

R&D for the production of innovative radioelements at GANIL

C. Barthe-Dejean, M. Fadil, G. de France, F. De Oliveira, H. Franberg, A.M. Frelin-Labalme, X. Ledoux, M. Michel, M.H. Moscatello, S. Perat, C. Stodel

- Motivations and opportunities

- Current projects
 - At-211
 - Other α emitters
 - Dosimetry
 - Liquid target

- Motivations and opportunities

- Current projects
 - At-211
 - Other α emitters
 - Dosimetry
 - Liquid target

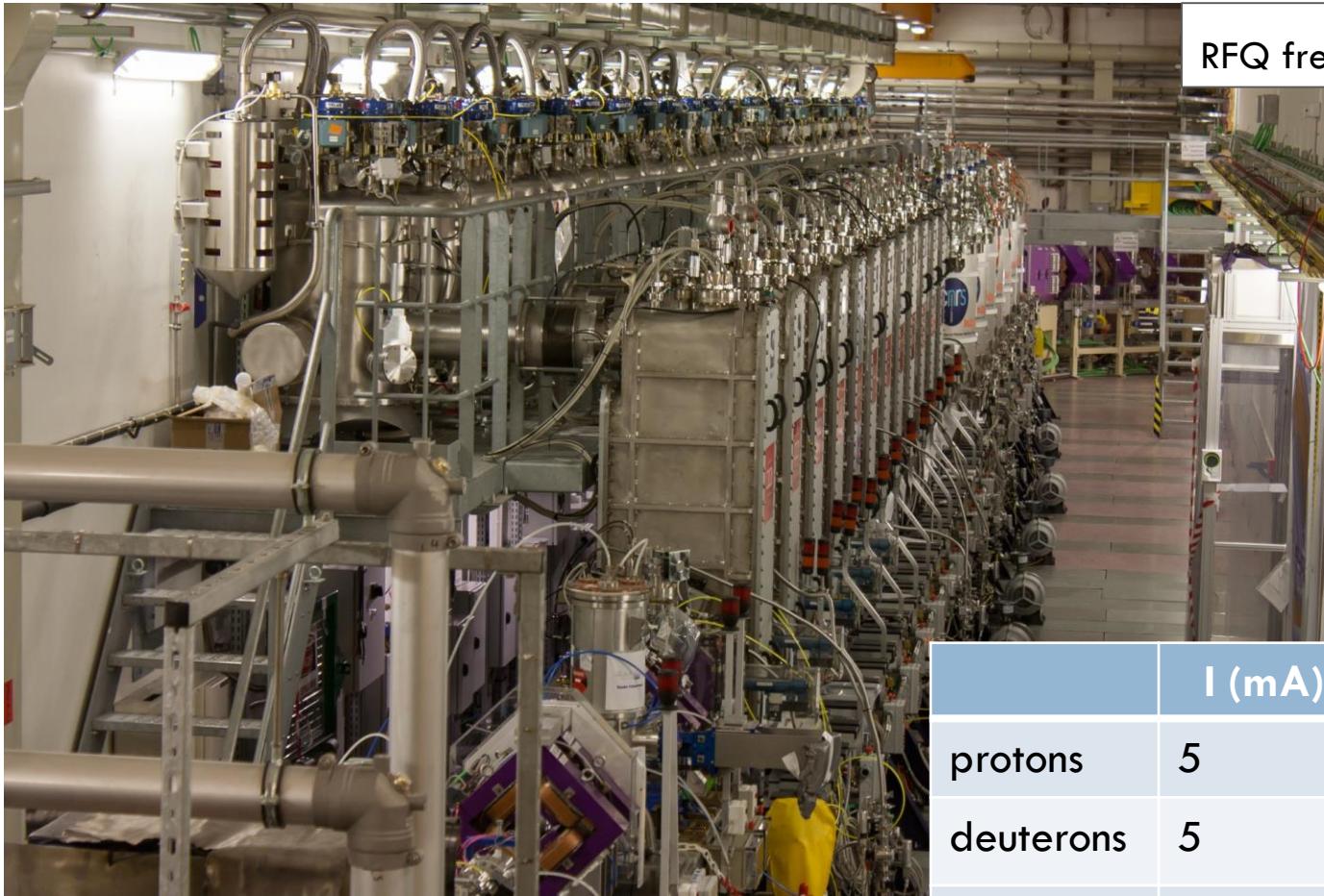
- A strong request from society which research labs like GANIL can partly address
- An R&D on the production of innovative radioelements for nuclear medicine
- A new field to exploit at best the potentialities of the new LINAC
- Endorsed by the GANIL Scientific Council
- In the SPIRAL2 blue paper
- Outcome might serve fundamental research

