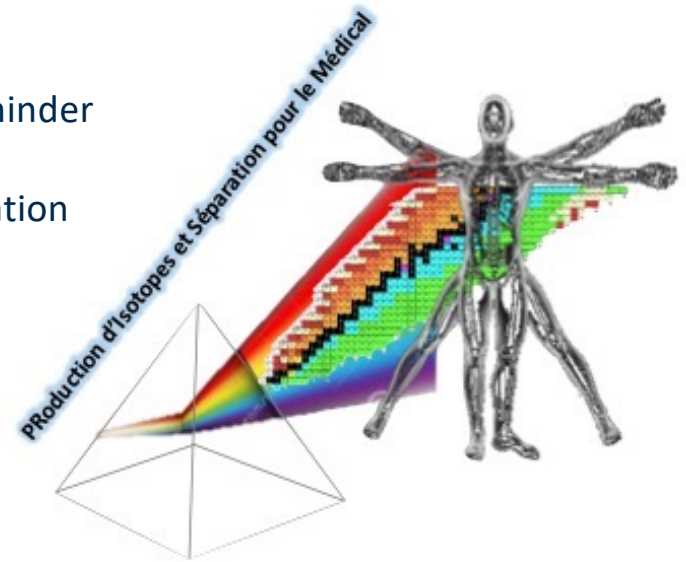
^{155}Tb production: a proof-of-concept method for an alternative production of medical isotope

TTRIP (Tools for Tb RadioIsotope Production for nuclear medicine)
ANR-21-CE19-0037

- Internal vectorized radiotherapy (RIV): a brief reminder
- Project on radio-isotope production and vectorization
  - → radio-isotope production
  - → use of electromagnetic separation to improve the isotopic purification
  - → first results
- Outlook and conclusions

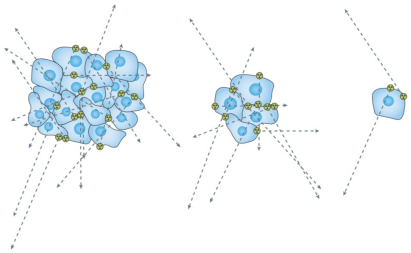


Radiotherapy

Use of radiations to
- → deliver energy into tumors
  - ↳ damage the DNA cancer cells
- → destroy their ability to divide and grow

→ **precise targeting of the tumor**
→ **limit damages to surrounding healthy tissues**

Internal Vectorized Radiotherapy
→ **introduction** of a radioactive therapeutic agent into the body **in direct contact with the tumour**



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isotope of medical interest

high purity

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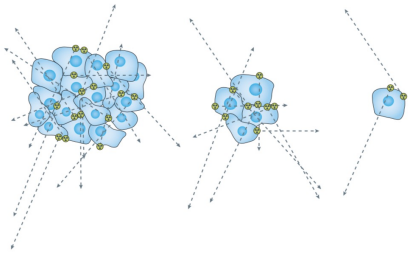

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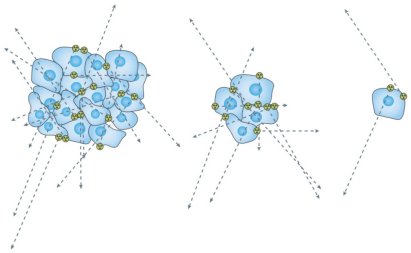

isotope of medical interest

biogical molecule able to target cancer cells

Peptide, antibody, …

high purity

precise targeting
spare healthy tissues

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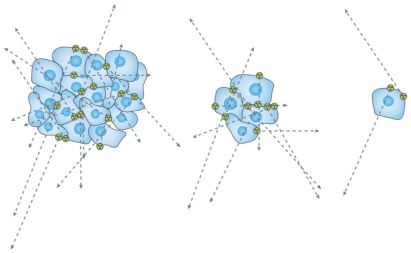

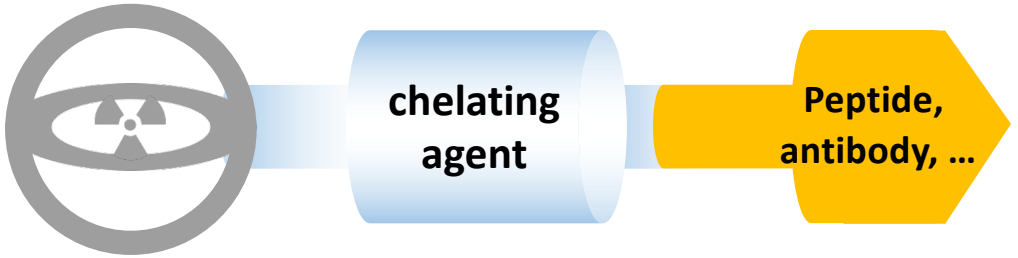

high purity

prevent release of the radioisotope
bifunctional compatibilty

precise targeting
spare healthy tissues

Internal Vectorized Radiotherapy

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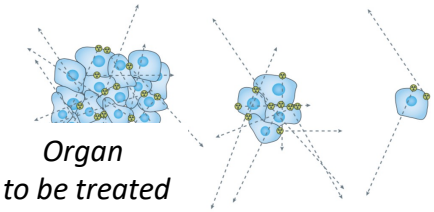
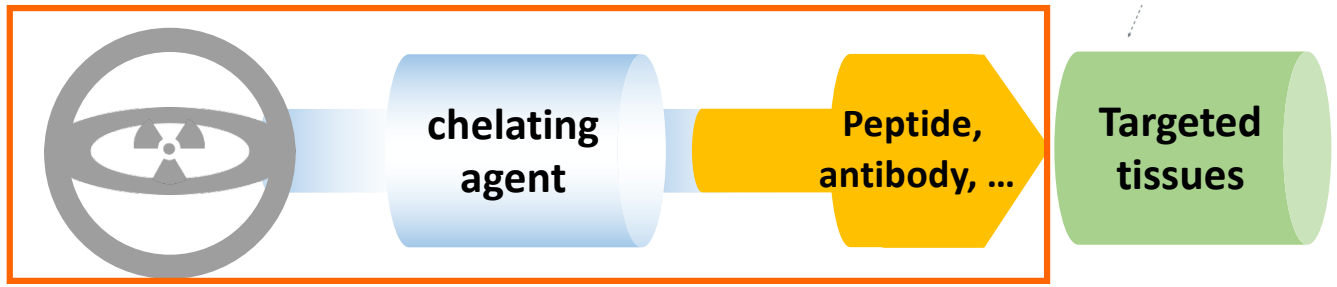


