UNIVERSITÉ DE NANTES

Subatech

ARRONAX

Development of Enriched Gadolinium Target for Cross Section Measurement and Future Production of Terbium

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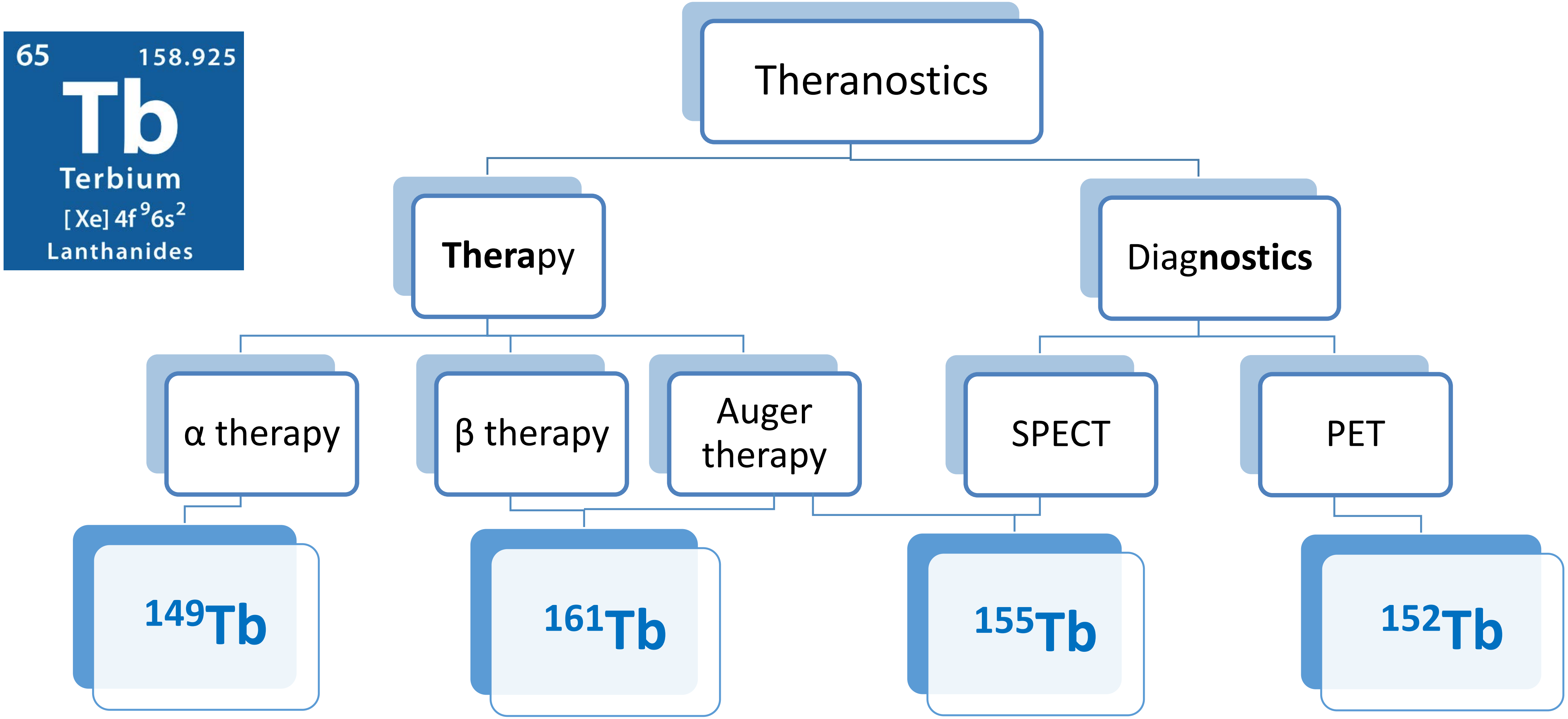
Terbium: a competitive candidate for theranostics



Production routes of Tb radionuclides

^{161}Tb

Produced by nuclear reactors: large quantity and good quality

- $^{160}Gd(n,\gamma)^{161}Gd \rightarrow ^{161}Tb$

☺

$^{149,152,155}Tb$

Produced by spallation reactions: limited quantity and complex procedure

- High energy (1,4 GeV) and many impurities
- Low efficiency to separate impurities (1%)

☹

This work: produce $^{149,152,155}Tb$ **with enriched Gd targets by cyclotrons**

$^{149,152,155}Tb$

Lower energy, lower cost, less impurities

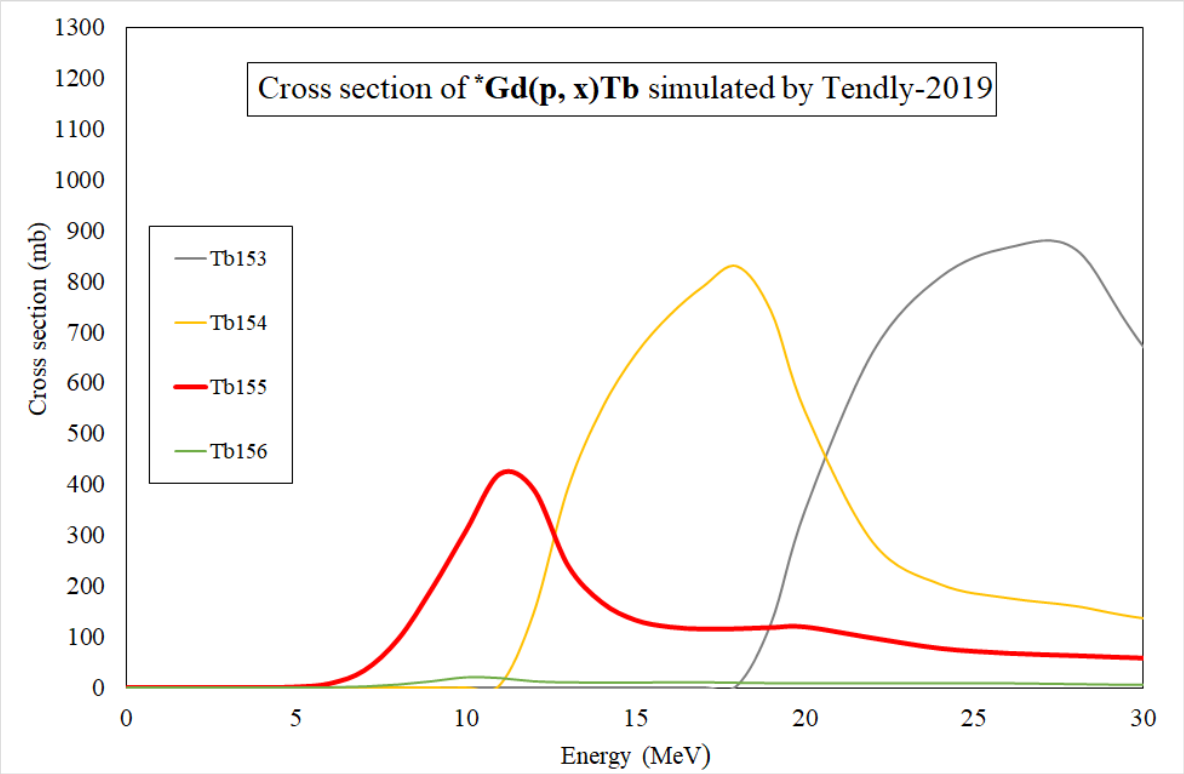
- ^{154}Gd (p, 6n) ^{149}Tb (enrichment of ^{154}Gd: 64%)
- ^{152}Gd (p, n) ^{152}Tb (enrichment of ^{152}Gd: 30%)
- **^{155}Gd (p, n) ^{155}Tb, ^{155}Gd (d, 2n) ^{155}Tb (enrichment of ^{155}Gd: 90%)**