Opportunities at SPIRAL2

- Limited current (cyclotrons):
 - Dedicated machines, not optimized for R&D production of new isotopes (fixed energy) => cross-section, contamination,...
 - Only proton, deuteron and/or alpha
 - Target not designed for high beam intensities

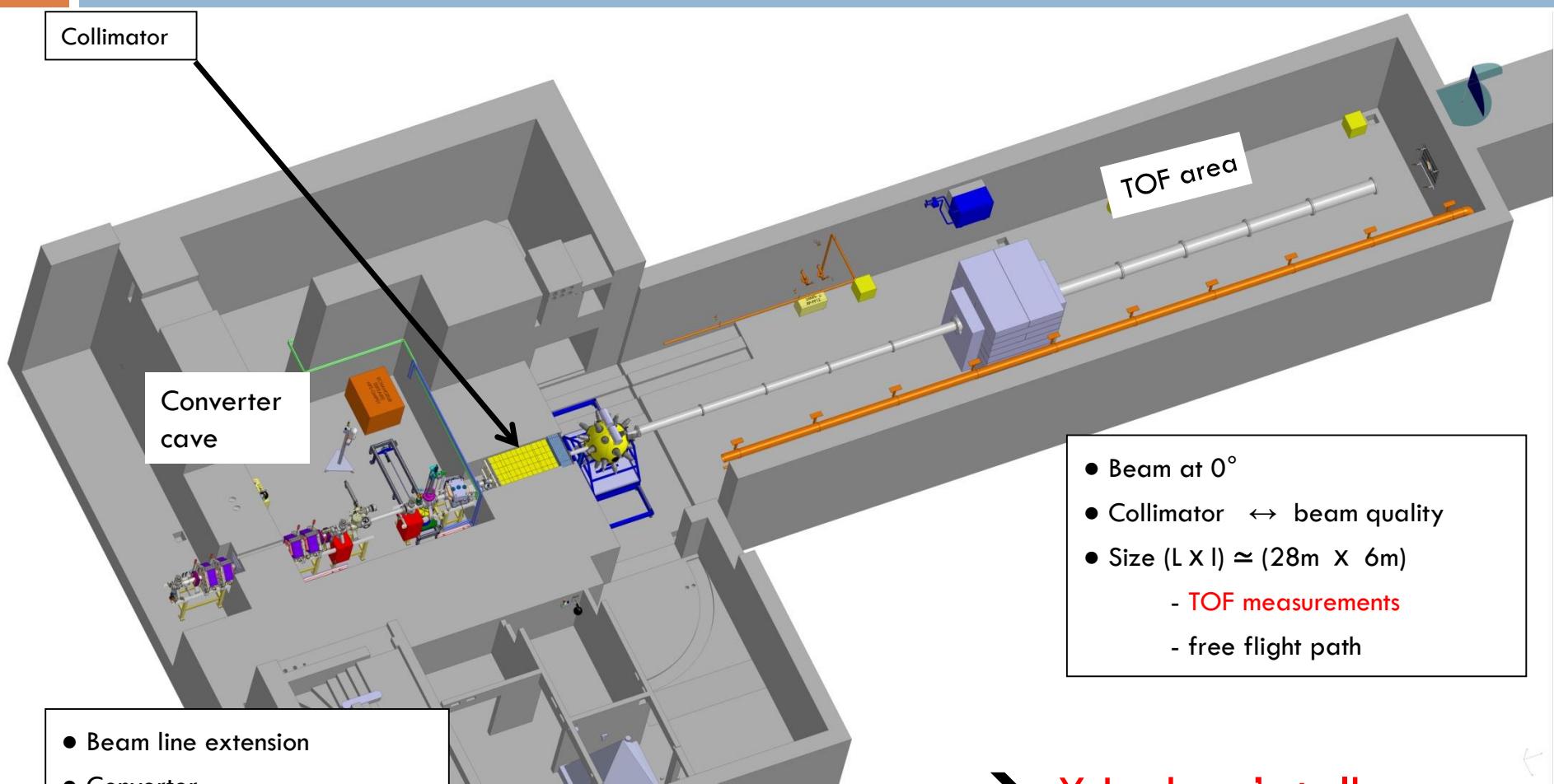
- New possibilities at SPIRAL2:
 - Very intense beams at optimal energy
 - New beams compared to existing production facility machines

The SPIRAL2 LINAC



RFQ frequency F = 88MHz

	I (mA)	Energy (MeV/u)
protons	5	2-33
deuterons	5	2-20
alpha	3	2-20
Ions (1/3)	1	2-14.5



