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 Nigron[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

$^{155}Gd(p,n)^{155}Tb$

Chemical separation of Gd and Tb

Collect all Tb radioisotopes

https://doi.org/10.1016/j.apradiso.2022.110175

$^{155}Gd(d,2n)^{155}Tb$

https://doi.org/10.1016/j.apradiso.2016.09.030 (Charlotte Duchemin at @ARRONAX (2016) with ^{nat}Gd)
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Indirect route

$^{155}Gd(\alpha,4n)^{155}Dy \rightarrow {}^{155}Tb$
$^{156}Gd(\alpha,5n)^{155}Dy \rightarrow {}^{155}Tb$

→ Chemical separation of Tb, Gd and Dy → Collect Dy radioisotopes → Dy decay to Tb → Chemical separation of Tb and Dy → Collect all Tb radioisotopes

https://doi.org/10.1016/j.nucmedbio.2021.12.004
PhD of Vanessa Rhoden (2023-2026)

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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

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E < 20 MeV to ...
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enriched ^{155}Gd exits only in **powder** form → The targets must be manufactured

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Targets manufacturing for Tb research

Thin targets for cross section data in nuclear research

Thickness	Conditions	Manufacture techniques	Nuclear data
≤ 20 µm	▪ Homogeneity ▪ Dense	▪ Co-electrodeposition ▪ Molecular plating	▪ Cross section measurement ▪ Yield and purity simulation

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

Thickness	Conditions	Manufacture techniques	Production
≥ 100 µm	▪ Homogeneity ▪ Dense	▪ Pelletizing technique	▪ Thick target irradiation ▪ Experimental yield and purity ▪ Chemical separations with LN resin

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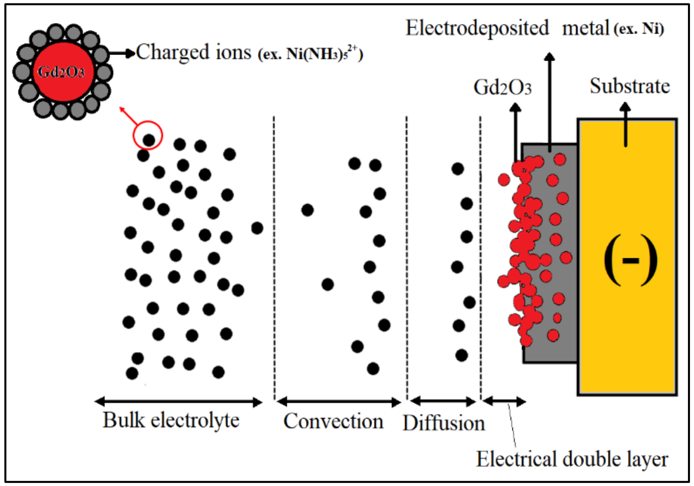
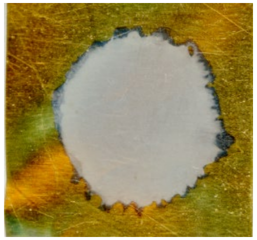
Thin targets for cross section data in nuclear research

E°(Gd^{3+}/Gd) = -2,279 V/SHE
=> To obtain metallic Gd by reducing Gd^{3+} in aqueous solution is impossible due to very low standart 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}Gd(d,2n)^{155}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)

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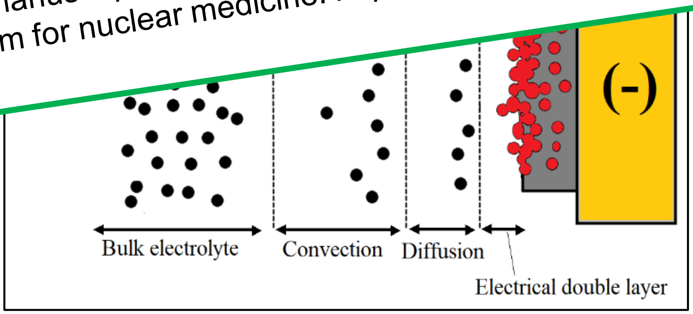
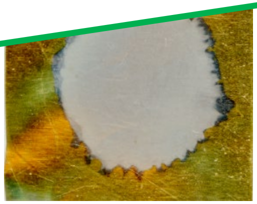
Gd_2O_3 particl…