☺

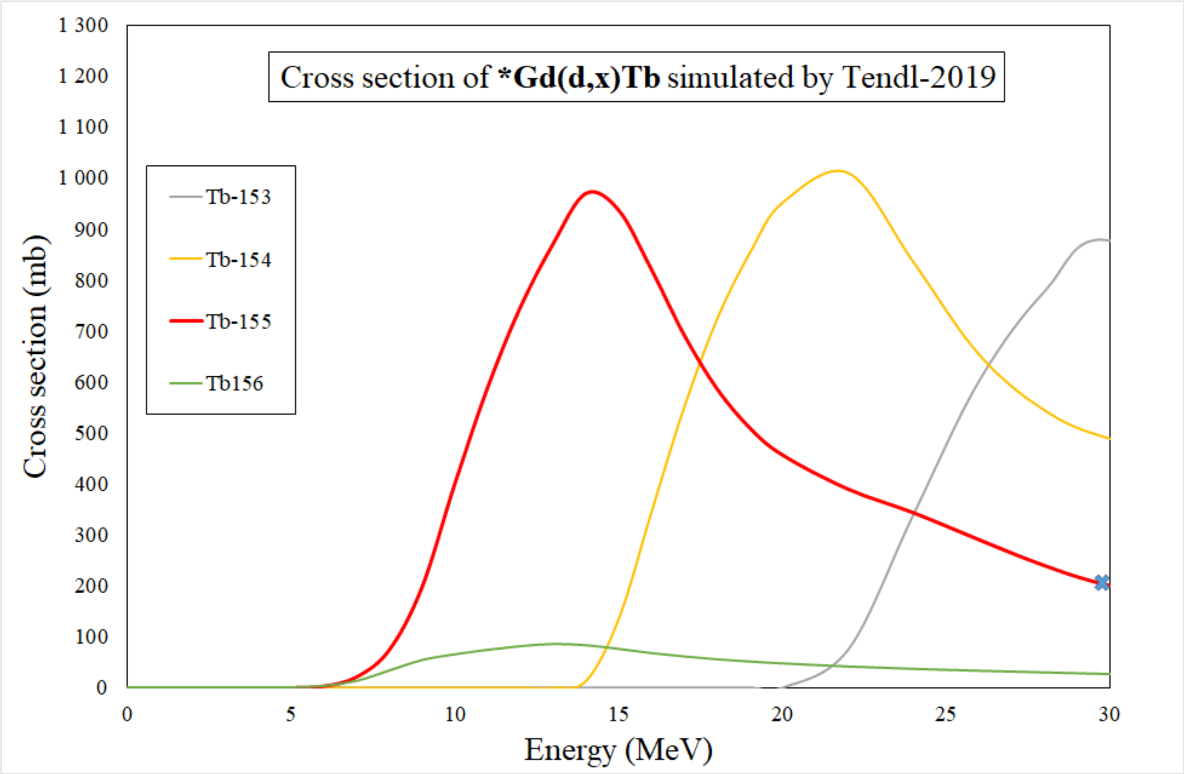
Objectives : produce ^{155}Tb using enriched Gd

Prepared enriched Gd_2O_3:

Isotopes	Gd-155	Gd-156	Gd-157	Gd-158	Gd-160
Proportion (%)	**92,8**	5,7	0,8	0,5	0,2



By proton: 400 mb



By deuteron: 1000 mb

Objectives : produce ^{155}Tb using ^{155}Gd

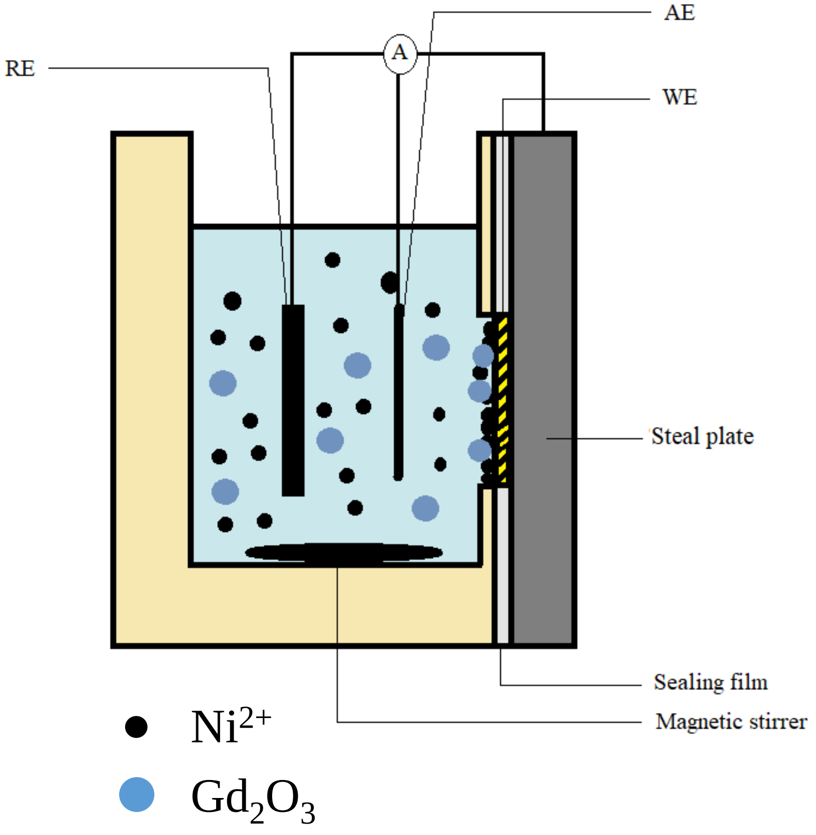
1 **Cross section measurement of the reaction $^{155}Gd(d,n)^{155}Tb$ to estimate production yield**

