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Studies of different production paths for ^{155}Tb using Gd targets : from target manufacturing to Tb/Gd separation

Nadia Audouin^c, Charlotte Duchemin^{b,c}, Roberto Formento-Cavaier^b, Arnaud Guertin^b, Etienne Nigrón^{b,c}, Férid Haddad^{b,c}, Nathalie Michel^b, **Thomas Sounalet^b**, Vanessa Rhoden^b and Yizheng Wang^{b,c}

^a CERN-MEDICIS

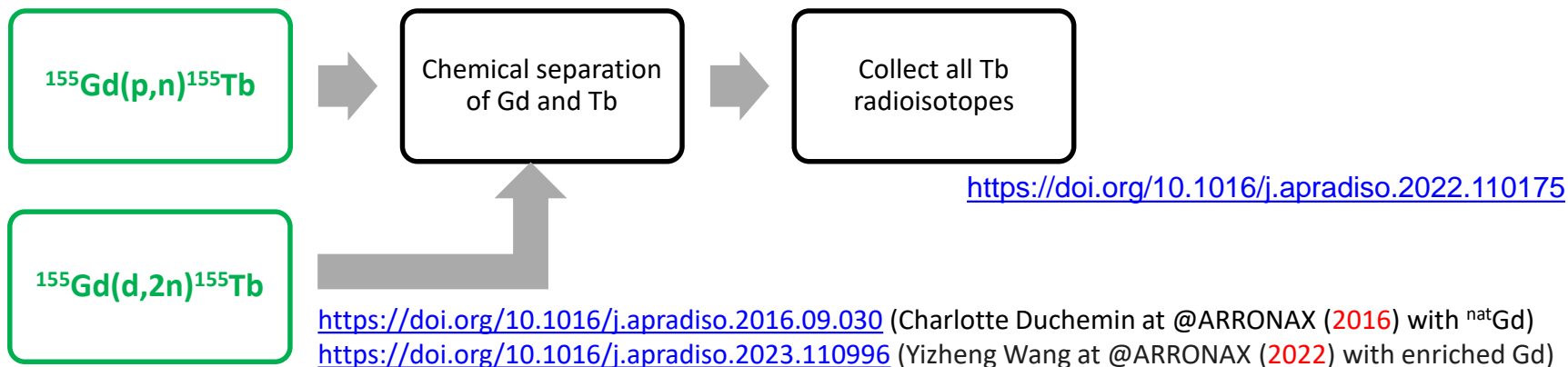
^b Subatech, UMR 6457, IMT Atlantique, CNRS/IN2P3

^c GIP ARRONAX

I am hard of hearing,
SO
in some slides, I will read the text to ensure I am
well understood, as my accent is very strong.

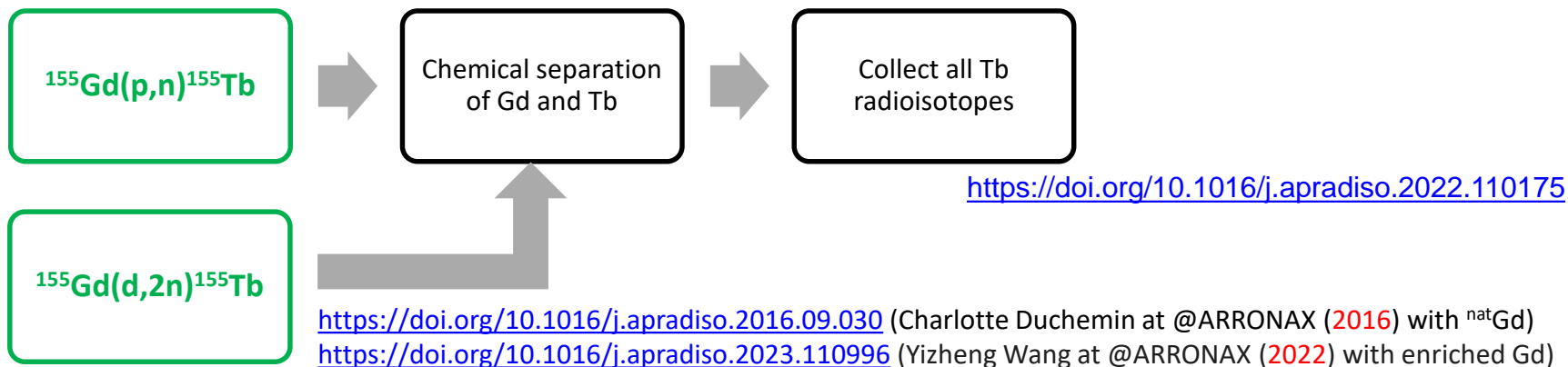
Different paths for ^{155}Tb production : proton or deuteron or alpha particles at @ARRONAX?