Organ to be treated

radiopharmaceutical



high purity

prevent release of the radioisotope
bifunctional compatibilty

precise targeting
spare healthy tissues

need for a large choice of {radionucleide,vector}
→ « toolbox » to propose the most appropriate treatment

Internal Vectorized Radiotherapy « strategy »

THERAPY

→ **Optimize tumour treatment**

radioisotope adapted to tumour size and geometry

decaying mode
energy of emitted radiation
adapted linear energy transfer (LET)
radiation path

→ **Adapt to the bio-distribution time of the vector**

choice of radioactive period ($T_{1/2}$)

β⁻ emitter	***α emitter***	***conversion e⁻***	***Auger e⁻***
~ 500 – 2500 keV	~ 4000 – 9000 keV	~ 10 – 200 keV	~ 0,5 – 5 keV
~ 0,2 keV/µm	~ 50-200 keV/µm	~ 0,5 keV/µm	~ 1 - 23 keV/µm
5 – 150 cells µm à qq cm	1 – 3 cells 40-100 µm	~ 10 cells 0,5 mm	< 1 cell 1 nm - 1 µm
^{90}Y, ^{177}Lu, ^{153}Sm, ^{131}I	^{211}At, ^{212}Bi, ^{213}Bi, ^{225}Ac, ^{212}Pb, ^{223}Ra, ^{149}Tb	^{111}In, ^{67}Ga, ^{195m}Pt, ^{123}I, ^{125}I	

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choice of radioactive period ($T_{1/2}$)

→ **deliver the right dose at the right place**
→ **avoid unnecessary doses**

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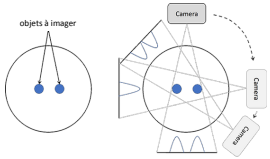
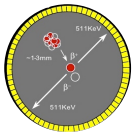
IMAGING : ***Personalize treatment***
→ better target the tumour
→ better estimate of the dose to be injected



SPECT
Single Photon Emission Computed Tomography

low energy γ emitter
^{99m}Tc, ^{123}I



PET
Positron Emission Tomography

positron emitter (β⁺)
^{15}O, ^{13}N, ^{11}C, ^{18}F

Theranostic strategy

THERApy - diag**NOSTIC**

Terbium ‘s family:
swiss knife of nuclear medicine

- ^{149}Tb ($T_{1/2}$ = 4.12 h, α therapy - 3.97 MeV)
- ^{152}Tb ($T_{1/2}$ = 17.5 h, PET 1140 keV)
- ^{155}Tb ($T_{1/2}$ = 5.32 d, SPECT and Auger therapy)
- ^{161}Tb ($T_{1/2}$ = 6.9 d, β^- therapy 154 keV and Auger therapy)

→ same chemical properties
↳ Identical bio-kinetic and pharmaco-kinetic

Quantitative data from imaging
=
Precise absorbed dosis estimation
into
→ targeted **lesion** (therapeutic effects)
→ non targeted **tissues** (secondary effects)

toward
more personalised treatment

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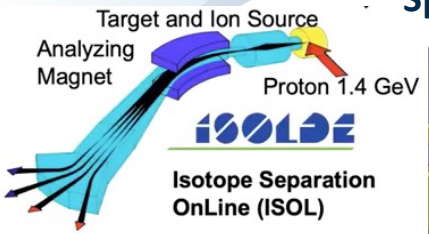
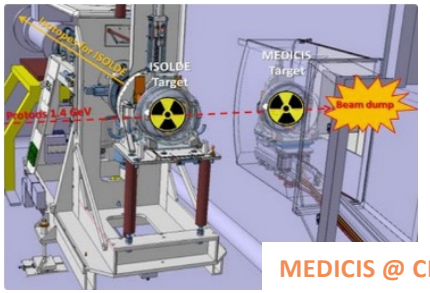
Production route for ^{155}Tb

Spallation reaction



« ISOLDE technique »



MEDICIS @ CERN

p+Ta → **^{155}Tb**
+ off line mass separation and radiochemical purification

high grade purity (> 99.9%)

not suitable for medical requirements
→ not sustainable production
(too important and costly infrastructure needed)
→ too limited amount of activity for clinical studies

Production route for ^{155}Tb

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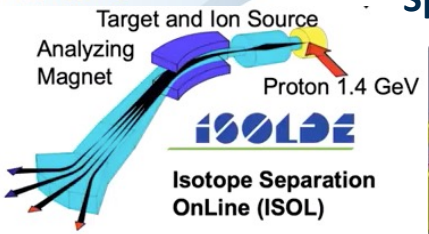

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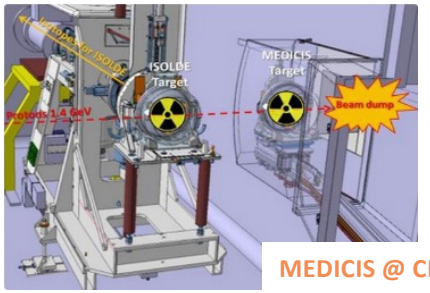

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Medical cyclotron production

^{159}Tb(p,5n)^{155}Dy(ε)^{155}Tb
^{155}Gd(p,n)^{155}Tb
^{155}Gd(d,2n)^{155}Tb
^{152}Sm(^{7}Li,4n)^{155}Tb
^{nat}Dy(d,x)^{155}Tb(cum)

cyclotron based studied are mostly with ^{nat}Gd or commercial $^{enr.\ 155}$Gd

	^{152}Gd	^{154}Gd	**^{155}Gd**	^{156}Gd	^{157}Gd	^{158}Gd	^{160}Gd
^{nat}Gd	0,2 %	2,18 %	**14,8 %**	20,47 %	15,65 %	24,84 %	21,86 %
commercial $^{enr.\ 155}$Gd			**92,8 %**	5,7 %	0,8 %	0,5 %	0,2 %

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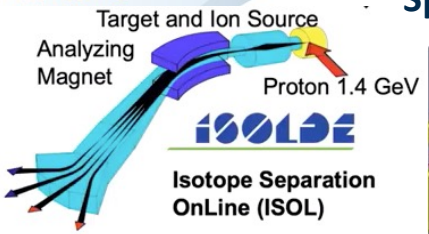

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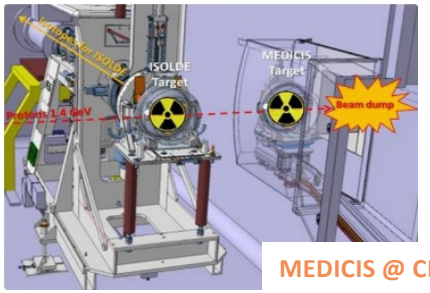

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→ numerous opened reaction channel
↳ high level of contaminant production