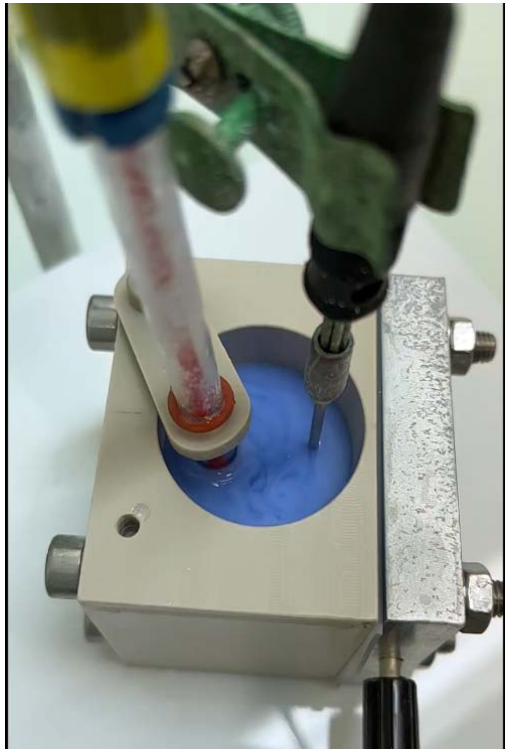
- Make very thin targets (10-20 µm)

2 **Manufacture a complete and uniform Gd-containing target for the routine production of Tb**

- Make thicker targets (≥ 200 µm)

Thin targets fabrication: Co-electrodeposition method





WHY?

The reduction potential of Gd^{3+}/Gd is too negative (-2,3 V/ENH) to obtain an adhered deposit in aqueous solutions because of the HER.

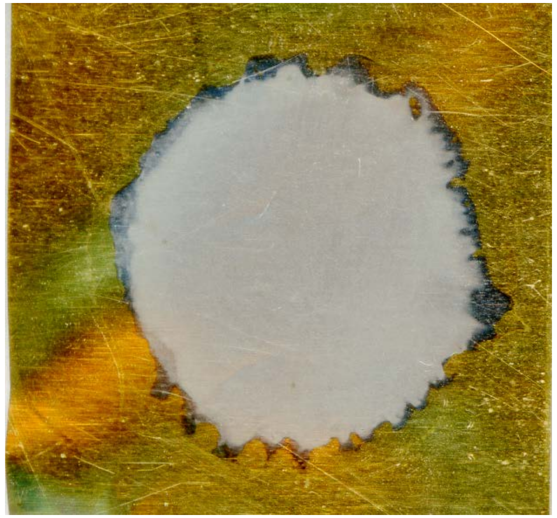
HOW?

Insoluble Gd_2O_3 particles in alkaline solution are mixed in the Ni plating bath, by applying a potential, the Gd_2O_3 particles will move with Ni^{2+} and trapped in the Ni metal matrix (mechanical process).

WHAT?

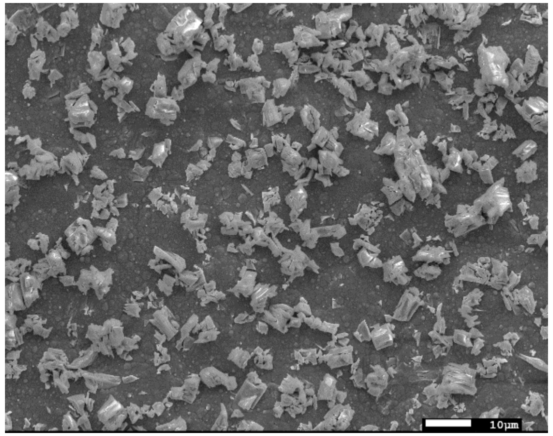
A Ni-Gd_2O_3 composite target with a thickness of 10-20 µm can be obtained.

Obtained targets: thin and homogenous

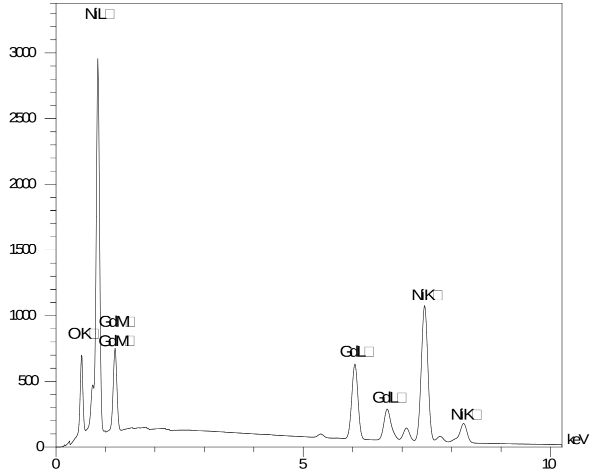


Picture of a prepared thin target
Thickness = 12,96 µm
Gd trapped in the deposit: 2,61 mg

➢Morphological analysis: SEM



E= 15kV, zoom x1000



E = 15kV, zoom x100
Temps de mesure = 60 s

➢Composition analysis: DRX et ICP-OES

According to the ICP-OES results, the target contains 3 mg of Gd atoms.

The cross section measurement

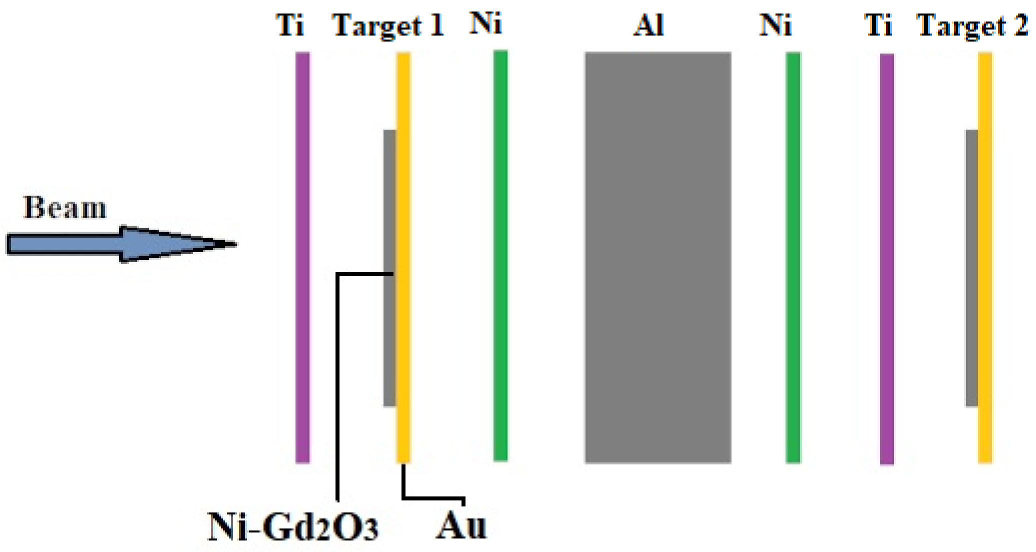
- The cross section represents the probability that a nuclear reaction will occur.
- $\sigma = \dfrac{\text{number of reactions per unit time per target nucleus}}{\text{number of incident particiles per unit time per unit area}}$

$$\sigma\,(E) = \frac{Act(E).\,A}{I \cdot N_A.\,\rho \cdot \delta x \cdot (1 - e^{-\lambda t})}$$