→ X Ledoux's talk

Strategy for radio-isotopes

- **Strategy:**
 - Only R&D
 - Collaboration
 - Targeted goals
 - Focus on alpha emitters

- **Current isotopes of interest:**
 - ^{211}At (ARRONAX et al)
 - $\alpha+\text{Th}$ reaction products

- Motivations and opportunities

- Current projects
 - At-211
 - Other α emitters
 - Dosimetry
 - Liquid target

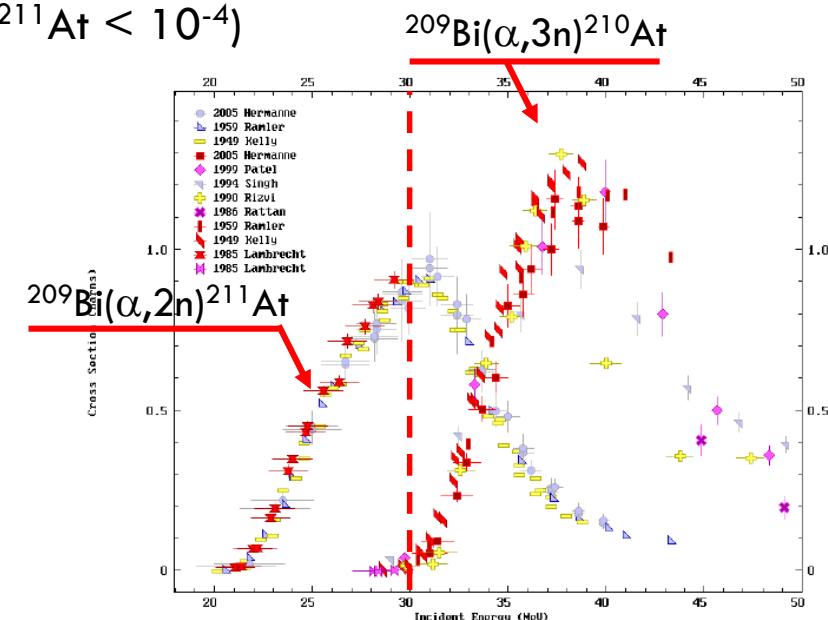
Production using $^{209}\text{Bi}(\alpha, 2n)^{211}\text{At}$

□ Réaction

- Reaction threshold 20.72 MeV
- Cross section max 900 mb at 30 MeV
- Production of contaminant ^{210}Po $^{209}\text{Bi}(\alpha, 3n)^{210}\text{At} \rightarrow ^{210}\text{Po}$, $t_{1/2} = 138$ d,
 $E_t = 28.61$ MeV
- Optimal energy ≈ 30 MeV (ratio $N^{210}\text{At}/N^{211}\text{At} < 10^{-4}$)

□ Production estimate:

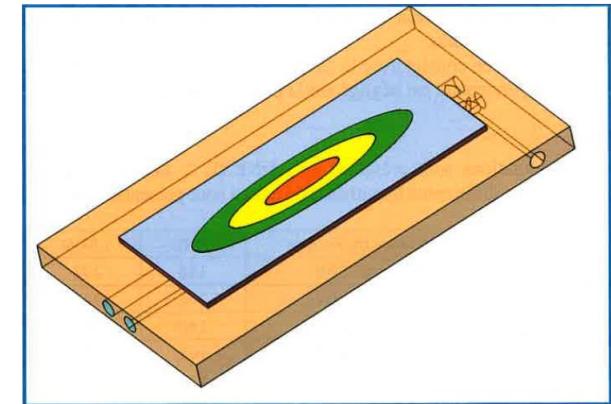
- Alpha 28 MeV, 1 kW
- Target : Bi on AlN backing
- At-211 production : 9.6 GBq in 8h



- Target from ARRONAX
- Irradiation at SPIRAL2
 - Irradiation station in the converter room
 - Beam: 28 MeV α , 70 μA ($3.15 \times 10^{14} \alpha/\text{s}$)
 - Tirr: 4 to 8h
 - Production of $\sim 10\text{GBq}$ of At-211
 - Dose Rate after 30 min cooling: 0.25 mSv/h at 30 cm
- Irradiated targets sent to ARRONAX
- Extraction, labelling, QC at ARRONAX

Thermal study

- 1kW deposited
- Two designs:
 - Target on cold copper
 - Direct backing water cooling
- Parameters:
 - Thermal resistance target-copper
 - Cooling liquid temperature



	résistance thermique	1KW	1,5KW	2KW
solution plaquée (eau 15°C)	100 mm ² .C/W	188	292	
	200 mm ² .C/W	252	387	
solution plaquée (éthanol -30°C)	100 mm ² .C/W	140	243	
	200 mm ² .C/W	202	340	
solution contact eau (eau 15°C)		156	228	309
solution contact eau (éthanol -30°C)		105	180	260