This technique, developped with Yizheng Wang (PhD from 2019-2022), is used to conduct cross section measurements to investigate ^{155}Tb production from a 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 155Tb 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 155Gd (d, 2n) 155Tb[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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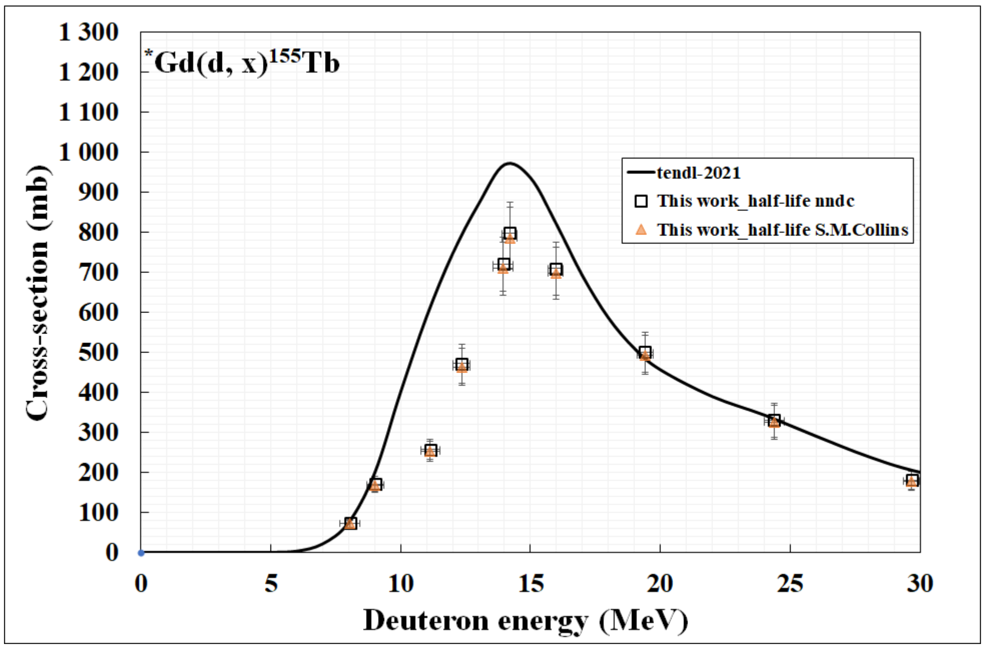
Thin targets for cross section data in nuclear research

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

From Trace Sciences International

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

^{156}Gd purity = 5.7%

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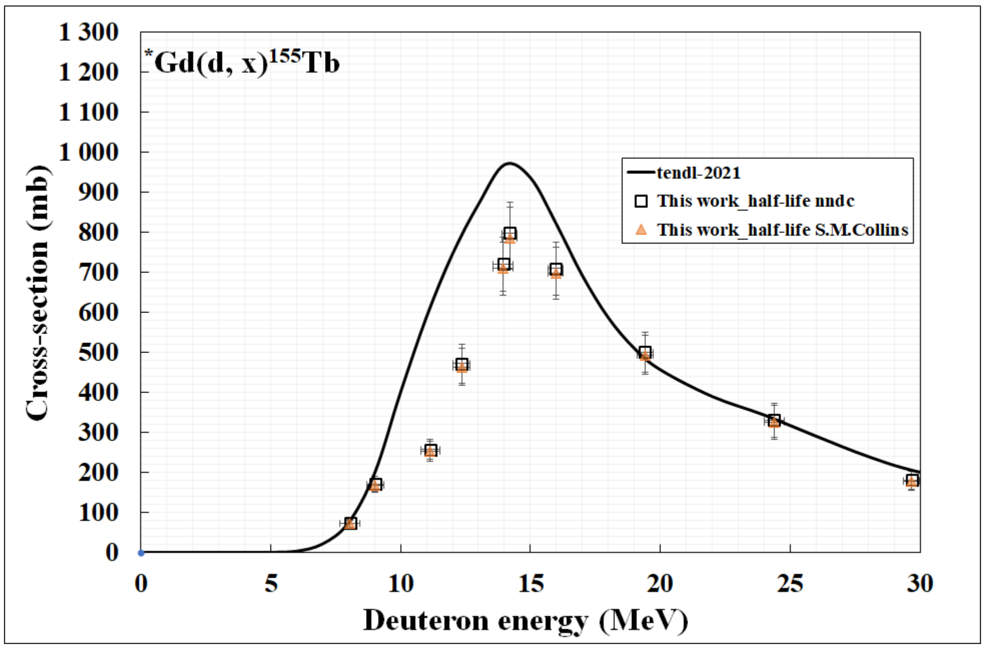

TENDL-2021 overestimates values at lower energies until 20 MeV and remains consistent at higher energies.