Direct route

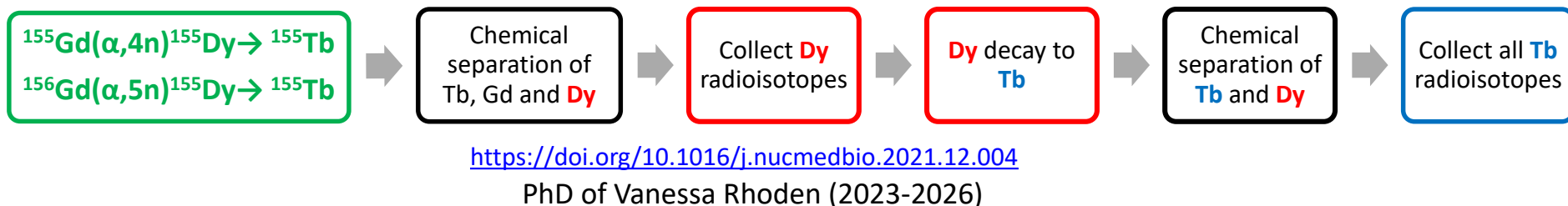


Different paths for ^{155}Tb production : proton or deuteron or alpha particles at @ARRONAX?

Direct route



Indirect route



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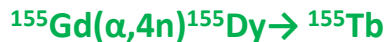


$E < 12$ MeV to avoid the ^{153}Tb ($T_{1/2} = 2.34$ d) production
At @GIP ARRONAX, the C70 cyclotron **cannot** deliver the beam at this energy



$E < 20$ MeV to avoid the ^{153}Tb ($T_{1/2} = 2.34$ d) production
At @GIP ARRONAX, the C70 cyclotron **can** deliver the beam at this energy

Indirect route



Work in ongoing with Vanessa Rhoden, PhD student 2023-2026

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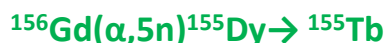


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$E < 20$ MeV to avoid the ^{153}Tb ($T_{1/2} = 2.34$ d) production
At @GIP ARRONAX, the C70 cyclotron **can** deliver the beam at this energy
enriched ^{155}Gd exists only in **powder** form → The targets must be manufactured

Indirect route



Work in ongoing with Vanessa Rhoden, PhD student 2023-2026

Targets manufacturing for Tb research

Thin targets for cross section data in nuclear research

Thickness	Conditions	Manufacture techniques	Nuclear data
$\leq 20 \mu\text{m}$	<ul style="list-style-type: none">▪ Homogeneity▪ Dense	<ul style="list-style-type: none">▪ Co-electrodeposition▪ Molecular plating	<ul style="list-style-type: none">▪ Cross section measurement▪ Yield and purity simulation

Thick targets for large-scale production of ^{155}Tb

Thickness	Conditions	Manufacture techniques	Production
$\geq 100 \mu\text{m}$	<ul style="list-style-type: none">▪ Homogeneity▪ Dense	<ul style="list-style-type: none">▪ Pelletizing technique	<ul style="list-style-type: none">▪ Thick target irradiation▪ Experimental yield and purity▪ Chemical separations with LN resin

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Poster of Vanessa Rhoden

Thick targets for large-scale production of ^{155}Tb

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Thin targets for cross section data in nuclear research

