15] and [16] have contributed with higher energy beams  
115 and our result at 22 MeV is in agreement with their values. In this case, the TALYS code gives satisfactory  
116 results, even if the shape is slightly smaller below 12 MeV.

### 117 3.2 Production of <sup>183</sup>Re

118            The decay of the <sup>183</sup>Re contaminant is followed by three main gamma radiations presented in Table 2.  
119 It can be produced by four of the tungsten isotopes constituting the natural target. Our results are plotted in  
120 Figure 2 with three other data sets.

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

### 126 4 Conclusions

127            In this work, new data sets concerning <sup>183,186</sup>Re production cross sections induced by deuterons have  
128 been obtained. Presented values are in good agreement with the few existing data.

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

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

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

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148

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192 **Illustrations and tables:**

Radioisotope	$T_{1/2}$ (days)	$E_{\gamma}$ (keV)	$I_{\gamma}$ (%)
$^{48}\text{V}$	15.9735 (25)	944.104	7.870 (7)
		983.517	99.98 (4)
		1312.096	98.2 (3)

193 Table 1: Vanadium-48 half-life and main  $\gamma$  rays

194

Radioisotope	T <sub>1/2</sub> (days)	E <sub>γ</sub> (keV)	I <sub>γ</sub> (%)	Contributing reactions	E <sub>threshold</sub> (MeV)
<sup>186</sup> Re	3.7183 (11)	137.157	9.42 (6)	<sup>186</sup> W(d,2n)	3.626
<sup>183</sup> Re	70.0 (11)	162.3219	23.3 (4)	<sup>182</sup> W(d,n)	0.0
		208.8057	2.95 (5)	<sup>183</sup> W(d,2n)	3.602
		292.7238	3.05 (16)	<sup>184</sup> W(d,3n)	11.095
				<sup>186</sup> W(d,5n)	24.180