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

Isotopes	^{155}Gd	^{156}Gd	^{157}Gd	^{158}Gd
$E_{threshold}$ (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

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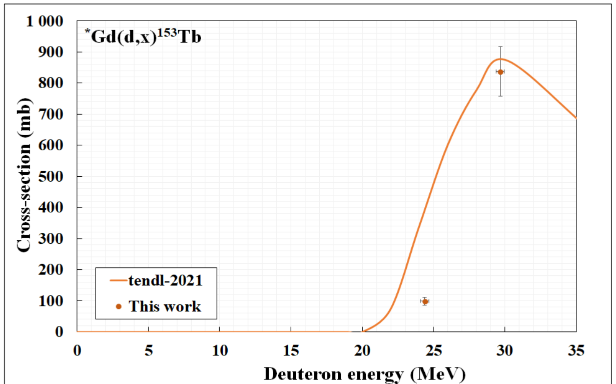
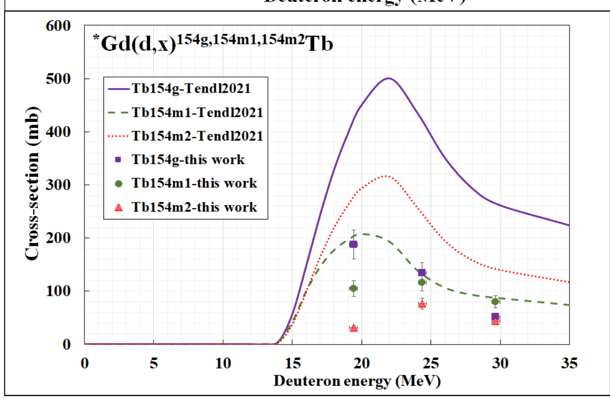
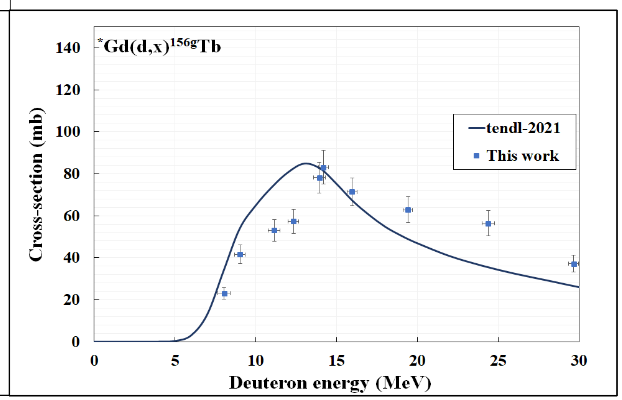


Isotope	Half-life	$E_{threshold}$ (MeV)
^{153}Tb	2.34 d	20.17
^{154g,m1,m2}Tb	21.5 h, 9.4 h, 22.7 h	13.16
^{156}Tb	5.35 d	0





- ❑ ^{153}Tb and ^{154}Tb can be avoided at lower energy, < 20 MeV.
- ❑ ^{156g}Tb is inevitable.
- ❑ The half-life of ^{156g}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 !

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Thin targets for new data nuclear research