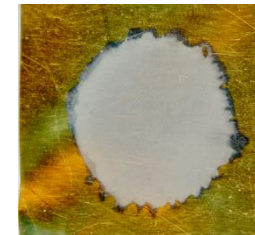
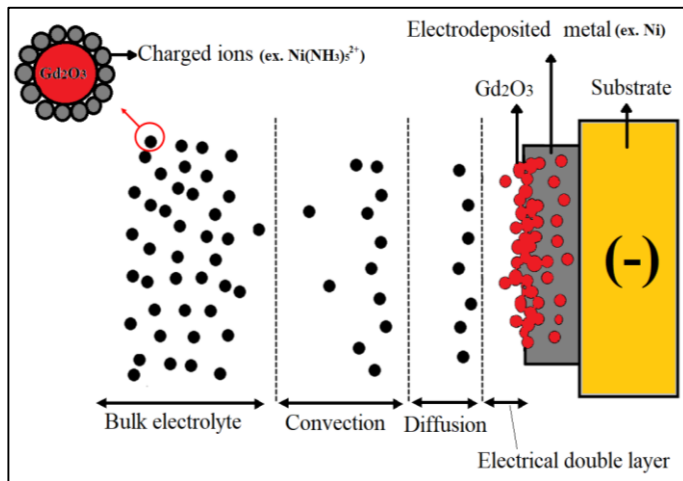
$$E^\circ(\text{Gd}^{3+}/\text{Gd}) = -2,279 \text{ V/SHE}$$

=> To obtain metallic Gd by reducing Gd^{3+} in aqueous solution is impossible due to very low standard hydrogen electrode of Gd^{3+}/Gd



Co-electrodeposition technique

Gd_2O_3 particles are trapped in Ni deposit by electrodeposition technique. This method has been used to prepare targets to measure cross sections of $^{155}\text{Gd}(d,2n)^{155}\text{Tb}$ nuclear reaction. To prevent the dissolution of Gd_2O_3 particles, the pH of the solution must be **greater than 7**. The solution used in our experiments is **ammoniacal** solution.



Thickness = 13 μm
(50- μm -thick gold substrat)
m(Gd): 2-3 mg (ICP-OES analysis)

Thin targets for cross section data in nuclear research

$$E^\circ(\text{Gd}^{3+}/\text{Gd}) = -2,279 \text{ V/SHE}$$

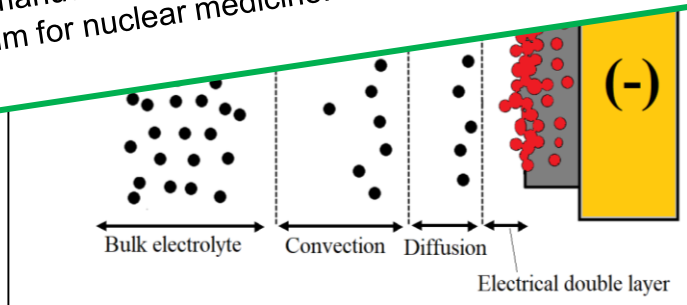
=> To obtain metallic Gd by reducing Gd^{3+} in aqueous solution is impossible due to very low standard potential of Gd^{3+}/Gd

Gd_2O_3 particles

This technique, developed with Yizheng Wang (PhD from 2019-2022), is used to conduct cross section measurements to investigate ^{155}Tb production from an enriched ^{155}Gd target with deuteron particles.

Articles:

1. Wang Y, Sounalet T, Guertin A, et al. Electrochemical co-deposition of Ni-Gd₂O₃ for composite thin targets preparation: Production of ^{155}Tb as a case study[J]. Applied Radiation and Isotopes, 2022, 186: 110287.
2. Wang Y, Sounalet T, Guertin A, et al. Study of terbium production from enriched Gd targets via the reaction $^{155}\text{Gd}(d, 2n)^{155}\text{Tb}$ [J]. Applied Radiation and Isotopes, 2023, 201: 110996.
3. PhD manuscript : Development of enriched gadolinium target for cross section measurement and future production of terbium for nuclear medicine. <https://www.theses.fr/2022NANU4062>.