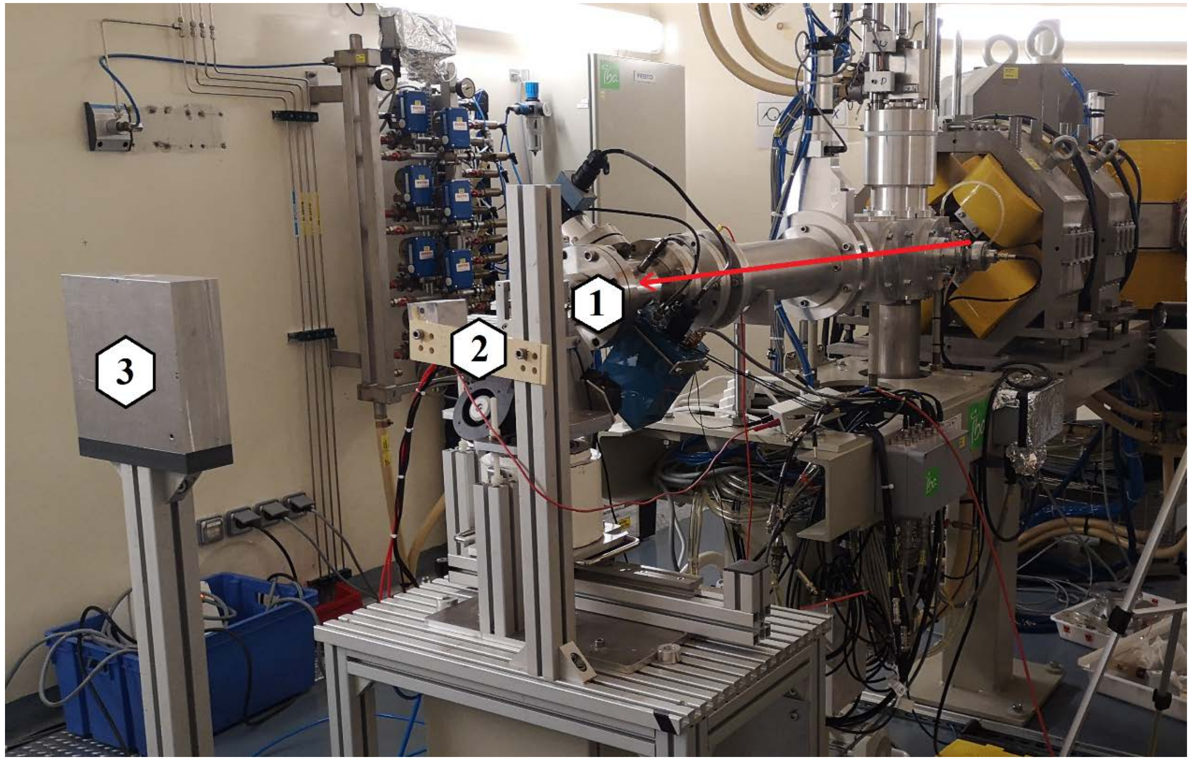
$Act(E)$ is the activity of nuclei produced at energy E (Bq),
A is the molar mass of the target nuclei (g·mol^{-1}),
I is the number of the projectiles per second hitting the target (s^{-1}),
N_A is the Avogadro number (at·mol^{-1}),
ρ is the target material bulk density (g·cm^{-3}),
δx is the thickness of the target (cm),
λ is the decay constant of produced nuclei (s^{-1}),
t is the irradiation time (s).

The « stacked foils » method

Target: Ni/Gd_2O_3 composite target deposited on the Au substrate
Monitor: Ti and Ni foils
Degrador: Al foils



Schematic diagram of the stack composition



Picture of the beam line used at GIP ARRONAX cyclotron

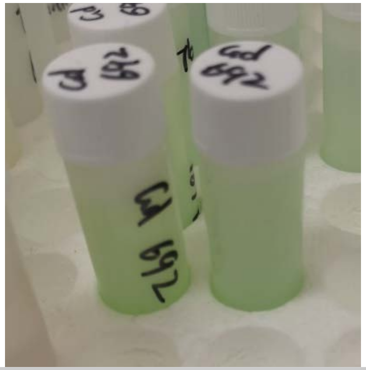
Irradiation and data acquisition

Irradiation:

Energy: 5-30 MeV

Beam intensity: 50-75 nA

Irradiation duration: 30 min



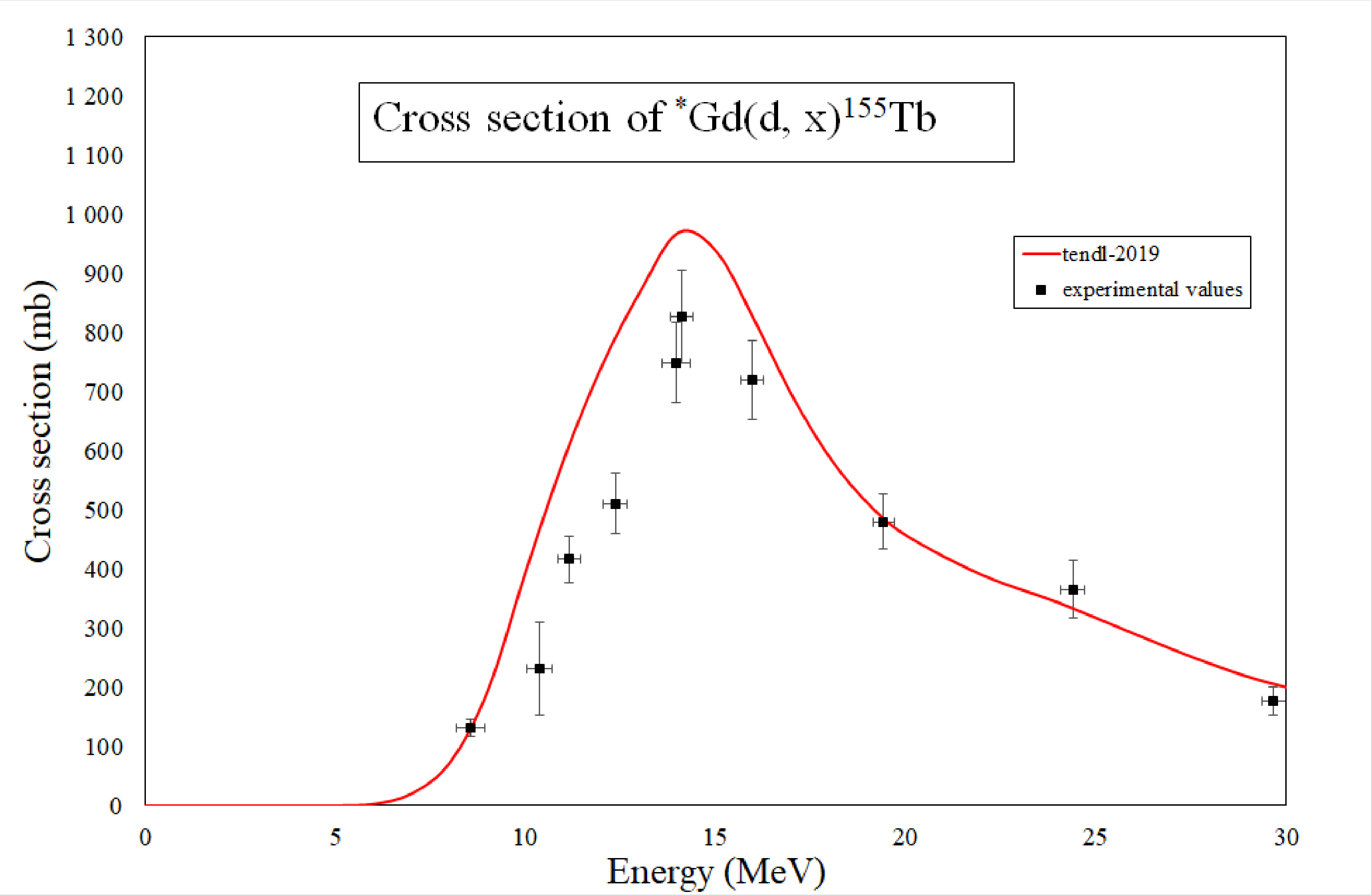
Data acquisition:

Determine the activity of targets and monitors by HPGe gamma detector.

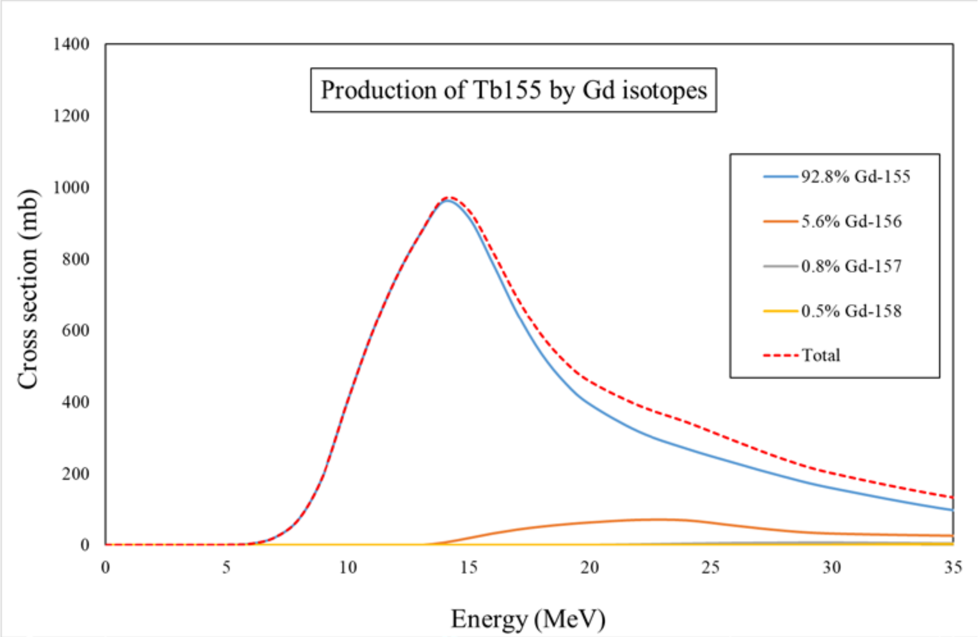
Dissolution:

Dissolved the target with 10 ml of concentrated HCl (12 M) for counting and for ICP analysis.