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

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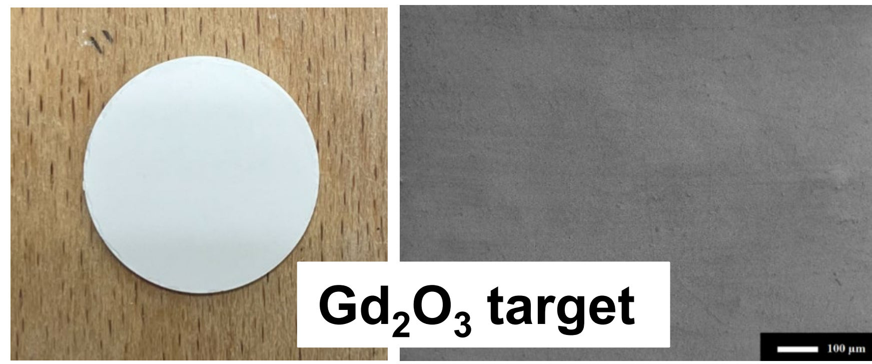
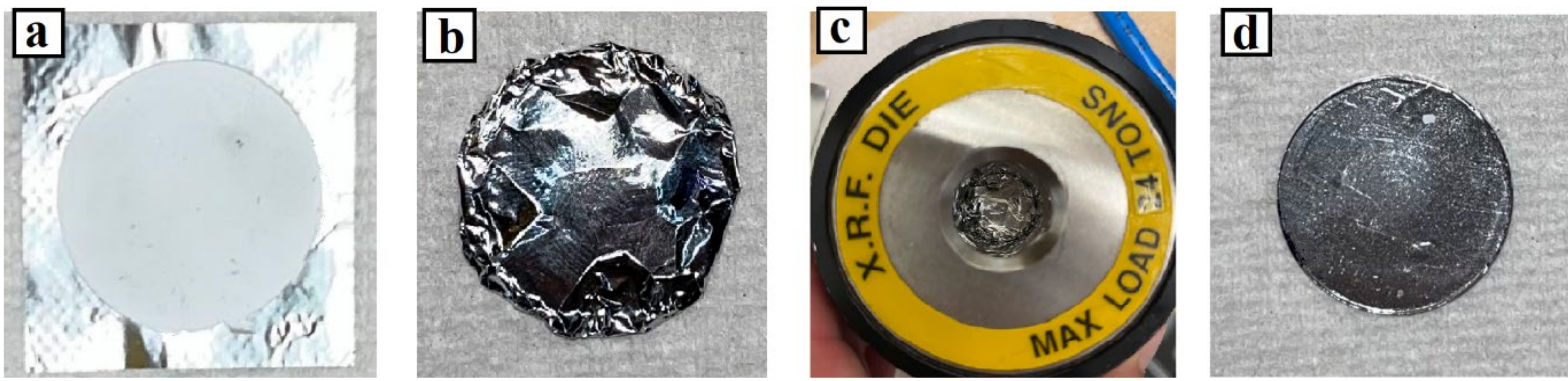
≥ 100 µm	▪ Homogeneity ▪ Dense	▪ Pelletizing technique	▪ Thick target irradiation ▪ Experimental yield and purity ▪ Chemical separations with LN resin

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

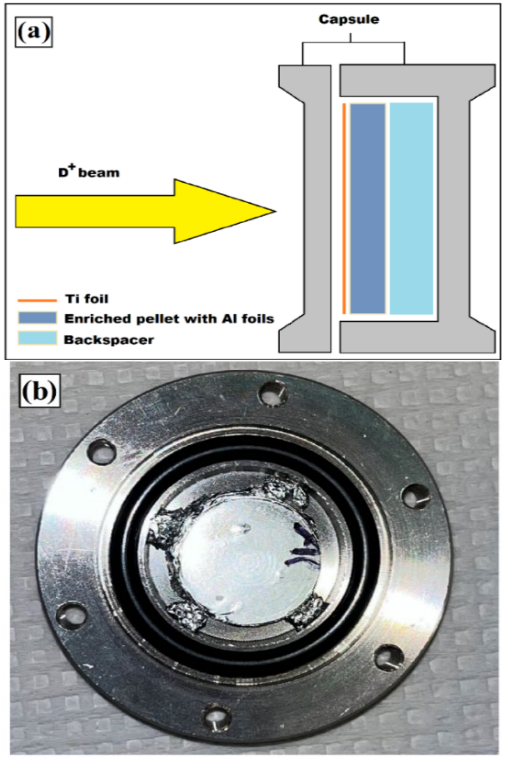
pelletizing technique





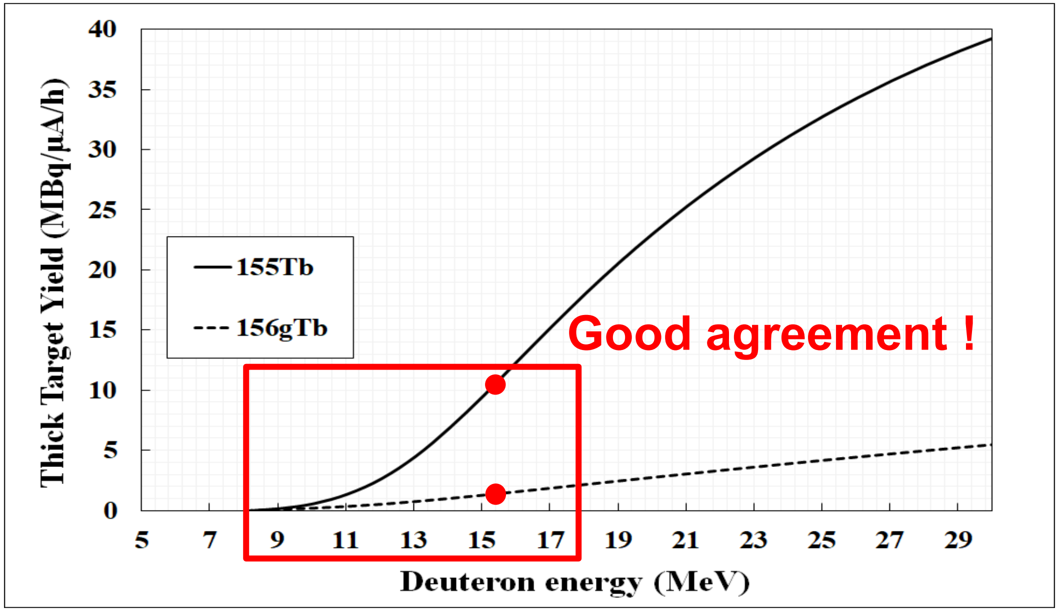
Mass = 0.6 g, diameter = 2 cm, thickness = **390 µm**