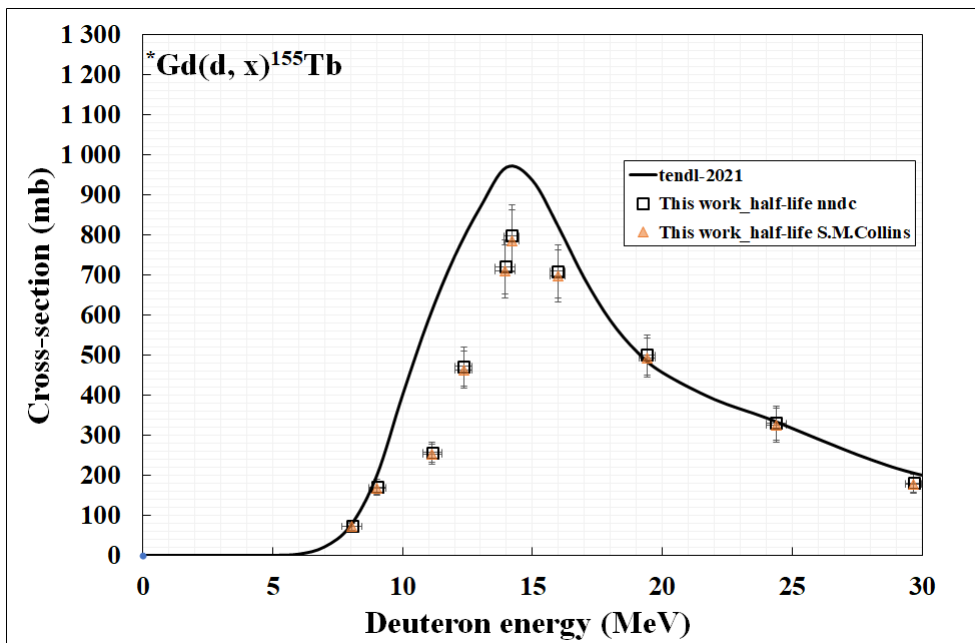
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 m(Gd): 2-3 mg (ICP-OES analysis)

Thin targets for cross section data in nuclear research

Co-electrodeposition

cross section measurements: irradiation on ^{155}Gd with deuteron particles (direct route)

Cross section measurements: ^{155}Tb



TENDL-2021 overestimates values at lower energies until 20 MeV and remains consistent at higher energies.

Isotope	152	154	155	156	157	158	160
Enrichment (%)	-	-	92.80	5.7	0.8	0.5	0.2

^{155}Gd enrichment = 92.80%
(noted as *Gd)

^{156}Gd purity = 5.7%

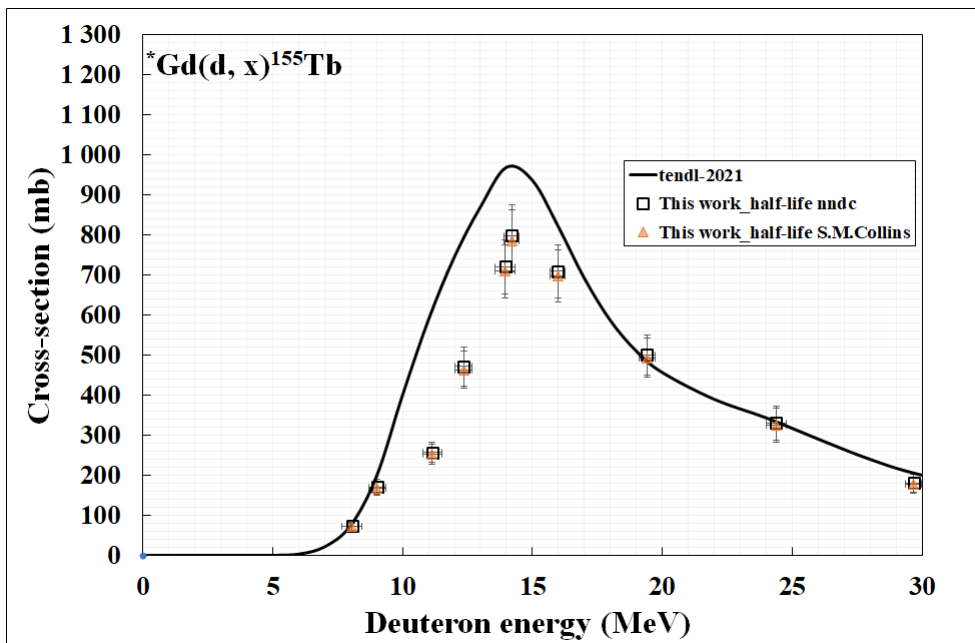
From Trace Sciences International

Thin targets for cross section data in nuclear research

Co-electrodeposition

cross section measurements: irradiation on ^{155}Gd with deuteron particles (direct route)

Cross section measurements: ^{155}Tb



TENDL-2021 overestimates values at lower energies until 20 MeV and remains consistent at higher energies.