Tableau 4 : températures maximum de la cible calculées en °C

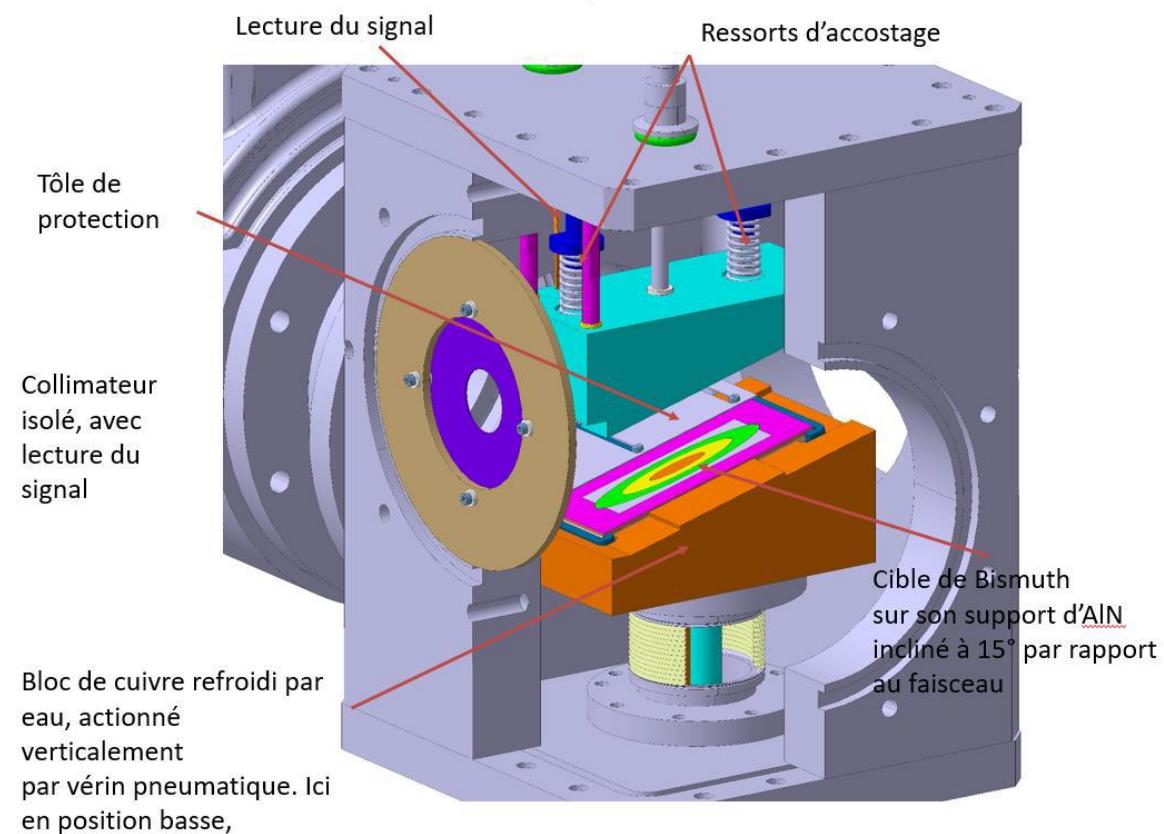
1 kW OK. Measurement of thermal resistance

T_{fusion} Bi = 271 °C

The irradiation station

- Manual load/unload
- Target on actuator (beam focusing before irradiation)
- Unload using vinyl confinement
- Dosimetry measurements
 - extremity : $480\mu\text{Sv}$
 - Full body : $20\mu\text{Sv}$
- Shipment : A type parcel

