Study of Liquid Bismuth as an alternative target for At211 production

Theo BIGOURDAN (SUBATECH) on behalf of the REPARE collaboration
29/09/2022, INTDS 2022

Summary

- Context
  - REPARE project
  - Astatine 211 production
- Capsule targets
  - Concept
  - Computation and results
- Fluid loop
  - Concept
  - Computation and results
- Windowless fluid loop
  - Concept
  - Risk assessment
- Conclusion

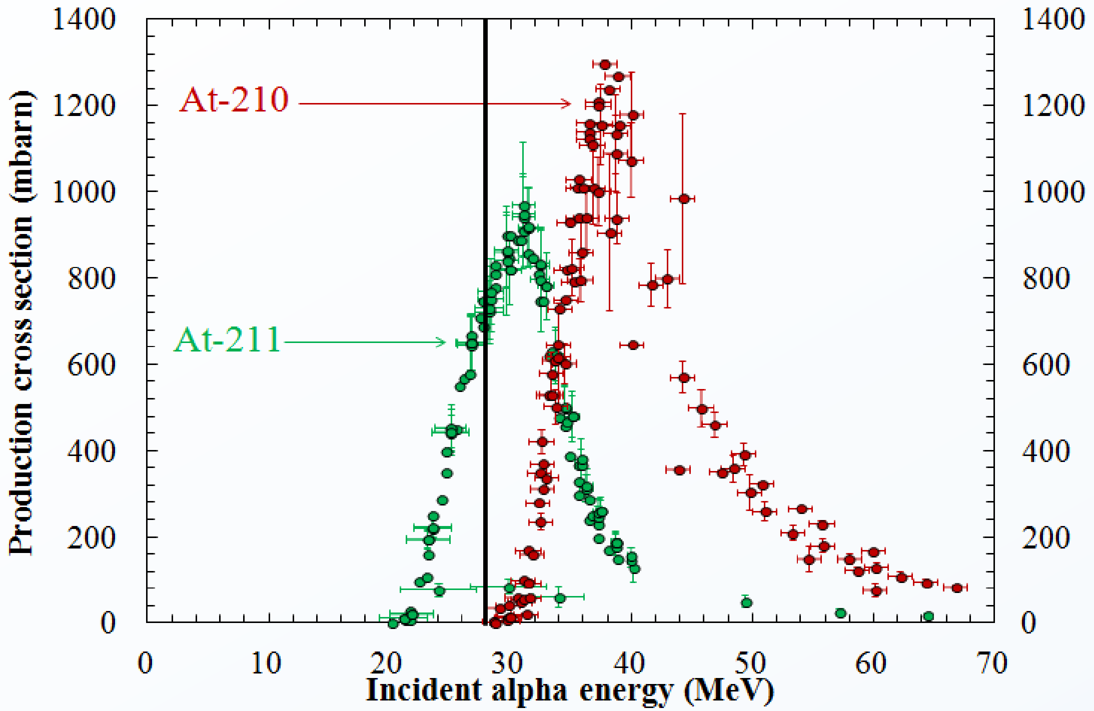
REPARE Project

- Research and dEvelopements for the Production of innovAtive RadioElements
  - Astatine 211 ($T_{1/2}$ = 7.2h): promising α-emitter for **Targeted α Therapy**
  - WP1: Inventory calculations and cross section measurements
  - WP2: High power solid target (*see our other presentation in this conference*)
  - **WP3: High power liquid target**
  - WP4: Rn211 generator

- **Our objective are:**
  - To study ways to increase At211 production through the ^{209}**Bi(α,2n)** reaction
  - To take advantage of the characteristics of GANIL's **SPIRAL 2** beam (up to 80MeV and **3mAe of α**)
  - To take advantage of **liquid bismuth properties** to design a liquid target

Beam properties for At211 production

- Nuclear reaction : ^{209}**Bi + α → ^{211}At + 2n**
- Energy :
  - In the bismuth, energy needs to be between **28.6MeV and 20.7MeV** to produce At211
  - **Strictly <28.6MeV** to avoid production of **At210** (which decreases in **Po210**)
- Intensity :
  - Value to **maximize** to increase production
  - **Limited** by heat generation effects on the **mechanical resistance** in the structure
  - Gaussian distribution as a function of the radial coordinate, cut to 2σ (which correspond to 90% of total power)