Cross sections measured for ^{155}Tb

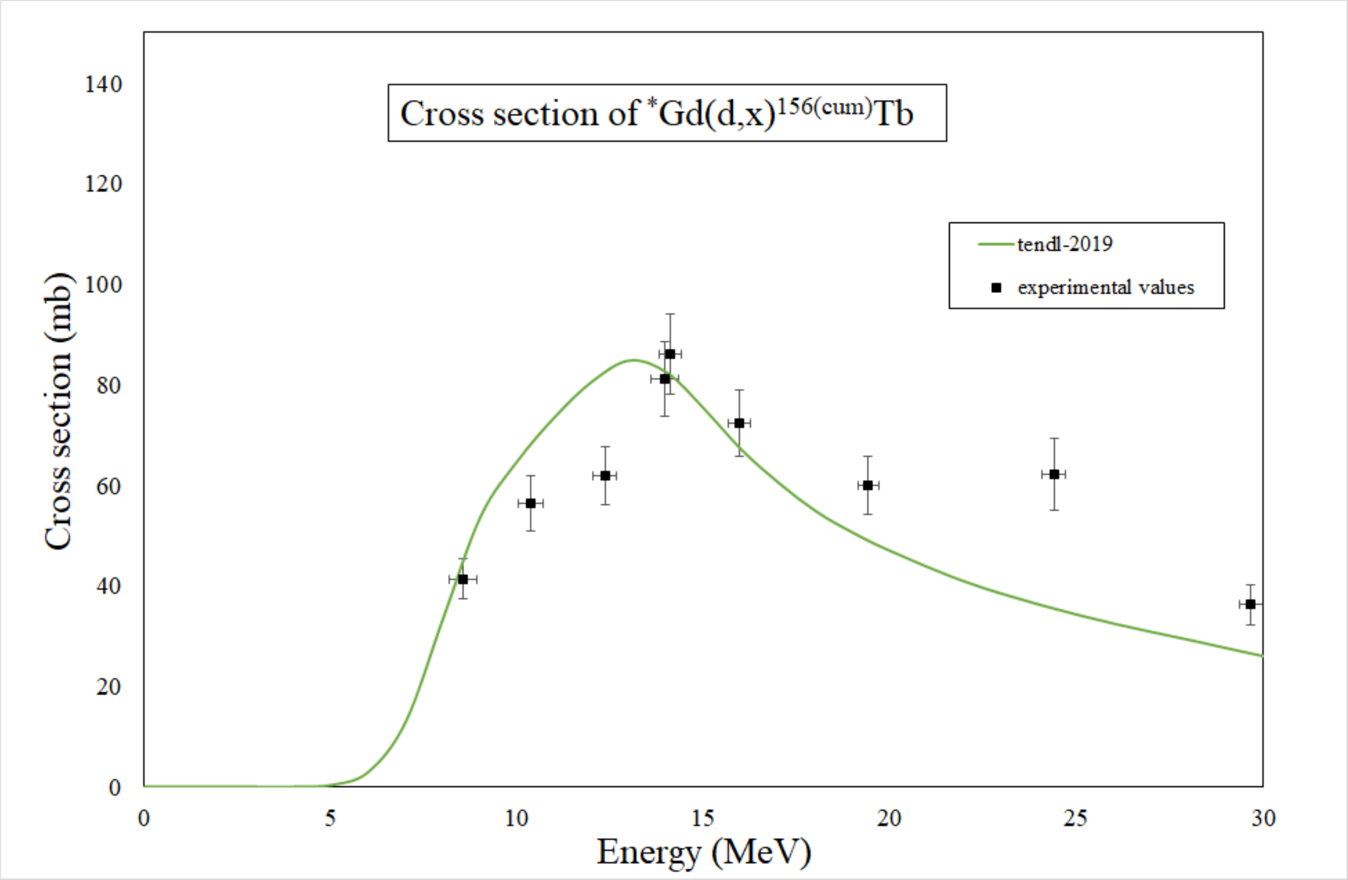


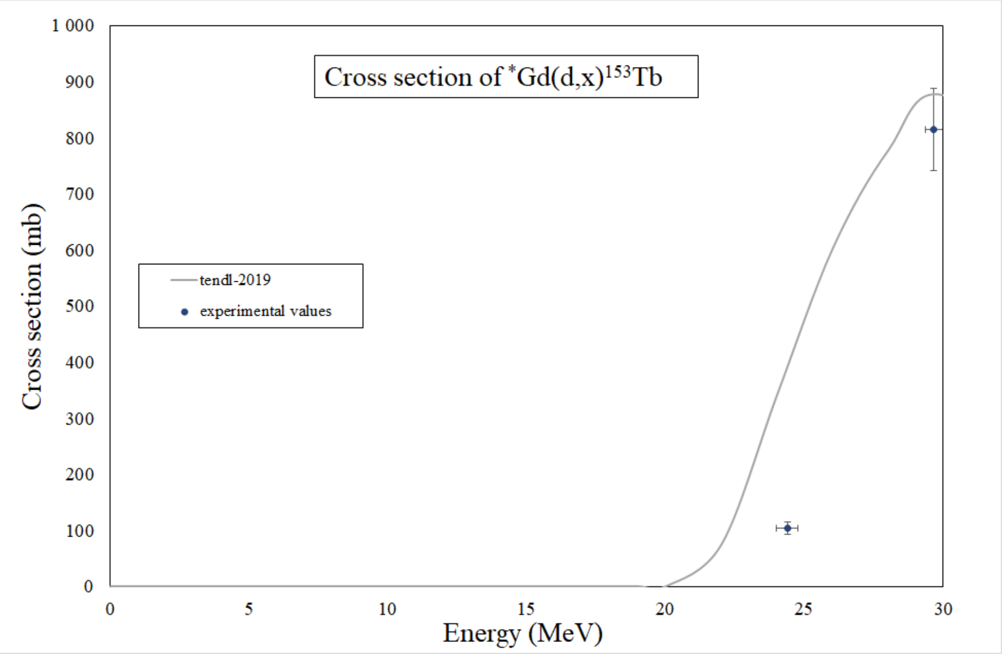
Results according to Ti monitors.
Gamma line to characterize ^{155}Tb: E_γ = 180,080 keV, I_γ = 7,5%

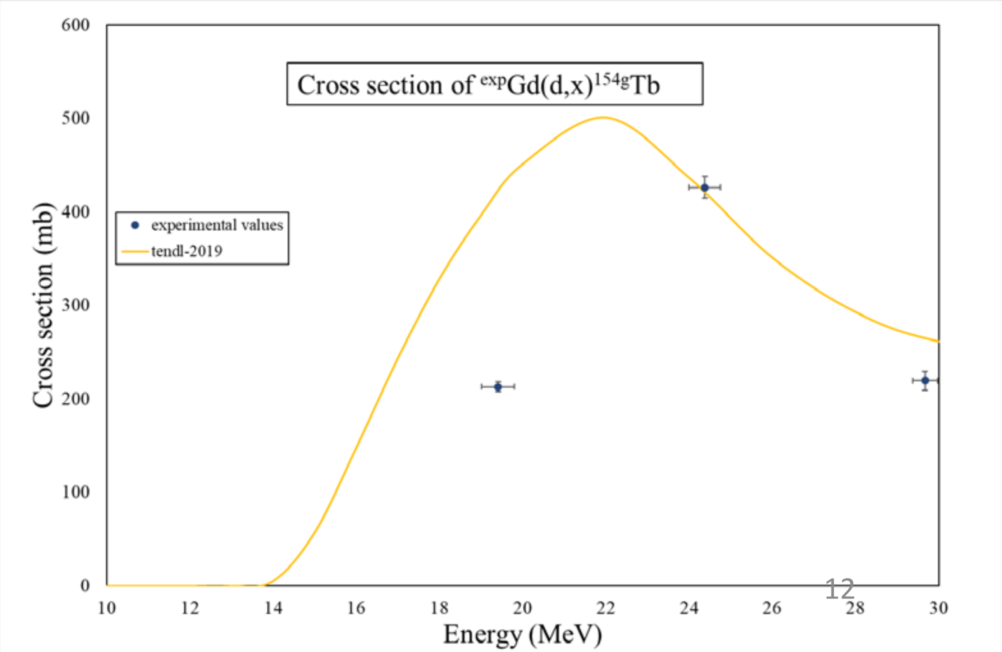


The contribution of ^{156}Gd starts when energy is bigger than 13 MeV.
We got 4 measured cross section of the reaction $^{155}Gd(d,2n)^{155}Tb$.

Cross sections measured for $^{153,154,156}Tb$







When E is smaller than 14 MeV, only ^{156}Tb will be produced as impurity.
The minimum ratio of ^{156}Tb and ^{155}Tb ($r = \frac{A_{Tb156}}{A_{Tb155}}$) is 7% at 0-14 MeV.