➤ **Before 13 MeV**, the ^{155}Tb production was produced from $^{155}\text{Gd}(d,2n)^{155}\text{Tb}$.

Isotopes	^{155}Gd	^{156}Gd	^{157}Gd	^{158}Gd
$E_{\text{threshold}}$ (MeV)	3.88	12.52	18.96	27.00

➤ **From 13 MeV to 19 MeV**, ^{155}Gd and ^{156}Gd contributed to produce ^{155}Tb (maximum cross section = 800 mb at 14.5 MeV)

Isotope	152	154	155	156	157	158	160
Enrichment (%)	-	-	92.80	5.7	0.8	0.5	0.2

^{155}Gd enrichment = 92.80%
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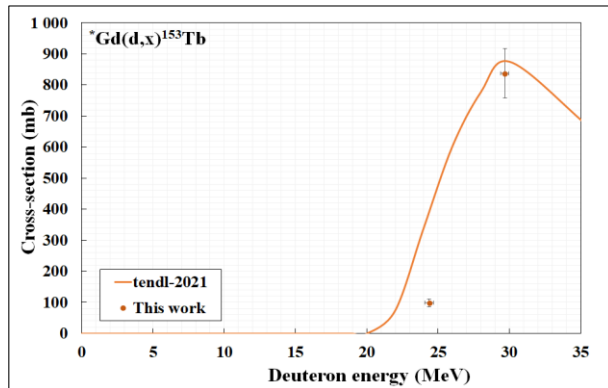
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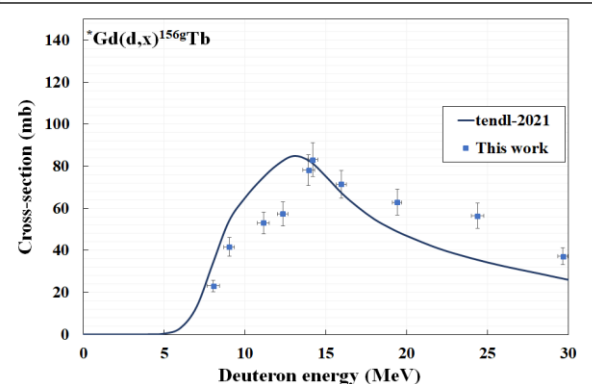
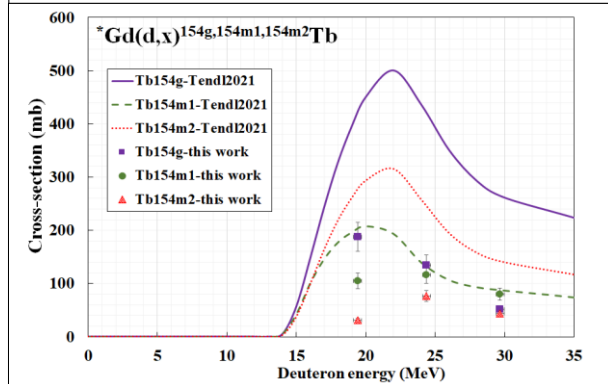
Thin targets for cross section data in nuclear research

Co-electrodeposition

cross section measurements: irradiation on ^{155}Gd with deuteron particles (direct route)



Isotope	Half-life	$E_{\text{threshold}}$ (MeV)
^{153}Tb	2.34 d	20.17
$^{154\text{g,m1,m2}}\text{Tb}$	21.5 h, 9.4 h, 22.7 h	13.16
^{156}Tb	5.35 d	0



- ❑ ^{153}Tb and ^{154}Tb can be avoided at lower energy, < 20 MeV.
- ❑ $^{156\text{g}}\text{Tb}$ is inevitable.
- ❑ The half-life of $^{156\text{g}}\text{Tb}$ is similar to that of ^{155}Tb (5.32 d).

Important to study the ^{156}Tb cross section as well as the impact of ^{156}Tb impurity on images !