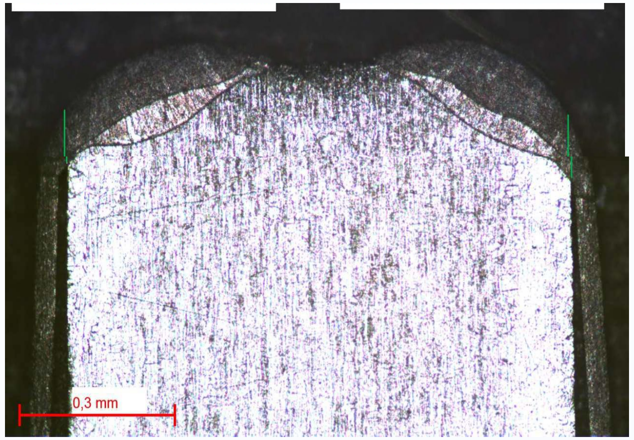
Production cross section for At211 and At210 as a function of energy

CAPSULE CONCEPT

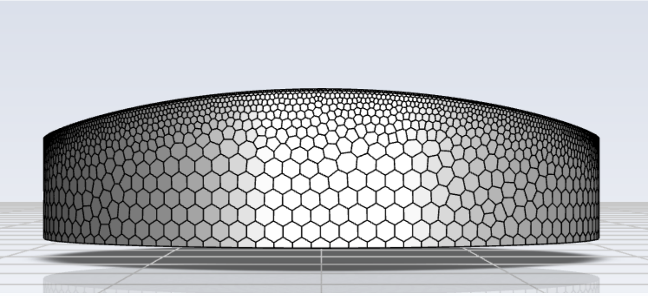
Capsule target

- Concept
  - Metallic capsule containing bismuth
  - Cooled on the back plate
  - Bismuth melts under the beam heat generation
- Opportunity:
  - Curved window to improve stress distribution