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



$E_{incident}$ = 15.1 MeV, E_{out} = 8.6 MeV (maximum production of ^{155}Tb)

500 nA for 1 h



Experimental yield:
- **^{155}Tb: 10.2 ± 0.7 MBq/µA/h**
- **^{156}Tb: 1.3 ± 0.1 MBq/µA/h**

Purity of ^{155}Tb:
Experiment: 88% (9 d after EOB)
Estimation: 89% (9 d after EOB)

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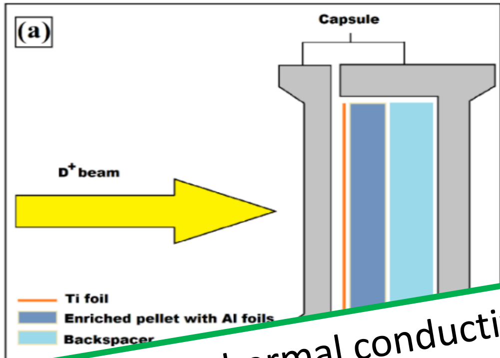

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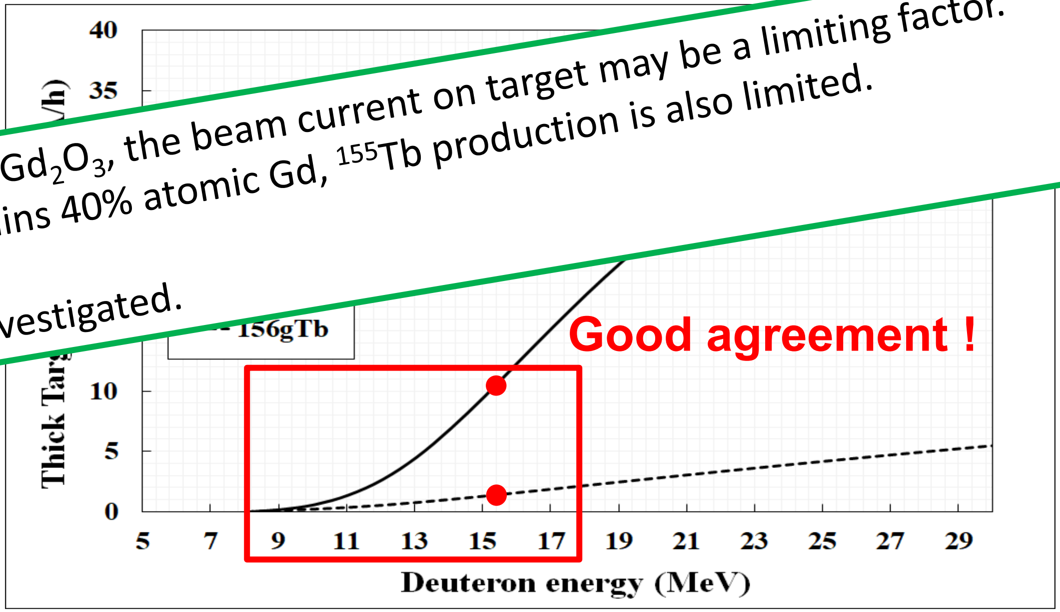

Due to low thermal conductivity of Gd_2O_3, the beam current on target may be a limiting factor.
Furthermore, because Gd_2O_3 contains 40% atomic Gd, ^{155}Tb production is also limited.

=> Metallic Gd target should be investigated.

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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 1O 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 149Tb production, Nuclear Inst. and Methods in Physics Research B 478 (2020) 174–181, Roberto Formento-Cavaier.

- Electrochemical co-deposition of Ni–Gd_2O_3 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}Gd(d,2n)^{155}Tb, Applied Radiation and Isotopes 201 (2023) 110996, Yizheng Wang.

Thank you for your attention

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