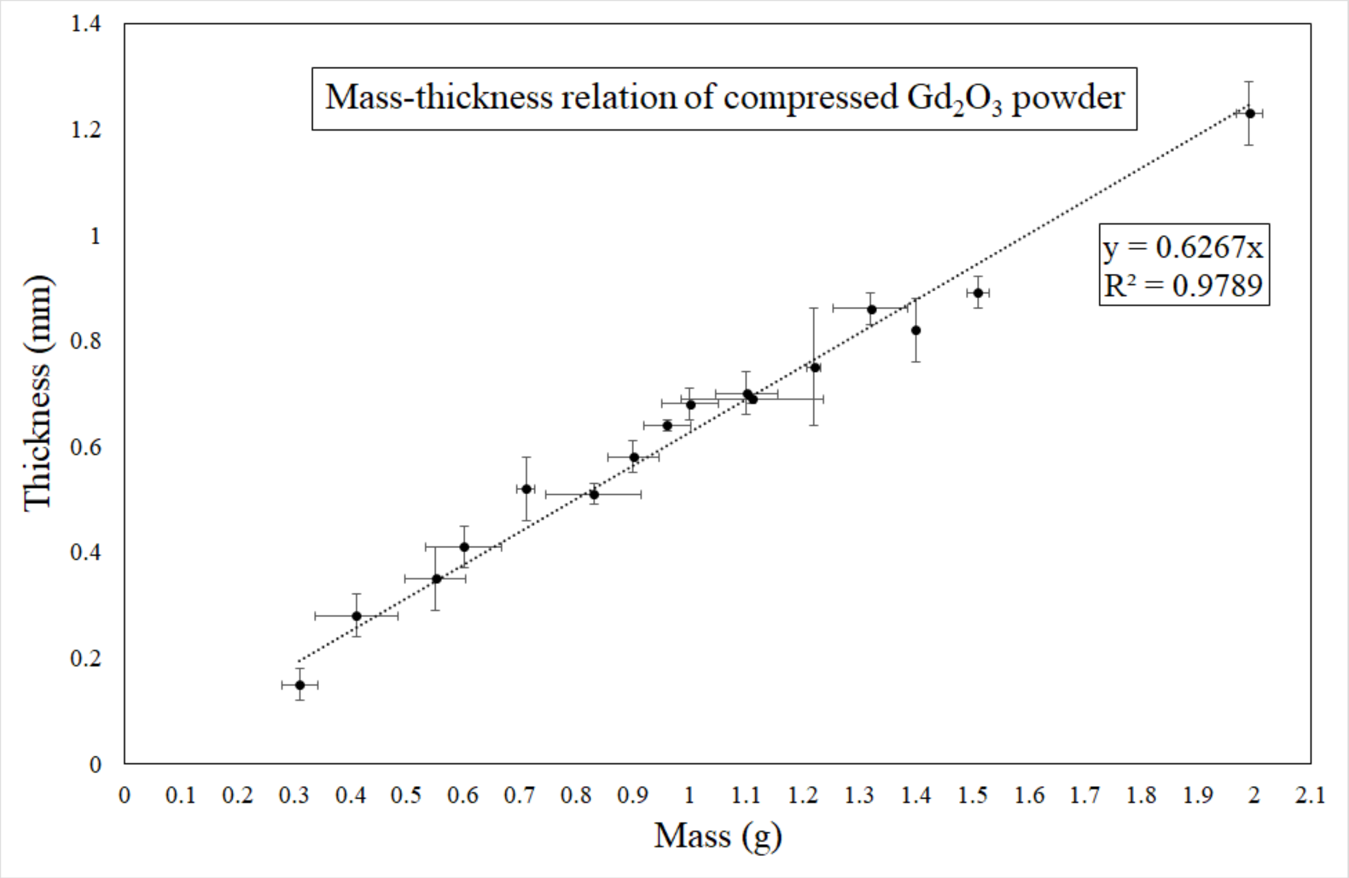
Thick targets fabrication: pelletizing method

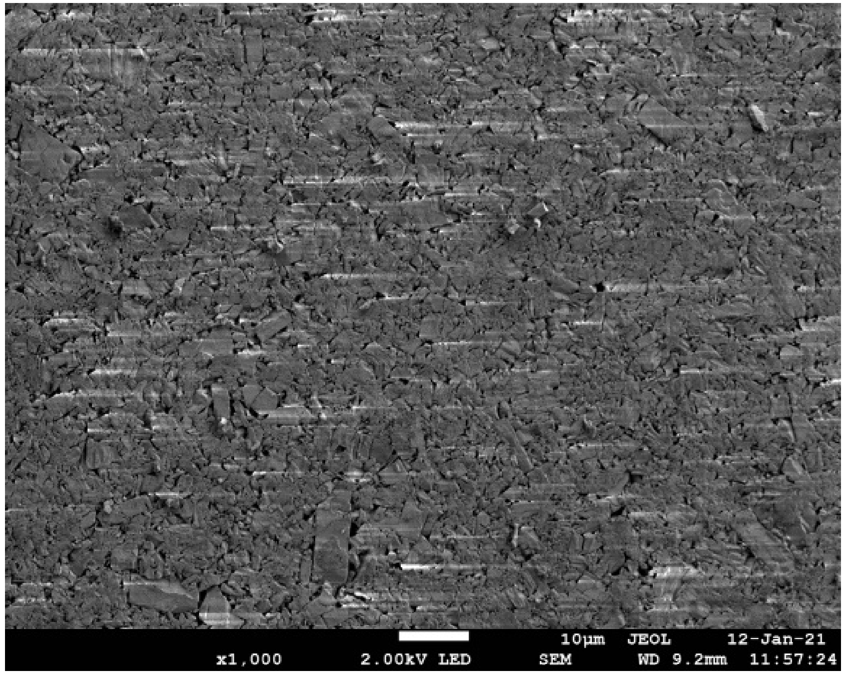
Presse hydraulique
Pression max = 38 t

The 20-mm-diameter pellet die

Pastille de Gd_2O_3 naturel
Épaisseur 1,07mm
Diamètre 20 mm

Obtained pellet: compact and homogenous





SEM image of the surface of a pellet
E= 15kV, zoom x1000

Pressure: 600 bar (for 60 s)

Obtained density: 5,3 g/cm³ (Theoretical density: 7,1 g/cm³)

Conclusion and perspectives

Work completed:

✓Cross section:

The cross sections of *Gd(d,x)^{155}Tb and other impurities have been measured using Ni-Gd_2O_3 composite targets.