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need for very enriched targets

limiting reaction channel number
↳ less contaminants after the production reaction
↳ should make easier purification step
↳ should make possible to achieve the necessary purity

*"Clearly, radionuclidically pure productions will require the use of the (p,n) reaction only and **highly enriched targets** with **careful selection of the production energy window**."*
C. Vermeulen et al., Nucl. Inst. Meth. B 2012, 275, 24-32

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PRODUCTION OPTIMIZATION OF A MOLECULE BIO-LABELED WITH ^{155}TB

Usual production methods are not always adapted to a ***large scale regular production*** *of* ***pure radioisotopes***

^{155}Tb : optimization of the production → M.Bouteculet PhD thesis (IJCLab)

- *Pure ^{155}Gd production (SIDONIE separator)*
- *Excitation function measurement of ^{155}Gd(p,n)^{155}Tb*
- *Recovery of other Gd isotopes*
  *(**^{152}Gd** for $^{149,\ 152}$Tb production, **^{154}Gd** for ^{152}Tb production, **^{160}Gd** for ^{161}Tb production)*
- *Quantify the effect of contaminant onto the image quality performed with ^{155}Tb*

TTRIP (Tools for Tb RadioIsotope Production for nuclear medicine)

ANR-21-CE19-0037

IJCLab Irène Joliot-Curie Laboratoire de Physique des 2 Infinis — Subatech — GANIL — IC Institut de Chimie de Strasbourg — icmub Institut de Chimie Moléculaire — ARRONAX — NEUTRONS FOR SOCIETY

cnrs — université PARIS-SACLAY — FACULTÉ DES SCIENCES D'ORSAY — Université Paris Cité

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Usual production methods are not always adapted to a ***large scale regular production*** *of* ***pure radioisotopes***

Bioconjugate radiolabelling *(like DOTA) with* ***metals*** *needs high temperature heating (> 80°C) to* ***accelerate complexation*** *⇒* ***protein vectorisation « excluded »*** *(denaturation)*

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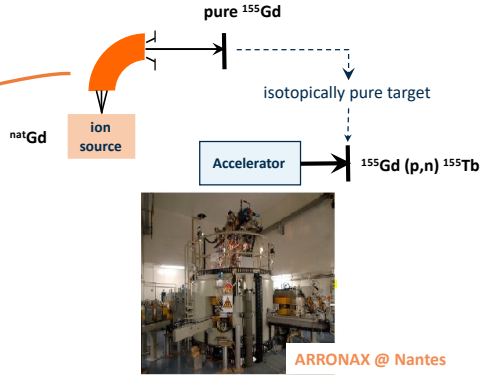
Chemistry to explore Tb chelation

- *Conception and synthesis of new model of Tb^{3+} chelators.*
- *Structural, thermodynamical and kinetic studies of chelators and their metallic complexes.*
- *Cytotoxicity of Tb*
- *Bioconjugation of functionalised chelators*

→ S.Lam PhD thesis (IJCLab + IRSN)

Radionuclide production : SIDONIE separator

SIDONIE separator @ IJCLab



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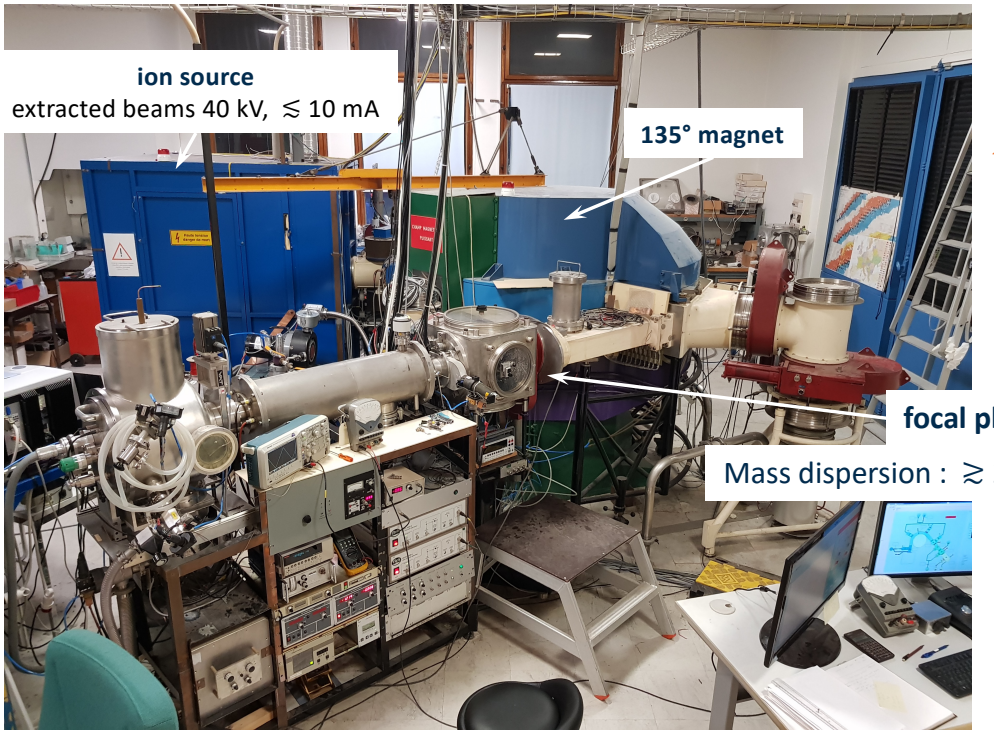
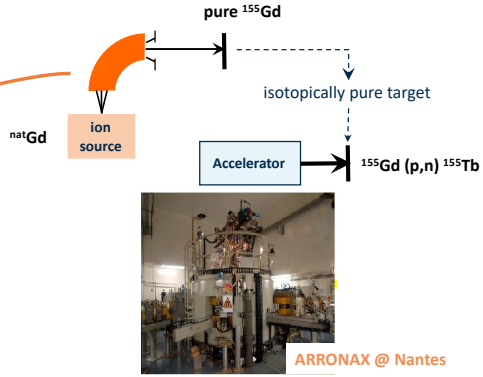
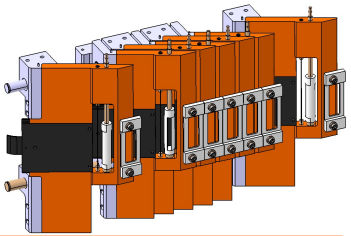


