

Production cross section measurements of the $natPd(\alpha, x)$ 111Ag reaction

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► To cite this version:

Laurine Puren, Arnaud Guertin, Etienne Nigron, Ferid Haddad, Vincent Métivier. Production cross section measurements of the nat $Pd(\alpha,x)111Ag$ reaction. Tumour targeting, Imaging, RadiotherapieS, Oct 2024, Erquy, FR, France. hal-04780637

HAL Id: hal-04780637 https://hal.science/hal-04780637v1

Submitted on 13 Nov 2024

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Physics of Radiation InteractionS with Matter and Applications





Production cross section measurements of the ^{nat}Pd(α,x)¹¹¹Ag reaction

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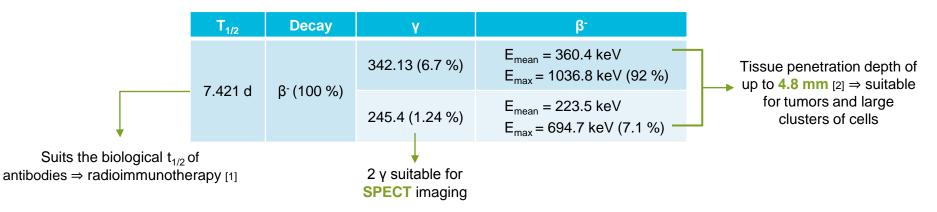
Tumour targeting, Imaging, Radiotherapies workshop

10th October 2024

Context and motivations

 \Box Low toxicity of silver in humans \Rightarrow already used in several pharmaceuticals [1]

 \Box ¹¹¹Ag can be used for β - therapy:



□ The appeal for ¹¹¹Ag:

- > Dose assessment and biodistribution monitoring with SPECT imaging
- Radiosynovectomy : treatment for arthritis. [2]

[1] K. M. El-Azony *et al.*, 'Advantages and disadvantages of nuclear reactions used in reactors or cyclotrons, in addition to a theoretical study based on photodisintegration on natural indium for 111Ag production' Feb. 2022

[2] S. Chattopadhyay et al., 'Preparation and evaluation of a new radiopharmaceutical for radiosynovectomy, 111Ag-labelled hydroxyapatite (HA) particles', Mar. 2008

Context and motivations

Production routes of ¹¹¹Ag :

¹⁰⁵ ln 5.07 min ε β ⁺ = 100 %	¹⁰⁶ ln 6.23 min ε β ⁺ = 100 %	¹⁰⁷ In 32.4 min ε β ⁺ = 100 %	¹⁰⁸ ln 58 min ε β ⁺ = 100 %	¹⁰⁹ ln 4.154 h ε β ⁺ = 100 %	¹¹⁰ ln 4.9 h ε β ⁺ = 100 %	¹¹¹ ln 2.80 d ε β ⁺ = 100 %	¹¹² In 14.88 m ε β ⁺ = 62 % β ⁻ = 38 %	¹¹³ In STABLE 4.281 %	¹¹⁴ In 71.9 s β ⁻ = 99.5 % εβ ⁺ = 0.5 %	¹¹⁵ In 4.41e14 y 95.719% β [.] = 100 %
¹⁰⁴ Cd 57.8 min ε β ⁺ = 100 %	¹⁰⁵ Cd 55.4 min ε β ⁺ = 100 %	¹⁰⁶ Cd STABLE 1.245 %	¹⁰⁷ Cd 6.52 h ε β ⁺ = 100 %	¹⁰⁸ Cd STABLE 0.888%	¹⁰⁹ Cd 461.98 d ε = 100 %	¹¹⁰ Cd STABLE 12.47 %	¹¹¹ Cd STABLE 12.795 %	¹¹² Cd STABLE 24.109 %	¹¹³ Cd 8.04e15 y 12.227 % β ⁻ = 100 %	¹¹⁴ Cd STABLE 28.754 %
¹⁰³ Ag 65.8 min ε β ⁺ = 100 %	¹⁰⁴ Ag 69.3 min ε β ⁺ = 100 %	¹⁰⁵ Ag 41.29 d ε β ⁺ = 100 %	¹⁰⁶ Ag 23.96 m ε β ⁺ > 99 % β ⁻ < 1 %	¹⁰⁷ Ag STABLE 51.83 %	¹⁰⁸ Ag 2.39 m β ⁻ = 97.15 % ε β ⁺ = 2.85 %	¹⁰⁹ Ag STABLE 48.161 %	¹¹⁰ Ag 24.56 s β [.] = 100 %	¹¹¹ Ag 7.421 d β ⁻ = 100 %	¹¹² Ag 3.15 h β ⁻ = 100 %	¹¹³ Ag 5.37 h β [.] = 100 %
¹⁰² Pd STABLE 1.02 %	¹⁰³ Pd 17 d ε β ⁺ = 100 %	¹⁰⁴ Pd STABLE 11.14 %	¹⁰⁵ Pd STABLE 22.33 %	¹⁰⁶ Pd STABLE 27.33 %	¹⁰⁷ Pd 6.50e6 y β [.] = 100 %	¹⁰⁸ Pd STABLE 26.46 %	¹⁰⁹ Pd 13.437 h β ⁻ = 38.5 %	¹¹⁰ Pd STABLE 30.85 %	¹¹¹ Pd 23.6 min β [.] = 100 %	¹¹² Pd 21.027 h β ⁻ = 100 %