✓Pelletizing:

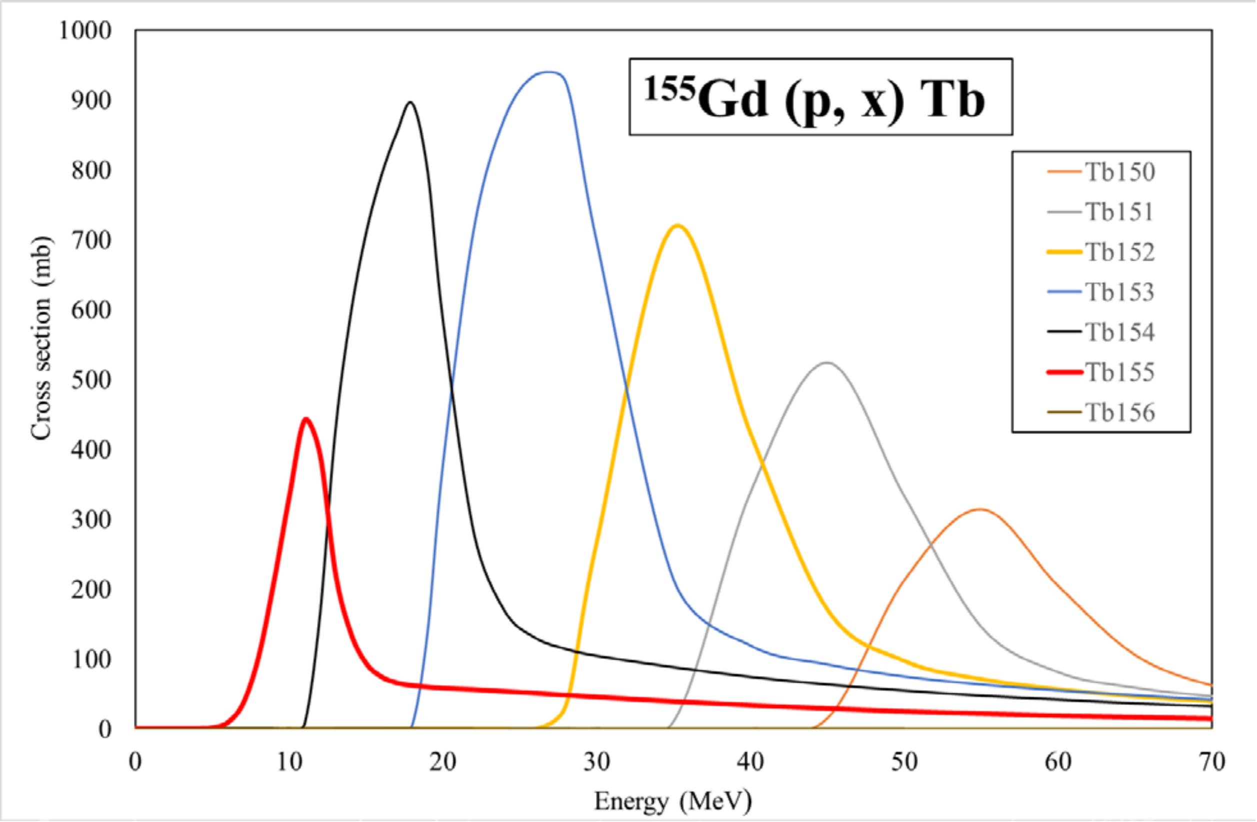
Solid, compact and thick targets have been manufactured.

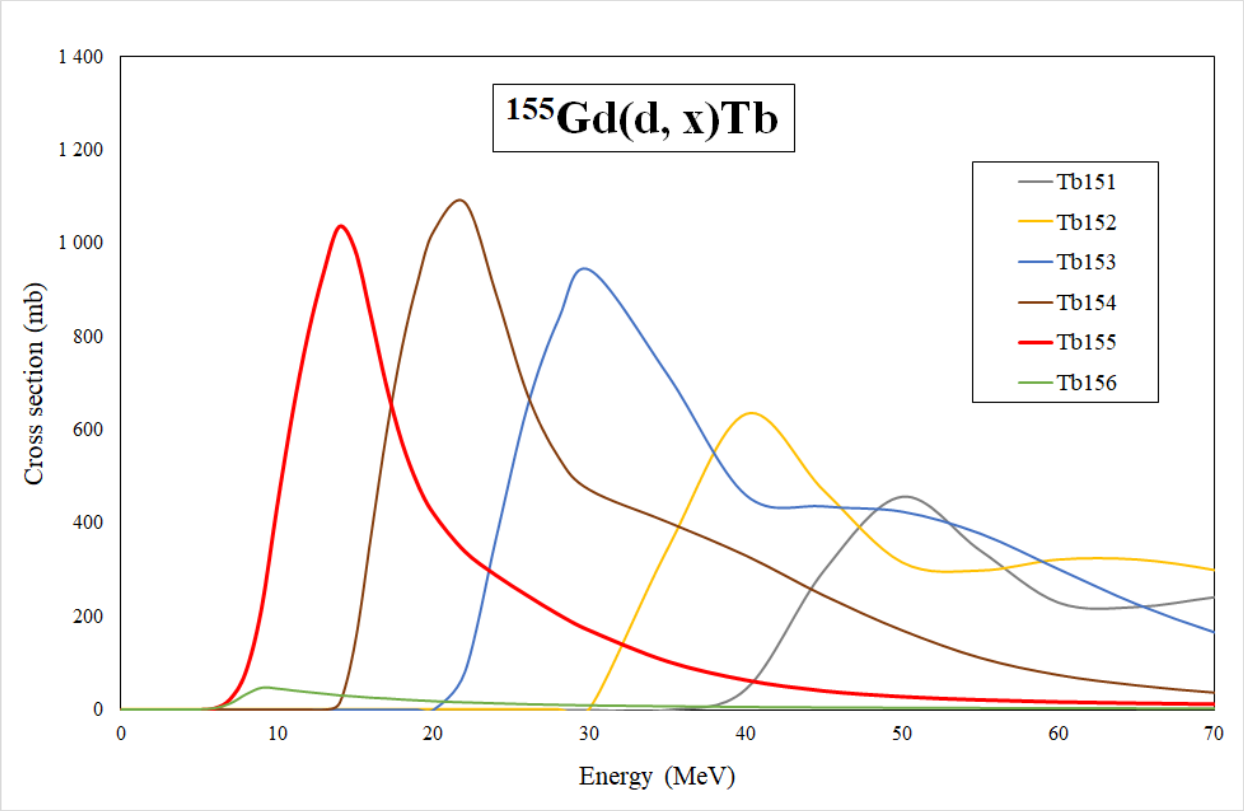
Follow-up:

❑ Irradiation of thick targets.

❑ Estimation of the production yield.

Production with 100% ^{155}Gd





Thank you!