Mass dispersion : ≳ 2000 mm x ΔM/M

SIDONIE separator @ IJCLab



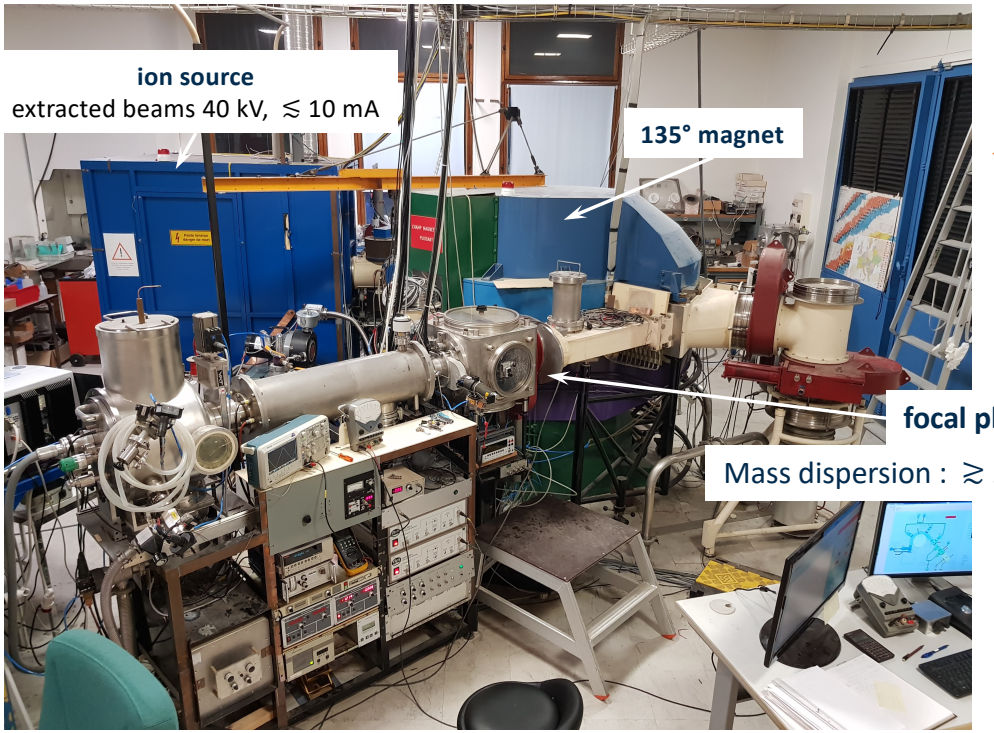


collection system to recover isotopes
(ex: ^{152}**Gd**(p,n)^{152}Tb, ^{152}**Gd**(p,4n)^{149}Tb , ^{154}**Gd**(p,6n)^{149}Tb, ^{160}**Gd**(n,γ)^{161}Gd (β^-) ^{161}Tb)

First test for 1 box: decembre 2023

ANR-21-CE19-0037

Radionuclide production : SIDONIE separator



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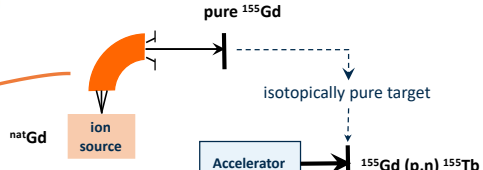

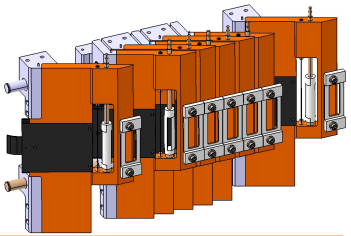

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ANR-21-CE19-0037

need to recover isotope to make target

1st production method

Development of targets at GANIL

Challenge: developing a deposition technique of with **isotopic Gd powder**
low losses of isotopic Gd → **high efficiency technique**

by courtesy of
G.Frémont
C.Stodel

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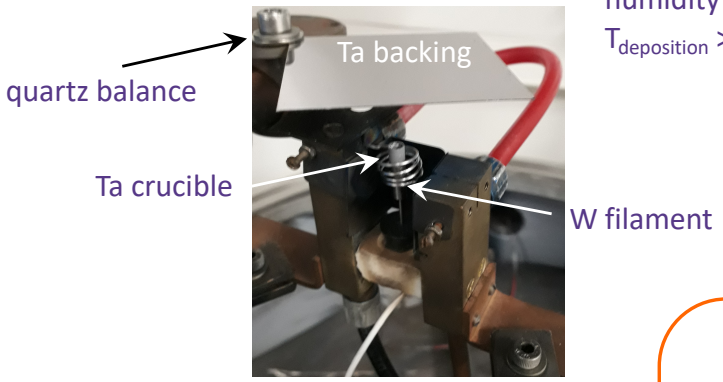
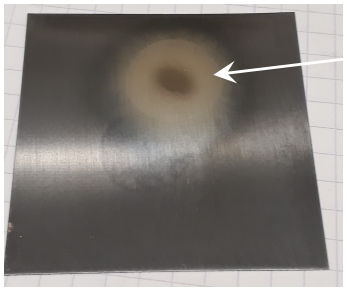
First tests : **electronic bombardment of Gd_2O_3 + Zr**