**Collaboration NPI
Under assembly**

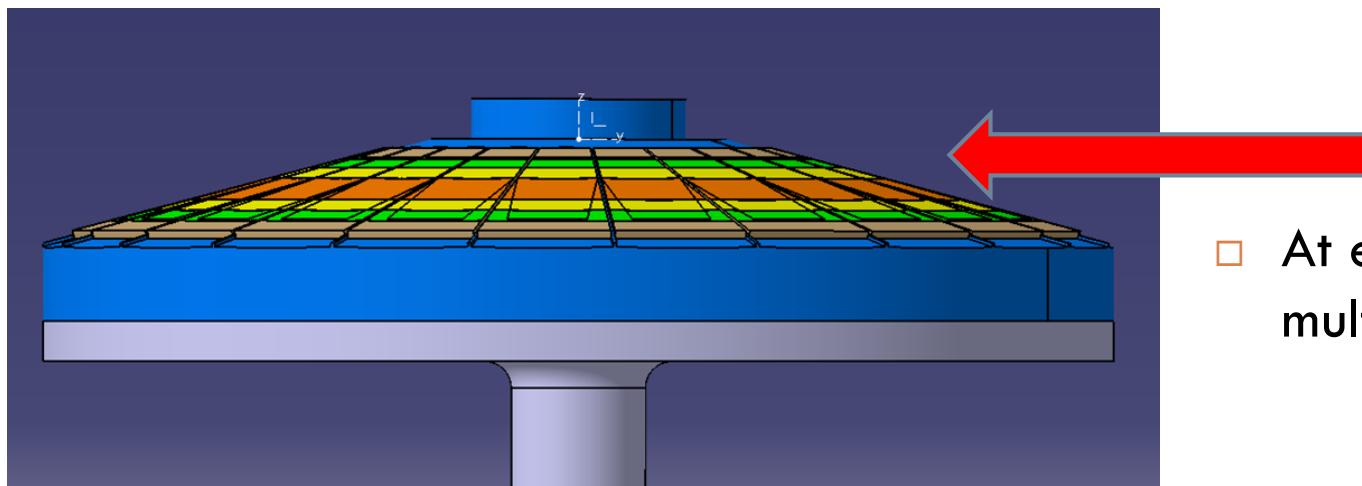


Higher power dissipation

Feasibility study

- Production via $\alpha + \text{Bi}$
- Collaboration NPI Rez
- Goal: $\sim 10 \text{ kW}$
- Rotating target+water cooling

\varnothing beam ($+/ - 3\sigma$) = 24mm
we kept the angle of the target of 15°



- At extraction with multiple targets?

^{211}At at NFS: generator

Design of a generator using $^{209}\text{Bi}(^6\text{Li},4\text{n})^{211}\text{Rn}$ or $^{209}\text{Bi}(^7\text{Li},5\text{n})^{211}\text{Rn}$

□ Reaction

□ Reaction threshold

- 28.5 MeV for $^{209}\text{Bi}(^6\text{Li},4\text{n})$
- 36.13 MeV for $^{209}\text{Bi}(^7\text{Li},5\text{n})$

□ ^{210}Po contamination

- $E(^6\text{Li}) < 36 \text{ MeV}$ for $^{209}\text{Bi}(^6\text{Li},4\text{n})$
- $E(^7\text{Li}) < 48 \text{ MeV}$ for $^{209}\text{Bi}(^7\text{Li},5\text{n})$

□ Energy domain of SPIRAL2

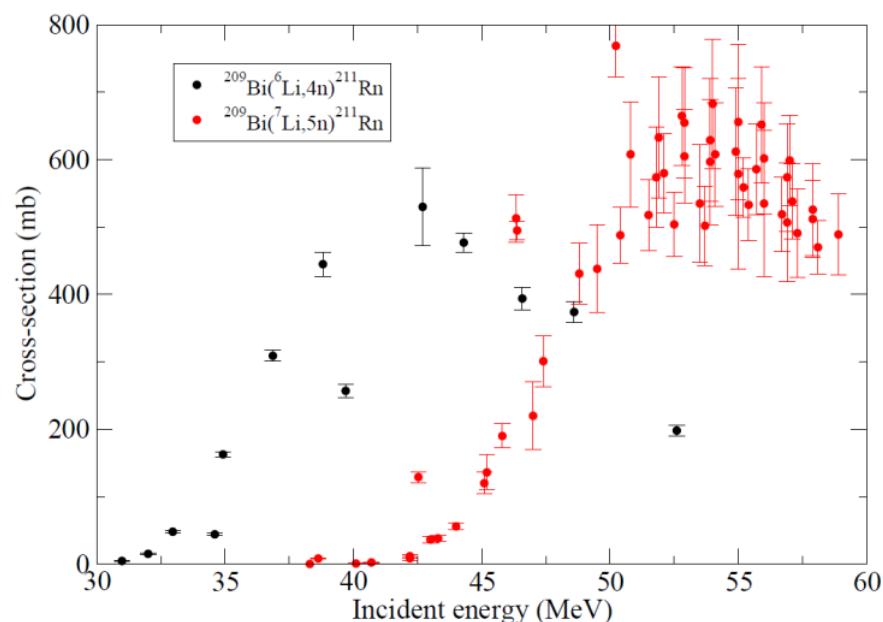
□ Production estimate:

□ LISE code

□ Lithium beam stopped in target

□ Optimal irradiation time : 14h

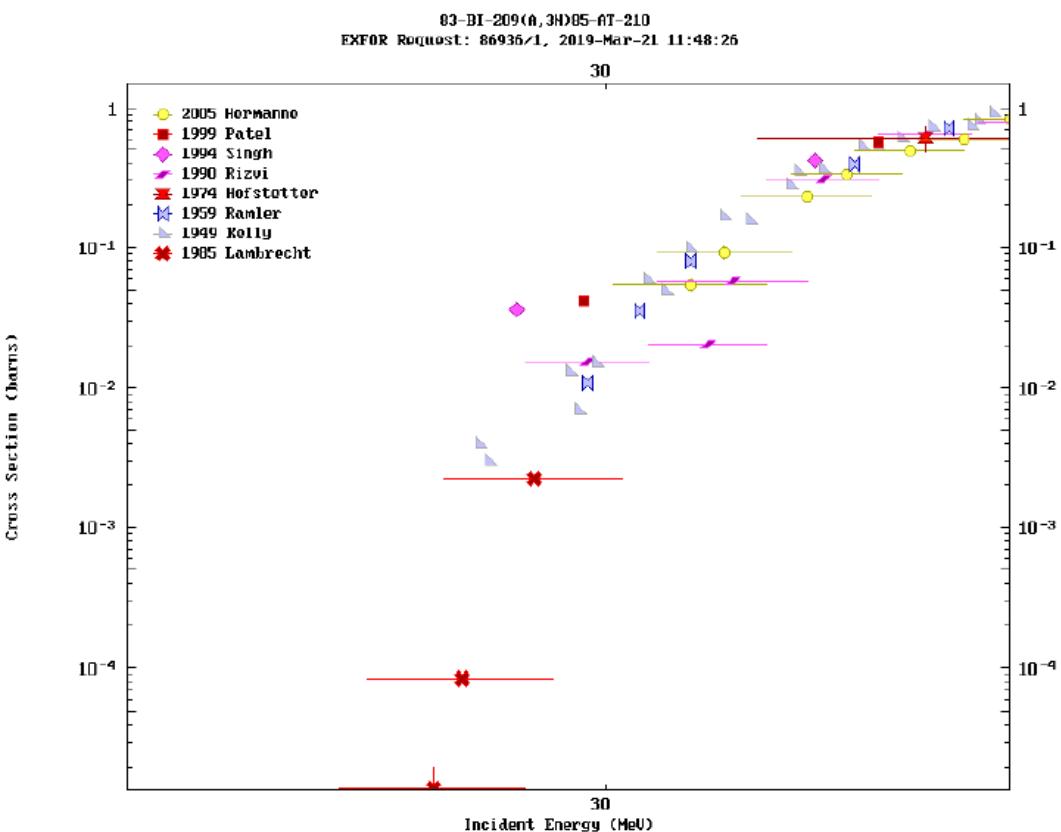
□ Best reaction $^{209}\text{Bi}(^7\text{Li},5\text{n})^{211}\text{Rn}$



48 MeV	36 MeV	Rapport activité
Act-utile (Bq/ μA)	Act-utile (Bq/ μA)	
^7Li	^6Li	$^7\text{Li}/^6\text{Li}$
4,84E+05	1,83E+05	2,65

^{211}At at NFS: contaminants

- ^{210}At :
 - $^{210}\text{At} \rightarrow ^{210}\text{Po}$
 - One single cross-section measurements in the alpha induced reaction
 - Nothing beyond 48 MeV with Li beam (max cross section at 55 MeV for ^7Li)
- $^{209,210}\text{Po}$:
 - Direct production in (α, t) or (α, dn) or $(\alpha, 2np)$
 - Incomplete fusion using ^7Li (t transfer followed by n evaporation) (10% of the yield involves α emission in $\text{Yb} + ^7\text{Li}$)
- $\alpha + \text{Pb}$:
 - PbBi eutectic liquid target. No EXFOR data