Neutron and photonuclear routes :

- $\label{eq:alpha} \square \ ^{110}\text{Pd}(n,\gamma)^{111}\text{Pd} \ (\beta^{-} \) \rightarrow \ ^{111}\text{Ag}$
- □ Isotope Separation On-Line technique (ISOL)
- \Box ^{nat}ln(γ ,x)¹¹¹Ag
- $\square \ ^{nat}Cd(\gamma,x)^{111}Ag$

Light-charged particle routes :

- □ ²³²Th(p,f)¹¹¹Ag (fission product)
- □ ^{nat}Pd(d,x)¹¹¹Ag, ¹¹⁰Pd(d,x)¹¹¹Ag
- $\Box ^{nat}Pd(\alpha,x)^{111}Ag, ^{110}Pd(\alpha,x)^{111}Ag, ^{108}Pd(\alpha,x)^{111}Ag$
 - Few experimental data available

Context and motivations

\Box a production routes of ¹¹¹Ag :

- Only two experimental datasets for ^{nat}Pd(α,x)¹¹¹Ag.
- Lack of data at high energy.

L, add data in the region of the peak and investigate what's happening after it.

Motivations

- 1. New measurements at high energy of ${}^{nat}Pd(\alpha,x)^{111}Ag$.
- 2. Comparison with the literature.
- 3. Comparison with the other production routes.

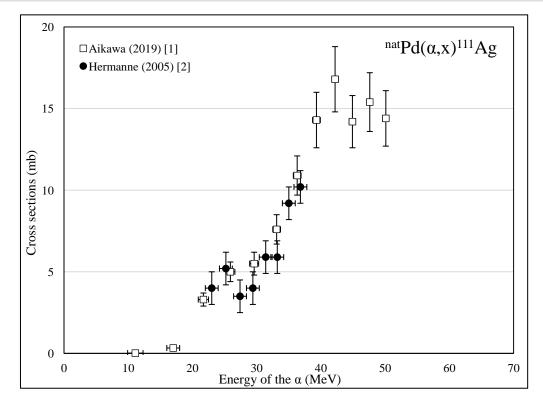


Figure 1 : Cross section measurements of the $^{nat}Pd(\alpha, x)^{111}Ag$ reaction

Method: Stacked-foils technique

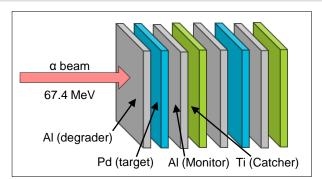
□ Stacked-foils technique: in one irradiation, several production cross sections can be measured.

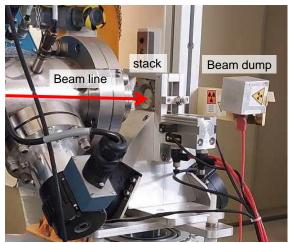
L One pattern composed : degrader, target, monitor + catcher

- **Given State and State and**
 - Alpha beam of 67.4 MeV
 - Monitor reactions : ${}^{27}AI(\alpha,x){}^{24}Na$, ${}^{27}AI(\alpha,x){}^{22}Na$

	1 st exp	2 nd exp	3 rd exp
Time of irradiation	1 h 30	2 h	2 h
Intensity of the beam	100 nA	150 nA	150 nA

Use of the γ-rays at 342 keV (6.7 %) and 245 keV (1.24 %) to extract ¹¹¹Ag activity with a **HPGe** detector.





<u>Figure 3:</u> Schematical representation of the stack and setup of the experiment



Results

Observations:

 8 measurements done in three different experiments: good consistency between the 1st and 3rd series.