humidity removing: 1h at 100°C
$T_{deposition}$ > 1650 °C

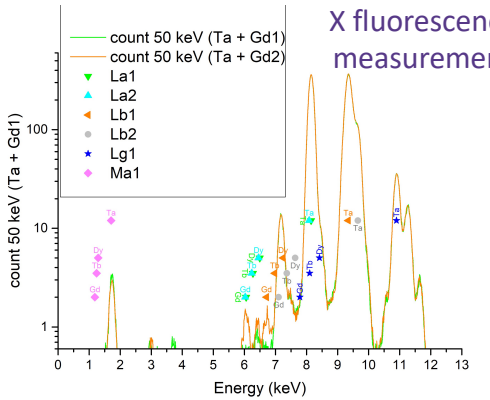


First result …

65 µg/cm² deposit



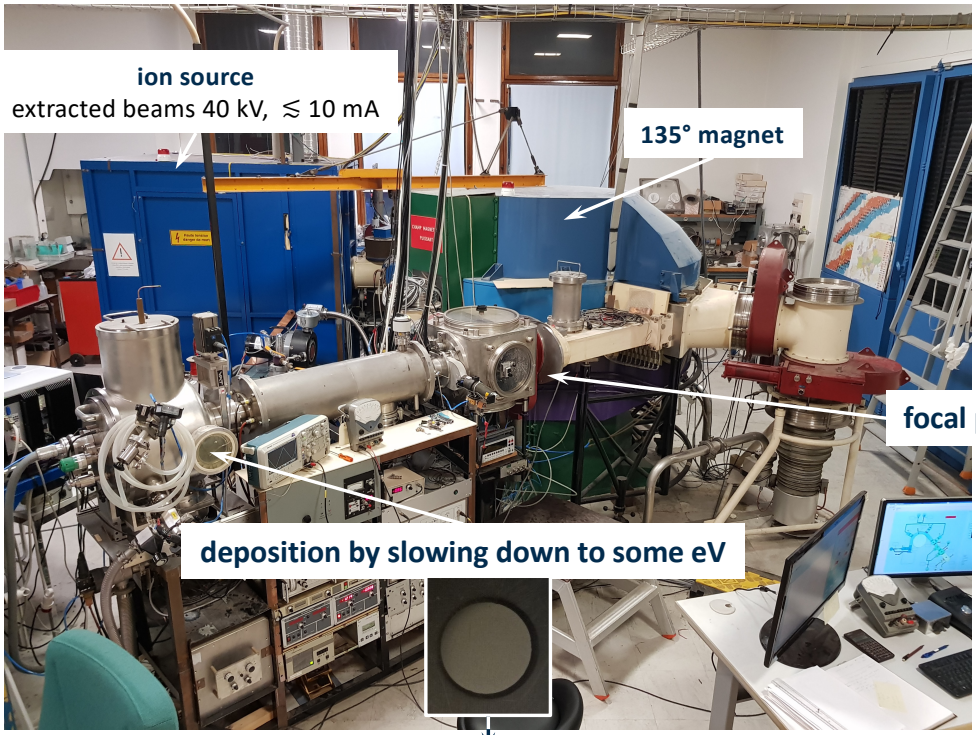
X fluorescence measurement



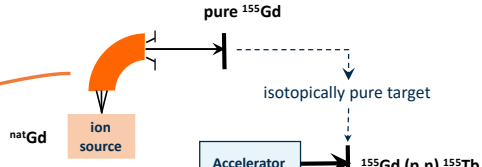
STILL OPENED QUESTIONS:

- Crucible temperature difficult to regulate
  - → T << 1600 °C : incomplete reduction ?
  - → T > 1700 °C : bonding of Gd and Ta
- Zr: purity and grain size ?
- Gd_2O_3 powder : hygroscopic + polluted ?
- homogeneity of the Gd/Zr mixture ?
- choice of the backing: Ta or … ?

Target production with SIDONIE separator



SIDONIE separator
@ IJCLab




ARRONAX @ Nantes

2nd production method
(compatible and complementary with the 1st one)

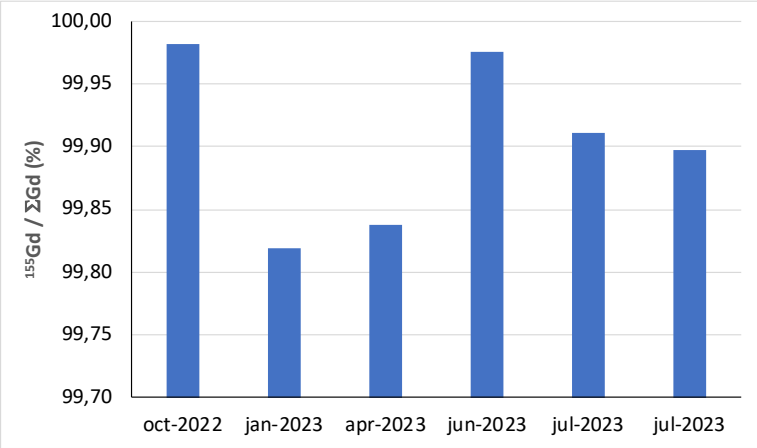
target characterisation :
- chemical purity, homogeneity: RBS, PIXE, MEB
- **isotopic purity** :
  neutron activation (PAA @ LVR-15, NPI-Řež),
  ICPMS (IRSN)

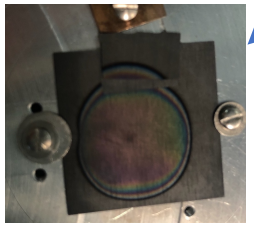
first measurements (PGAA) : $(9.6 \pm 4.1\ 10^{-6}) < {}^{157}Gd / {}^{158}Gd < (4.7 \pm 0.5\ \ 10^{-4})$
U. Köster et al.; Nucl. Inst. Meth. B 2020, 463, 111–114

first target: **^{155}Gd / ΣGd = (99.98 ± 0,04) %**

First ^{155}Gd targets with SIDONIE

6 SIDONIE targets currently available





^{152}Gd (%)	^{154}Gd (%)	^{155}Gd (%)	^{156}Gd (%)	^{157}Gd (%)	^{158}Gd (%)	^{160}Gd (%)	
0.2	*2.18*	*14.8*	*20.47*	*15.65*	*24.84*	*21.86*	*^{nat}Gd*
4.309 E-04	1.250E-03	99.9817	5.689E-03	5.329E-03	3.929E-03	1.640E-03	

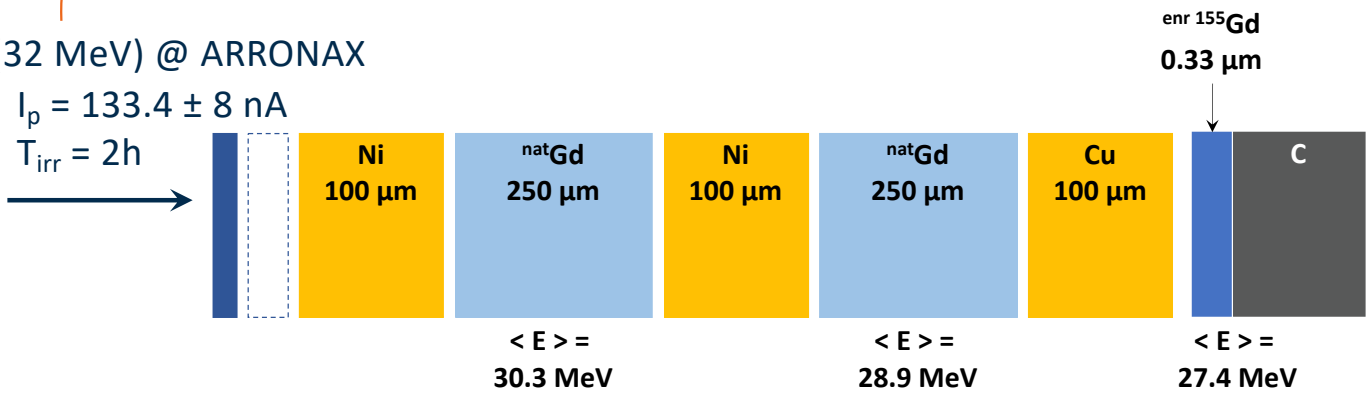
First test experiment (oct. 2022)