Targets manufacturing for Tb research

Thin targets for new data nuclear research

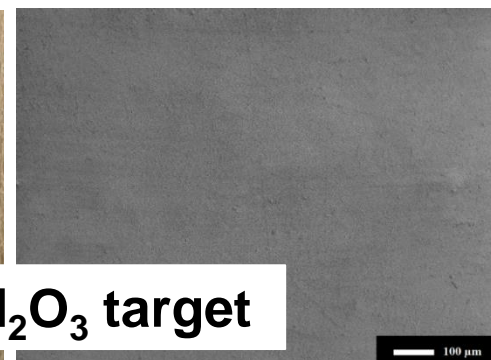
Thickness	Conditions	Manufacture techniques	Nuclear data
$\leq 20 \mu\text{m}$	<ul style="list-style-type: none">▪ Homogeneity▪ Dense	<ul style="list-style-type: none">▪ Co-electrodeposition▪ Molecular plating	<ul style="list-style-type: none">▪ Cross-section measurement▪ Yield and purity simulation

Thick targets for large-scale production of ^{155}Tb

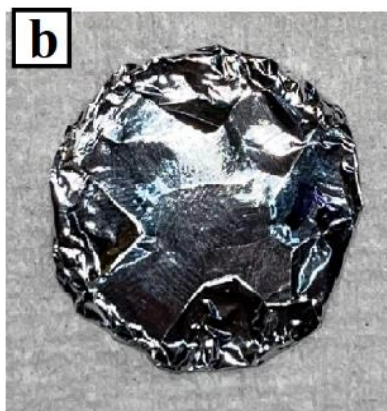
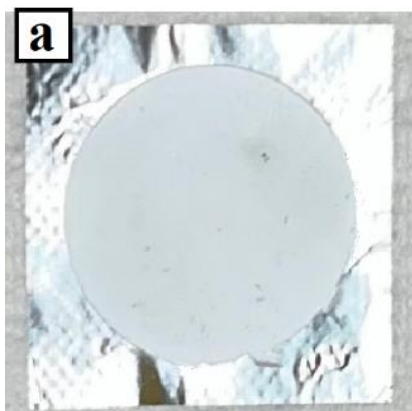
Thickness	Conditions	Manufacture techniques	Production
$\geq 100 \mu\text{m}$	<ul style="list-style-type: none">▪ Homogeneity▪ Dense	<ul style="list-style-type: none">▪ Pelletizing technique	<ul style="list-style-type: none">▪ Thick target irradiation▪ Experimental yield and purity▪ Chemical separations with LN resin