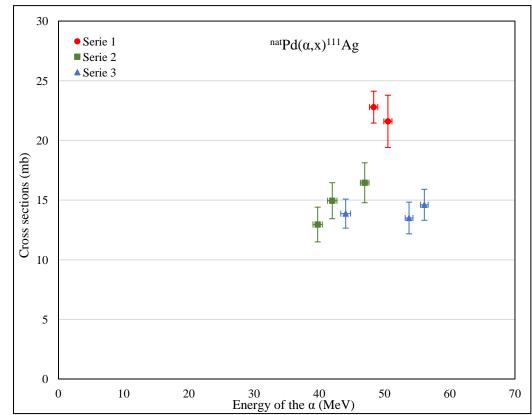


Figure 4: Cross section measurements of the ${}^{nat}Pd(\alpha,x)^{111}Ag$ reaction.

Results

Observations:

- 8 measurements done in three different experiments: good consistency between the 1st and 3rd series.
- The maximum cross section seems to be around 50 MeV
- Overall good agreement with the experimental values of Aikawa et al.
 - L, the points at 48 and 50.5 MeV (*serie 1*) seem questionable.

Make new measurements in the area around the peak to define it better.

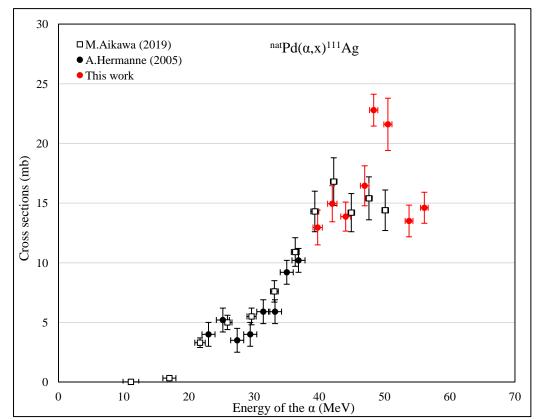


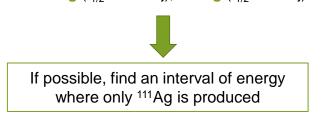
Figure 4: Cross section measurements of the ${}^{nat}Pd(\alpha,x)^{111}Ag$ reaction.

Results – Contaminants

Production of other silver isotopes:

¹⁰³ Ag 65.8 min ε β ⁺ = 100 %	¹⁰⁴ Ag 69.3 min ε β ⁺ = 100 %	¹⁰⁵ Ag 41.29 d ε β ⁺ = 100 %	¹⁰⁶ Ag 23.96 m ε β ⁺ > 99 % β ⁻ < 1 %	¹⁰⁷ Ag STABLE 51.83 %	¹⁰⁸ Ag 2.39 m β ⁻ = 97.15 % ε β ⁺ = 2.85 %		¹¹⁰ Ag 24.56 s β [.] = 100 %	¹¹¹ Ag 7.421 d β [.] = 100 %	¹¹² Ag 3.15 h β ⁻ = 100 %	¹¹³ Ag 5.37 h β ⁻ = 100 %
¹⁰² Pd STABLE 1.02 %	¹⁰³ Pd 17 d ε β ⁺ = 100 %	¹⁰⁴ Pd STABLE 11.14 %	¹⁰⁵ Pd STA BLE 22.33 %	¹⁰⁶ Pd STABLE 27.33 %	¹⁰⁷ Pd 6.50e6 y β [.] = 100 %	¹⁰⁸ Pd STABLE 26.46 %	¹⁰⁹ Pd 13.437 h β [.] = 38.5 %	¹¹⁰ Pd STABLE 30.85 %	¹¹¹ Pd 23.6 min β [.] = 100 %	¹¹² Pd 21.027 h β ⁻ = 100 %
	^{106m} Ag: $t_{1/2} = 8.28 \text{ j}$					^{110m} Ag: t _{1/2} = 249.83 j				

■ Most of the contaminants produced are short-lived \Rightarrow a cooling time of 1-2 days is enough for them to decay completely I, Not the case for the longer-lived ¹⁰⁵Ag (t_{1/2} = 41.29 j), ^{106m}Ag (t_{1/2} = 8.28 j) and ^{110m}Ag (t_{1/2} = 249.83 j)



Results – Contaminants

Observations:

٠

¹¹¹Ag and ^{110m}Ag have the lowest cross sections of all the long-lived silver isotopes.

l, more than 5 times lower than for 106m Ag and more than 10 times lower than 105 Ag.

- They are all produced in the same energy interval than ¹¹¹Ag.
- Ly their production is **unavoidable**.

□ Use of an **enriched target** of ¹⁰⁸Pd or ¹¹⁰Pd :

- Small production of ¹⁰⁵Ag and ^{106m}Ag.
- ¹¹¹Ag is **still** produced alongside ^{110m}Ag.

What about the other production routes ?