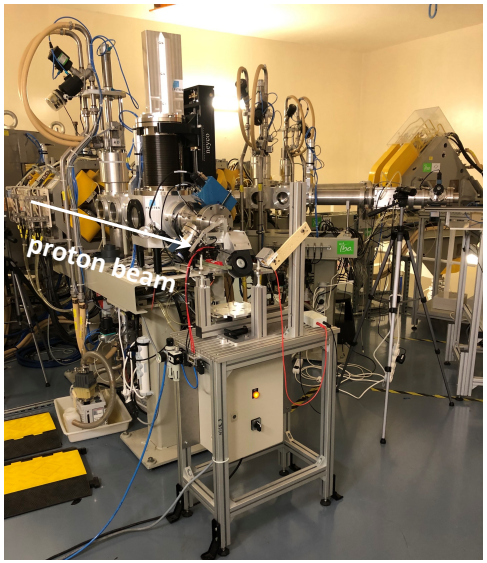
- Minimum E delivered by ARRONAX without degrading the beam
- Medical interest : Thick Target Yield → TTY ~ $\int_{threshold=10\ MeV}^{E_{\max\ medical\ cyclotron \lesssim 35\ MeV}} \sigma\ (E)$
- Optimum production ^{155}Gd (p,4n)^{152}Tb ~ 30 MeV

p (32 MeV) @ ARRONAX

I_p = 133.4 ± 8 nA

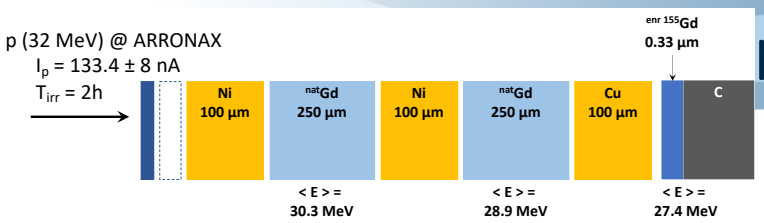
T_{irr} = 2h

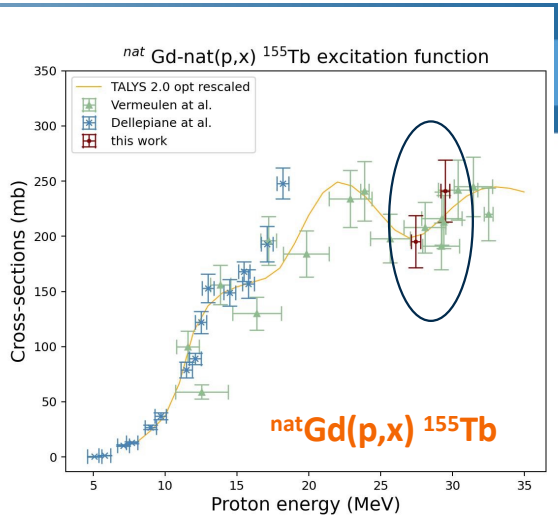


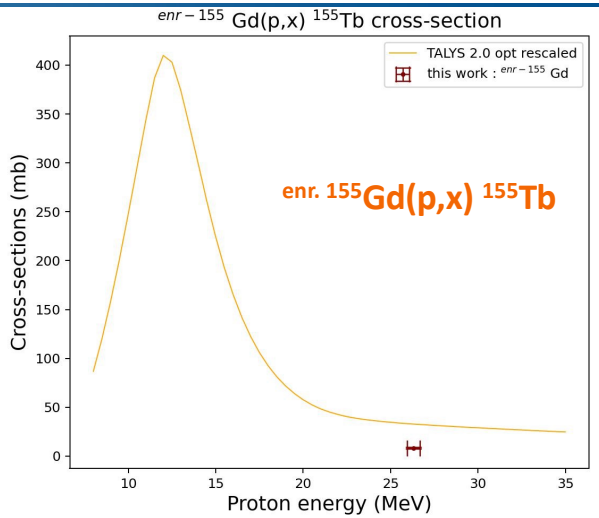


γ spectroscopy measurement

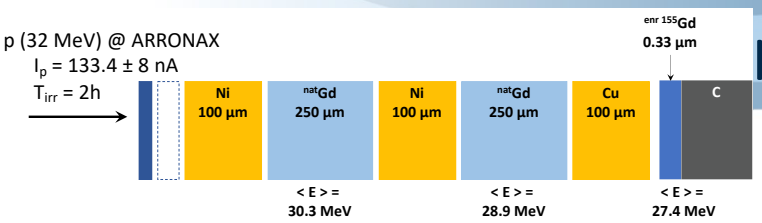
perimen

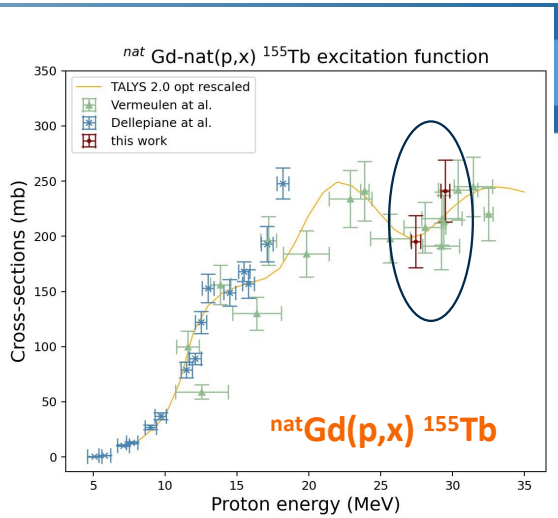

p (32 MeV) @ ARRONAX
I_p = 133.4 ± 8 nA
T_{irr} = 2h
Ni 100 µm
^{nat}Gd 250 µm
Ni 100 µm
^{nat}Gd 250 µm
Cu 100 µm
$^{enr\ 155}$Gd 0.33 µm
C
< E > = 30.3 MeV
< E > = 28.9 MeV
< E > = 27.4 MeV

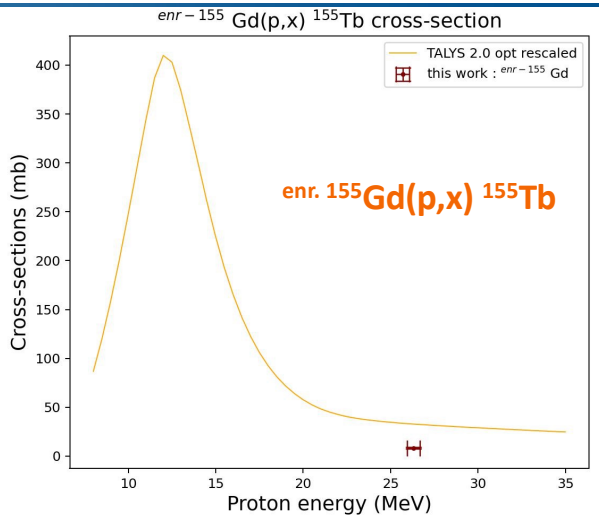

^{nat} Gd-nat(p,x) ^{155}Tb excitation function
TALYS 2.0 opt rescaled
Vermeulen at al.
Dellepiane at al.
this work
^{nat}Gd(p,x) ^{155}Tb
Cross-sections (mb)
Proton energy (MeV)