→ Cross section measurements at NFS

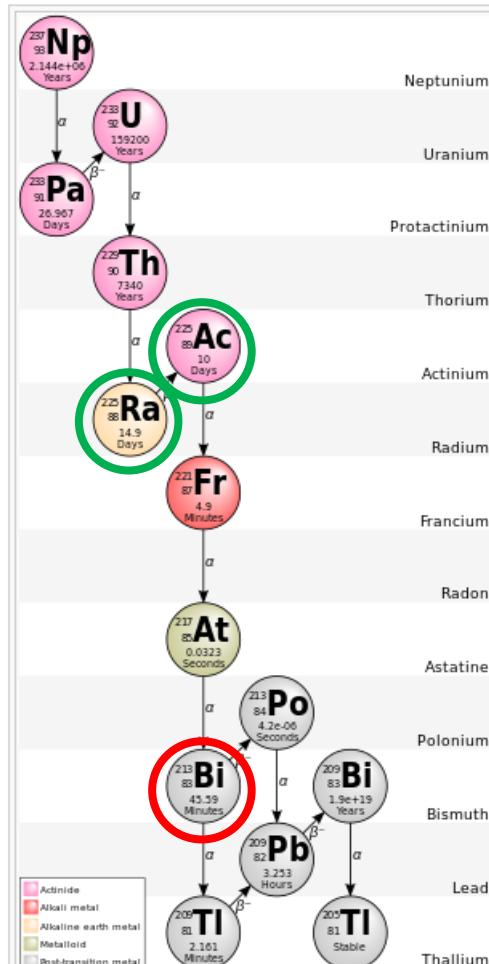
Agenda

- Motivations and opportunities

- Current projects
 - At-211
 - Other α emitters
 - Dosimetry
 - Liquid target

Other α emitters

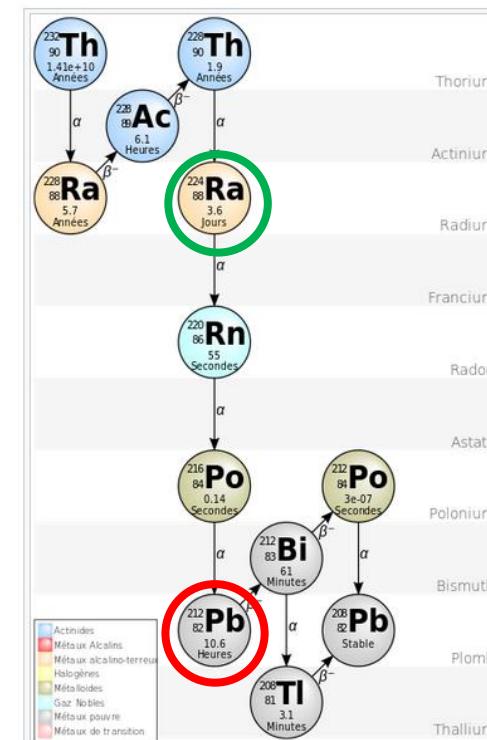
Bi-213 from Ra-225 or l'Ac-225



Radio-isotopes
of interest

Neptunium 237 decay chain

Pb-212 from Ra-224



Thorium 232 decay chain

Generators from
the $\alpha + \text{Th-232}$
reaction

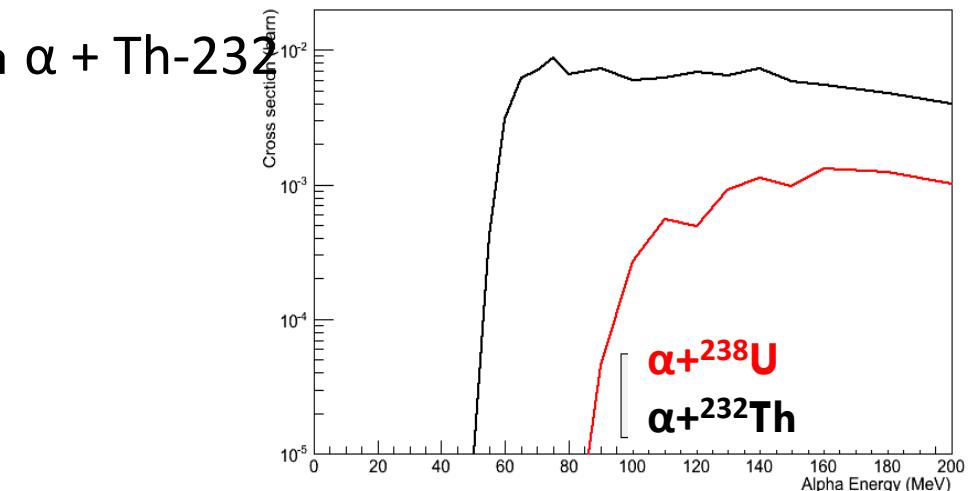
- Database TENDL-2014: Reaction $\alpha + \text{Th-232}$

- Reaction threshold

 - ≈ 50 MeV for $\alpha + \text{Th-232}$
 - ≈ 100 MeV for $\alpha + \text{U-238}$

- Production rate calculations :

 - Natural thorium target 0.05 cm so that $E_{\text{out}} \approx 50$ MeV
 - FISPACT-II vs PHITS : huge differences
 - $I=200\mu\text{A}$ ($6.2\text{e}14$ α/s)
 - $T_{\text{irr}}=1\text{d}$