Thick targets for large-scale production of ^{155}Tb

pelletizing technique

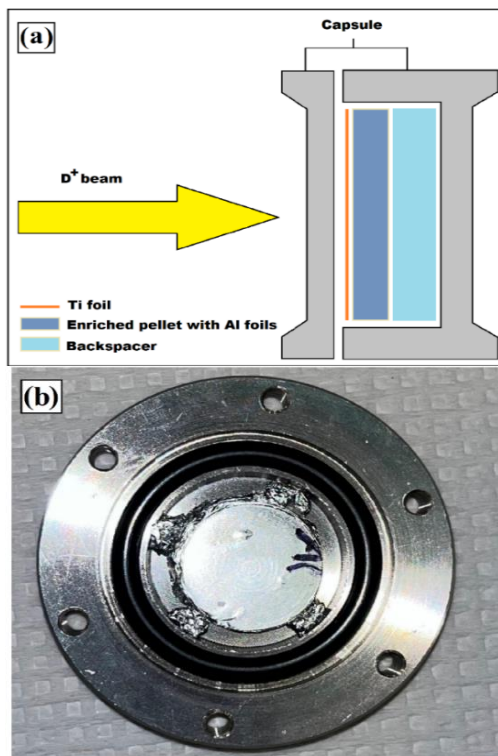


Gd_2O_3 target

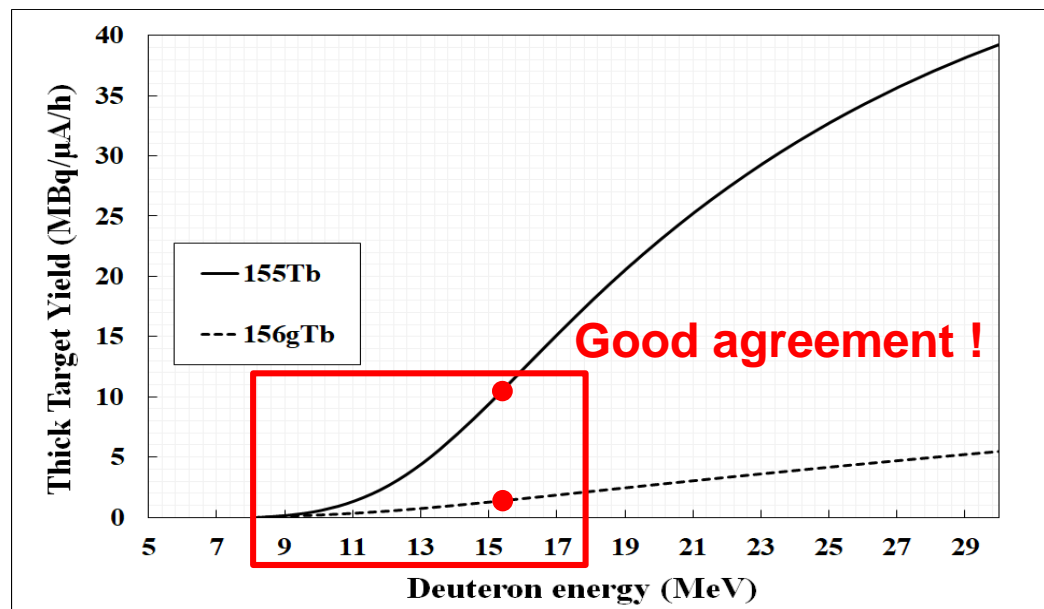


Mass = 0.6 g, diameter = 2 cm, thickness = 390 μm

Thick targets for large-scale production of ^{155}Tb pelletizing technique



$E_{\text{incident}} = 15.1 \text{ MeV}$, $E_{\text{out}} = 8.6 \text{ MeV}$ (maximum production of ^{155}Tb)
500 nA for 1 h



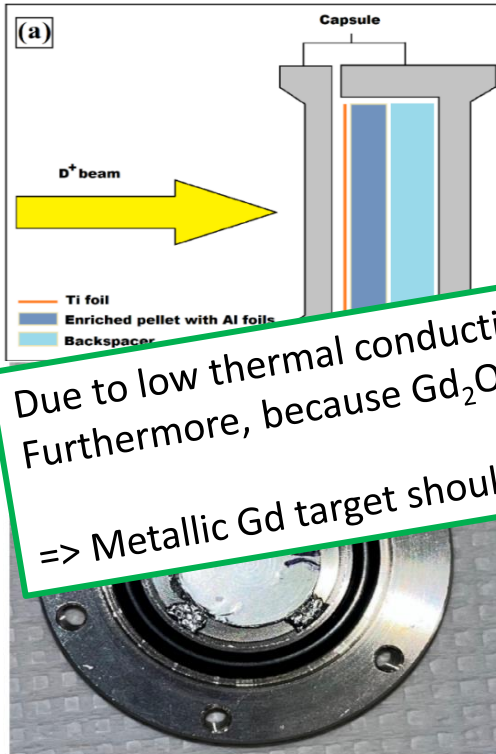
Experimental yield:

- ^{155}Tb : $10.2 \pm 0.7 \text{ MBq}/\mu\text{A}/\text{h}$
- ^{156}Tb : $1.3 \pm 0.1 \text{ MBq}/\mu\text{A}/\text{h}$