$^{enr-155}$ Gd(p,x) ^{155}Tb cross-section
TALYS 2.0 opt rescaled
this work : $^{enr-155}$ Gd
enr. ^{155}Gd(p,x) ^{155}Tb
Cross-sections (mb)
Proton energy (MeV)

perimen



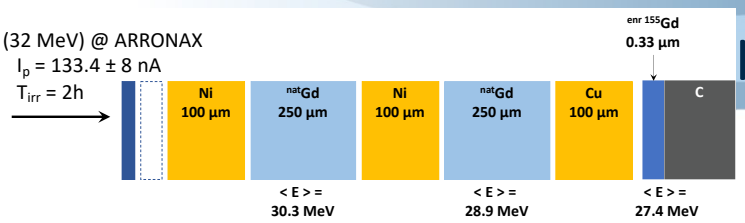




main pollution of concern : ^{156}Tb

High E γ rays could degrade image quality

	^{155}Tb		^{156}Tb	
$T_{1/2}$	5,32 days		5,34 days	
EC main γ rays	86,54 keV 105,3 keV 180,1 keV 262,3 keV	I = 32 % I = 25,1 % I = 7,5 % I = 5,3 %	88 keV 199,2 keV 356,3 keV 534,3 keV 1065,1 keV 1154,1 keV 1222,4 keV 1421,7 keV	I = 18 % I = 41 % I = 13.6% I = 67 % I = 10.8 % I = 10,4 % I = 31 % I = 12 %



perimen

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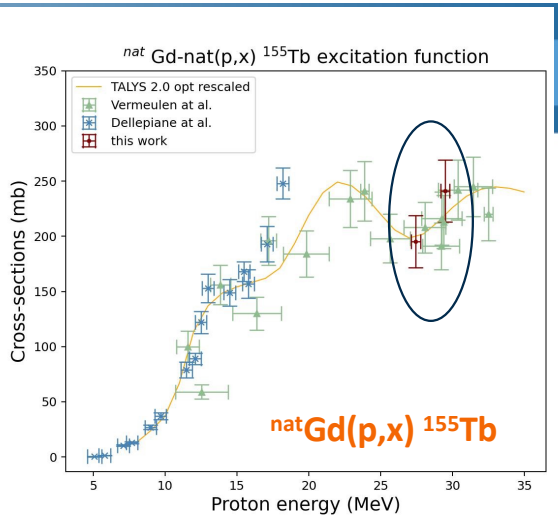
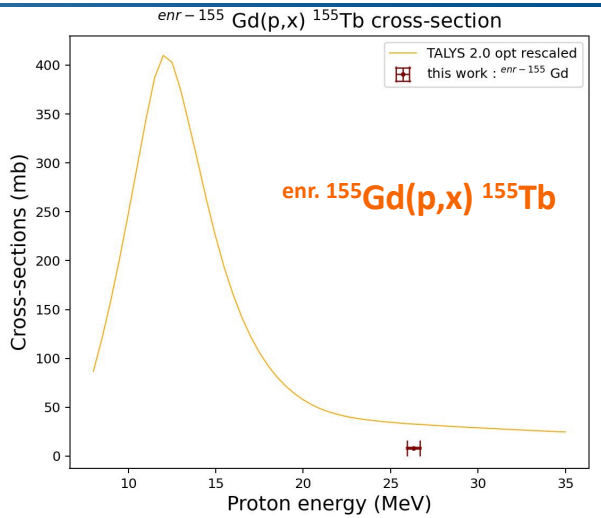
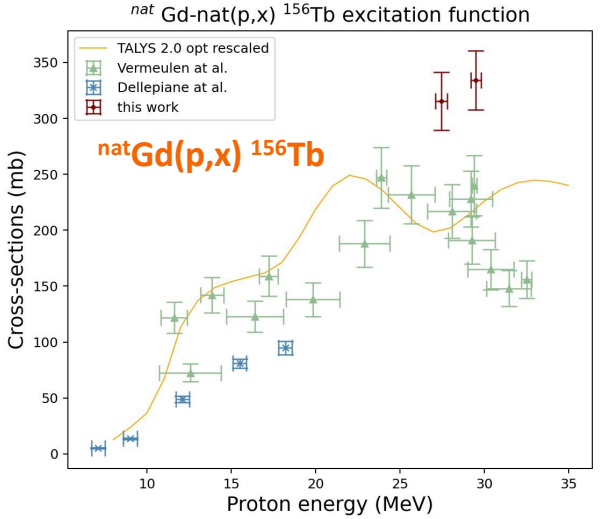
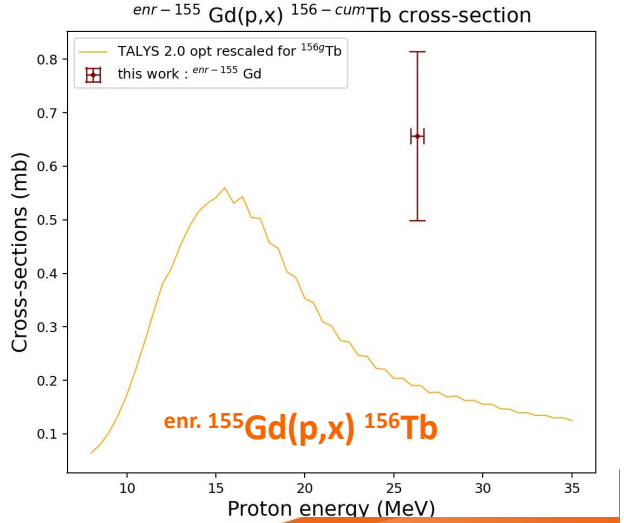
$\frac{Tb-155}{Tb-156}$ yield for ^{nat}Gd	$\frac{Tb-155}{Tb-156}$ yield for $^{enr\text{-}155}$Gd
0.6 ± 0.1	$14. \pm 5.$