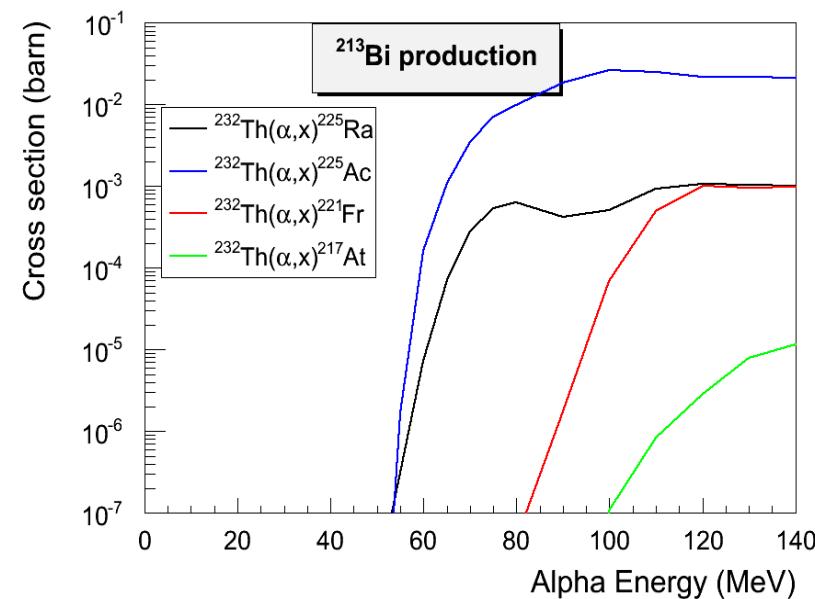


	MCNPX+FISPACT II	PHITS
^{224}Ra	$3.56 \times 10^8 \text{ Bq/g}$	$5.34 \times 10^6 \text{ Bq/g}$

⇒ First cross-section measurements to constrain models

⇒ Approved experiment

- Database TENDL-2014: Reaction α + Th-232
- Reaction threshold
 - ≈ 50 MeV for Ac-225 and Ra-225
 - ≈ 100 MeV for Bi-213
- FISPACT-II calculations:
 - Natural thorium target 0.05 cm so that $E_{\text{out}} \approx 50$ MeV
 - $I=200\mu\text{A}$ ($6.2\text{e}14$ α/s)
 - $T_{\text{irr}}=1$ and 10 d



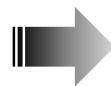
	80 MeV	
	A(Bq/g)	% Atot
Irradiation 1 i	1.24E+08	0.39%
Irradiation 10 i	9.30E+08	0.495%

⇒ First cross-section measurements to constrain models

- Motivations and opportunities

- Current projects
 - At-211
 - Other α emitters
 - **Dosimetry**
 - Liquid target

Development of a new
radiopharmaceutical



Studies /Evaluations
clinic /**preclinic**

BIOLOGICAL observable **vs.** PHYSICAL dose

→ AM Frelin's talk

- Motivations and opportunities

- Current projects
 - At-211
 - Other α emitters
 - Dosimetry
 - Liquid target

Liquid target

□ Liquid Bi target

- Design an «as simple as possible» liquid Bi target; not a loop
- Cooled Bi container; confinement issues
- Online and continuous extraction of At or Rn; how to concile with the previous point?...
- Design study in the next years (JRA ERINS; collaboration Czech Rep, ARRONAX, SUBATECH, INFN, HIL; ANR?)

Conclusions (1)

- SPIRAL-2 beams offer opportunities for R&D on production of innovative radioisotopes
- GANIL concentrate on target irradiations → collaborations
- A state (with ARRONAX, SUBATECH, NPI Rez):
 - Measurements of $\alpha +^{209}\text{Bi}$ et $+^{6,7}\text{Li}+^{209}\text{Bi}$ at NFS; impurities
 - Irradiation station for Bi under construction
 - Design study for high power (10 kW; solid Bi)
 - Long-term: liquid bismuth target with continuous online extraction of At-211 or Rn-211
 - Safety, transport, autorisation, manpower,...

Conclusions (2)

- Other alpha emitters : Bi-213, Pb-212:
 - Production study using $\alpha + {}^{232}\text{Th}$. Approved experiment.
 - Cross-section measurements \leftrightarrow nuclear data interest
- Development of dosimetry studies to evaluate new radioisotopes:
 - Focus on alpha dosimetry (detection methods; in-vitro distribution; effects;...). Very little done
 - Connection physics-radiobiology; interdisciplinary studies; CYCERON

Conclusions (3)

- Implementation:
 - Various possibilities evaluated (from optimal to simple)
 - Requires development (chopper) and safety aspects
- Extremely fruitful Czech Rep. – France collaboration
- **Many very interesting opportunities. New in-lab culture...**

THANK YOU