195

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

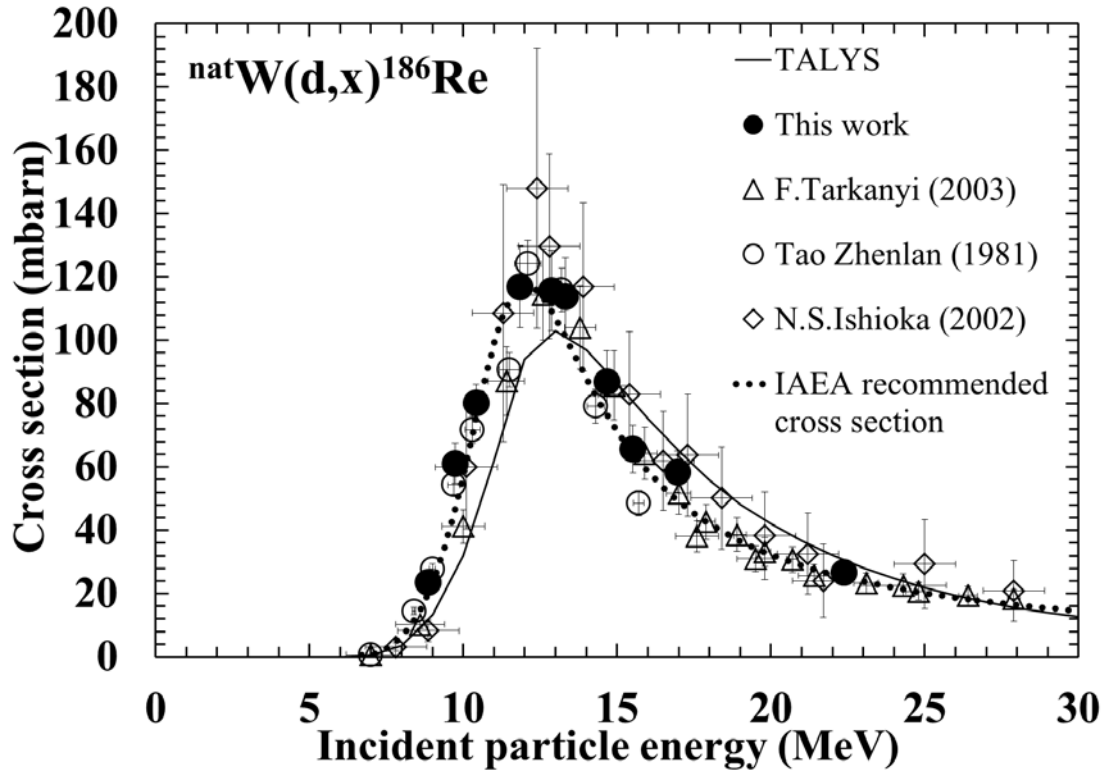


Figure 1: Production cross section of  $^{nat}\text{W}(d,x)^{186}\text{Re}$

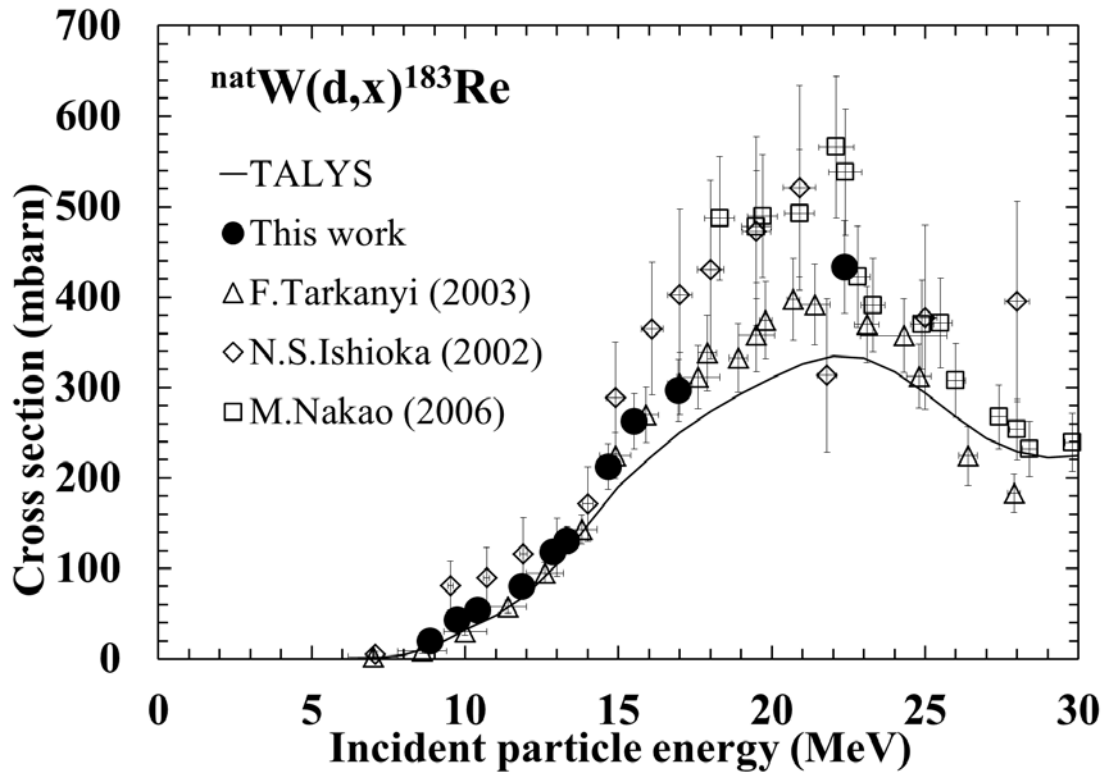


Figure 2: Production cross section of  ${}^{\text{nat}}\text{W}(\text{d},\text{x}){}^{183}\text{Re}$

196

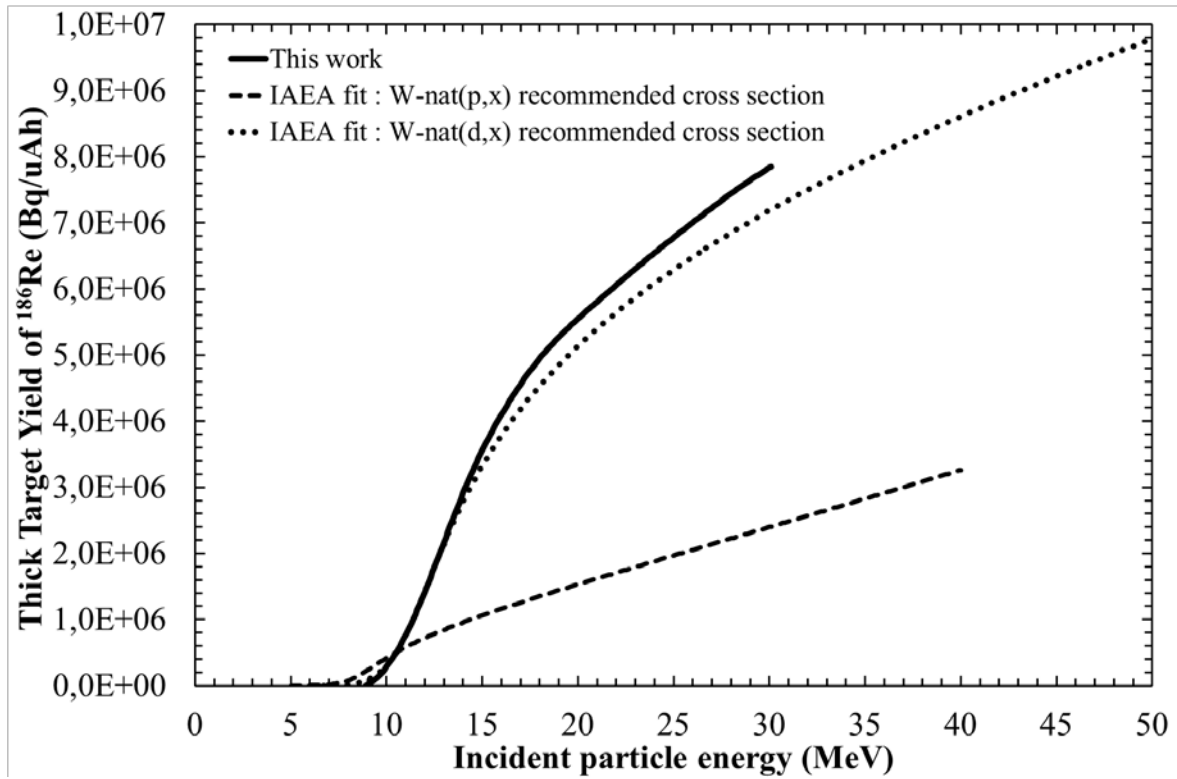


Figure 3:  ${}^{186}\text{Re}$  thick target yield for proton and deuteron production routes

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199