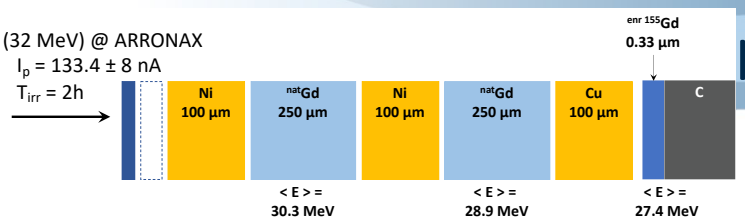
normalized for 1 µA current, 1h irradiation
^{nat}Gd thickness scaled to $^{enr\text{-}155}$Gd thickness



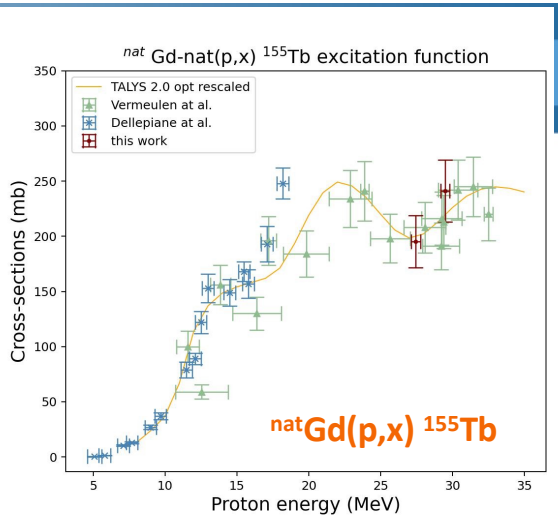


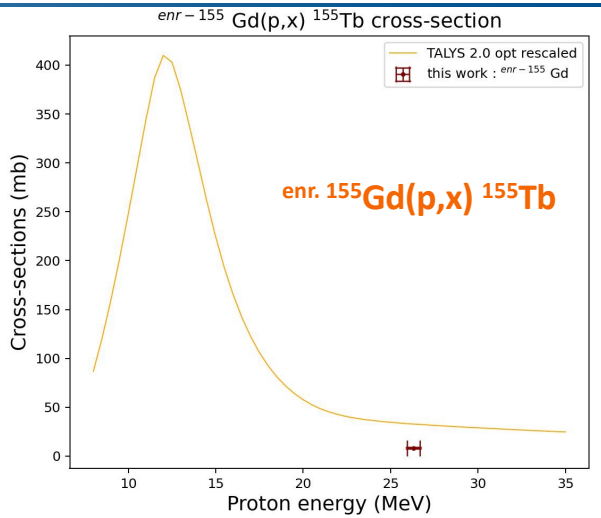






perimen





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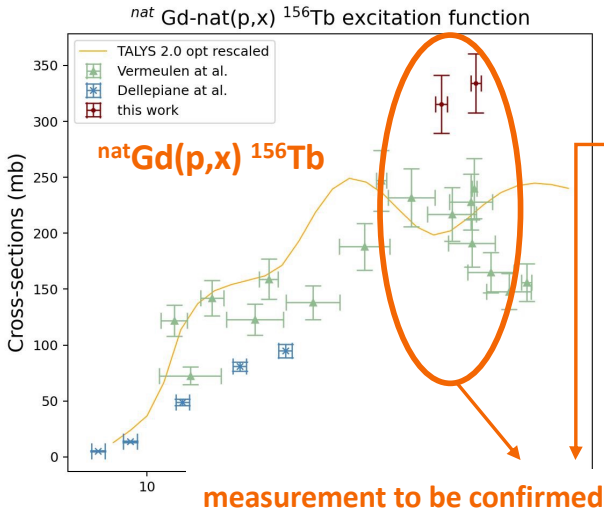

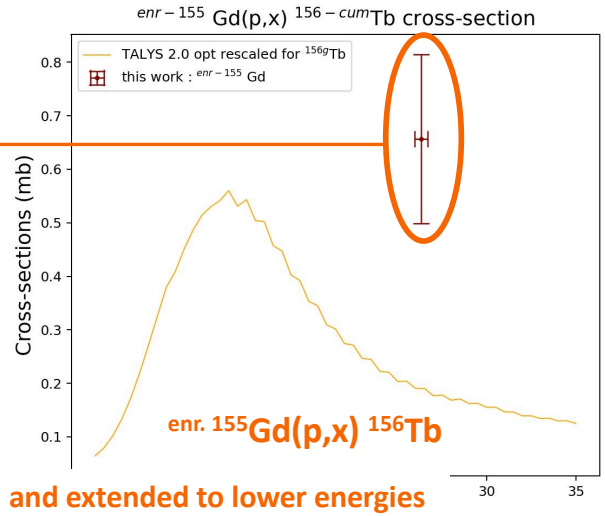

$\frac{Tb-155}{Tb-156}$ yield for ^{nat}Gd	$\frac{Tb-155}{Tb-156}$ yield for $^{enr\text{-}155}$Gd
0.6 ± 0.1	14. ± 5.

normalized for 1 µA current, 1h irradiation
^{nat}Gd thickness scaled to $^{enr\text{-}155}$Gd thickness

measurement to be confirmed and extended to lower energies
→ next experiment: Oct 2023 @ NPI-ReZ (J.Mrazek)

Outlook and conclusions

- Alternative method production with cyclotron of ^{155}Tb proposed
  - ➢ first pure targets produced: ^{155}Gd/ΣGd > 99,9 %
  - ➢ « test experiment » at 30 MeV
    - → development/validation of the methodology
    - → highlighted unavoidable pollution of ^{156}Tb

- Measurement for excitation function of ^{155}Gd(p,n)^{155}Tb (∼10 MeV – 30 MeV)
  optimization production rate % purity
  - ➢ next experiment on october (NPI @ ReZ)
  - ➢ foreseen experiments @ IPHC, Strasbourg
  - ➢ possible at NFS but planning not optimal …
- Project of imaging with mixture ^{155}Tb + ^{156}Tb (coll. CHUV, Lausanne)

… ^{155}Tb production « recipe » : → M.Bouteculet PhD Aug.-Sept. 2025
→ dec. 2025: end of ANR TTRIP

Thank you for your attention

C-O. Bacri
M. Bouteculet (PhD student from Sept. 2022)
M-A. Duval
H.Lefort
S.Lam (Ph.D student from Dec. 2022)
N.Pauwels
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V. Zinovyeva

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(G.Frémont)
C.Stodel

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M. Meyer

A.Guertin
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Y.Wang

J.C. Chambron
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G.Phan
Laboratoire de recherche en radiochimie, spéciation et imagerie (LRSI)
Laboratoire sur le devenir des pollutions des sites radioactifs
A.Gourgiotis

ANR-21-CE19-0037-01
TTRIP
Tools for Terbium RadioIsotopes Production for nuclear medicine

Thank you for your attention

and all the technical staff

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and

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