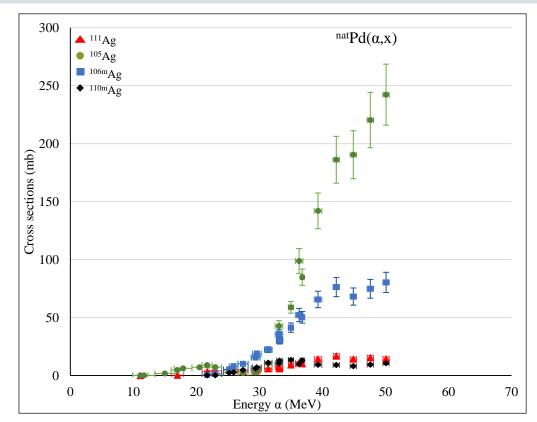


Figure 5: Cross section measurements of the $^{nat}Pd(\alpha,x)^{111}Ag$, ^{105}Ag , ^{106m}Ag , ^{110m}Ag reaction.

Results – Production routes

Deuteron route :

- Maximum cross section of ^{nat}Pd(d,x)¹¹¹Ag is at lower energy than with the alpha route : 10 MeV.
- The same contaminants : ^{105, 106m, 110m}Ag
 Low purity and low specific activity
- A theoretical production could be between 16 and 6 MeV.
 - L Target thickness : ≈ 225 μm
 - l, Yield at EOB : ≈ 1.21 MBq/µAh

Reactor route:

- Indirect production : $^{110}\text{Pd}(n,\gamma)^{111}\text{Pd}~(\beta^{-}~) \rightarrow \,^{111}\text{Ag}$
- · Contaminants : only short-lived silver isotopes
- High specific activity
- Production of 100 MBq of ¹¹¹Ag [1] :
 - L neutron flux : $3 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$
 - L 26 h of irradiation

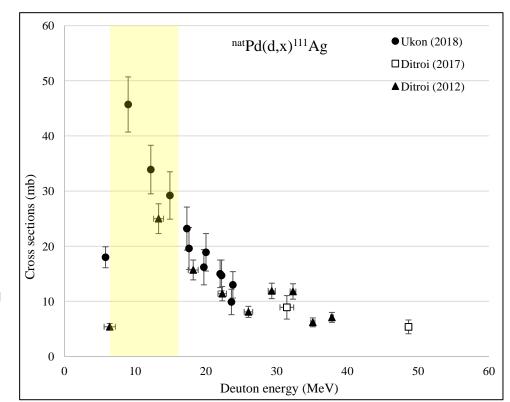
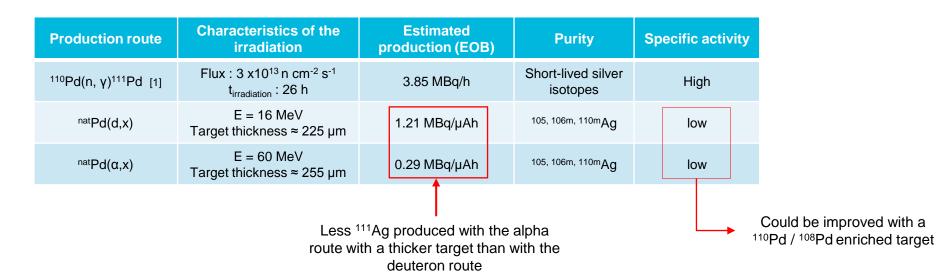


Figure 6: Cross section measurements of the $^{nat}Pd(d,x)$ reaction.

[1] R. Alberto et al. 'An improved method for the separation of 111Ag from irradiated natural palladium', Jul. 1992



The most promising route seems to be the neutron route.

[1] L. Morselli et al., 'Production and characterization of 111Ag radioisotope for medical use in a TRIGA Mark II nuclear research reactor', Jul. 2023

Conclusion

- New production cross section measurements of the reaction $^{nat}Pd(\alpha,x)^{111}Ag$ have been performed at the GIP Arronax at high energy:
 - \rightarrow Good agreement with the data of Aikawa *et al.*, apart from the points of my first experiment.
 - \rightarrow The maximum cross section seem to be around 50 MeV
- Comparison with the most accessible production routes:
 - \rightarrow low purity and specific activity with the alpha and deuteron routes
 - \rightarrow Good purity and high specific activity with the neutron route \Rightarrow the most promising
- **Outlook:**
 - → Measure again the peak area to understand the difference between the data from Aikawa my first experiment
 - \rightarrow Future experiments between 55 and 65 MeV to have experimental data after the peak
 - \rightarrow Extract the production cross sections of the silver contaminants (¹⁰⁵Ag, ^{106m}Ag and ^{110m}Ag)







THANK YOU FOR YOUR ATTENTION





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