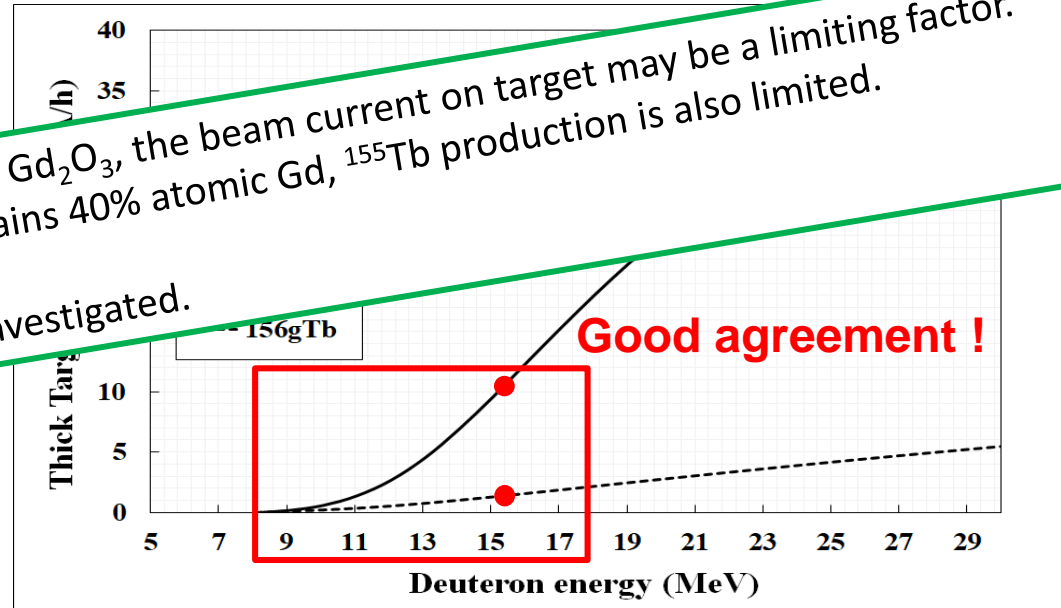
Purity of ^{155}Tb :

- Experiment: 88% (9 d after EOB)
- Estimation: 89% (9 d after EOB)

Thick targets for large-scale production of ^{155}Tb pelletizing technique



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Due to low thermal conductivity of Gd_2O_3 , the beam current on target may be a limiting factor. Furthermore, because Gd_2O_3 contains 40% atomic Gd, ^{155}Tb production is also limited.
=> Metallic Gd target should be investigated.

Experimental yield:

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Purity of ^{155}Tb :

Experiment: 88% (9 d after EOB)
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Conclusion

- ✓ We are able to manufacture thin targets for nuclear research : co-electrodeposition and molecular plating (Poster of Vanessa Rhoden)
- ✓ We are able to manufacture thick targets for production : a pellet of Gd_2O_3 wrapped with Al.
Perspective : obtain a metallic Gd -> molten salts technique?
- ✓ Cross section measurements have been studied with deuteron as particles
Perspective : measure cross section with alpha particles (Vanessa Rhoden's PhD thesis).
- ✓ Knowing the values of cross section measurements allows us to determine which deuteron energy is optimal to produce ^{155}Tb with *good* radioisotopic purity (90% in this work) : less than 20 MeV to avoid producing ^{153}Tb .
- Proton or deuteron irradiation? It depends on the acceptable value of ^{156}Tb .
- The radioisotopic purity of ^{155}Tb depends on the purity of ^{155}Gd .
- Alpha irradiation is interesting; however, only few cyclotrons can deliver this particle.

Publications

For 10 years, we are studying on production of Tb radionuclides (Tb-149, Tb-155). Below are the peer-reviewed articles that we have published :

- Deuteron induced Tb-155 production, a theranostic isotope for SPECT imaging and auger therapy, Applied Radiation and Isotopes 118 (2016) 281–289, Charlotte Duchemin.
- Terbium Radionuclides for Theranostics Applications: A Focus On MEDICIS-PROMED, Physics Procedia 90 (2017) 157 – 163, Roberto Formento-Cavaier.
- New excitation functions for proton induced reactions on natural gadolinium up to 70 MeV with focus on ^{149}Tb production, Nuclear Inst. and Methods in Physics Research B 478 (2020) 174–181, Roberto Formento-Cavaier.
- Electrochemical co-deposition of Ni–Gd₂O₃ for composite thin targets preparation: Production of ^{155}Tb as a case study, Applied Radiation and Isotopes 186 (2022) 110287, Yizheng Wang.
- Study of terbium production from enriched Gd targets via the reaction $^{155}\text{Gd}(d,2n)^{155}\text{Tb}$, Applied Radiation and Isotopes 201 (2023) 110996, Yizheng Wang.

Thank you for your attention

The ARRONAX cyclotron is a project promoted by the Regional Council of Pays de la Loire financed by local authorities, the French government and the European Union.

This work has been supported in part by a grant from the French National Agency for Research called “Investissements d’Avenir”, Equipex ArronaxPlus n°ANR-11-EQPX-0004 and labex IRON n°ANR-11-LABX-18-01.