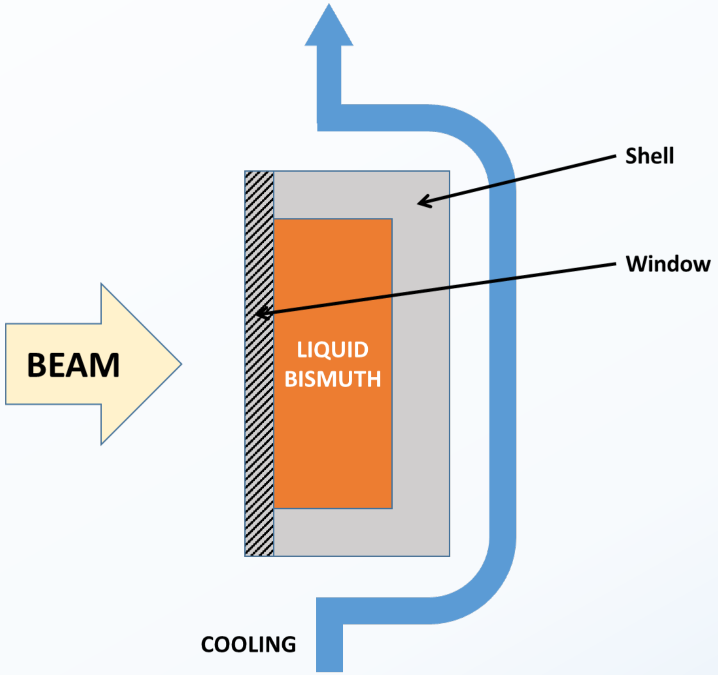
Existing capsule



Capsule cross-section



Capsule with curved window side view - CFD Mesh

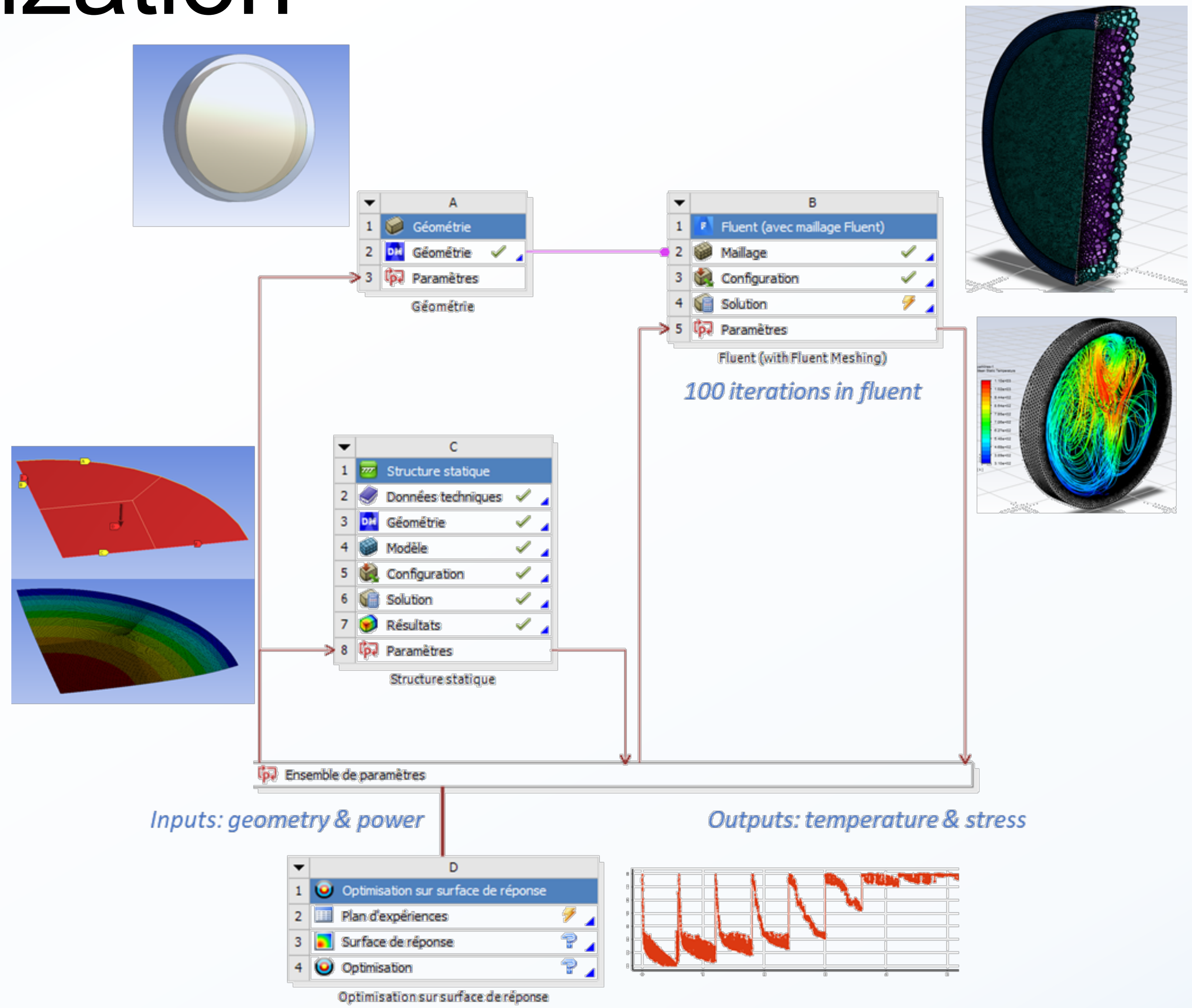


Window optimization

Parameter ranges

Target safety factor	1.1 +/- 0.1
Radius range	10 to 20mm
Window thickness range	100 to 700µm
Curvature radius range*	50 to 80mm

**For cylindrical window solution only.*



=> Maximal efficient power/production with safety factor>1

Capsule target – optimization results

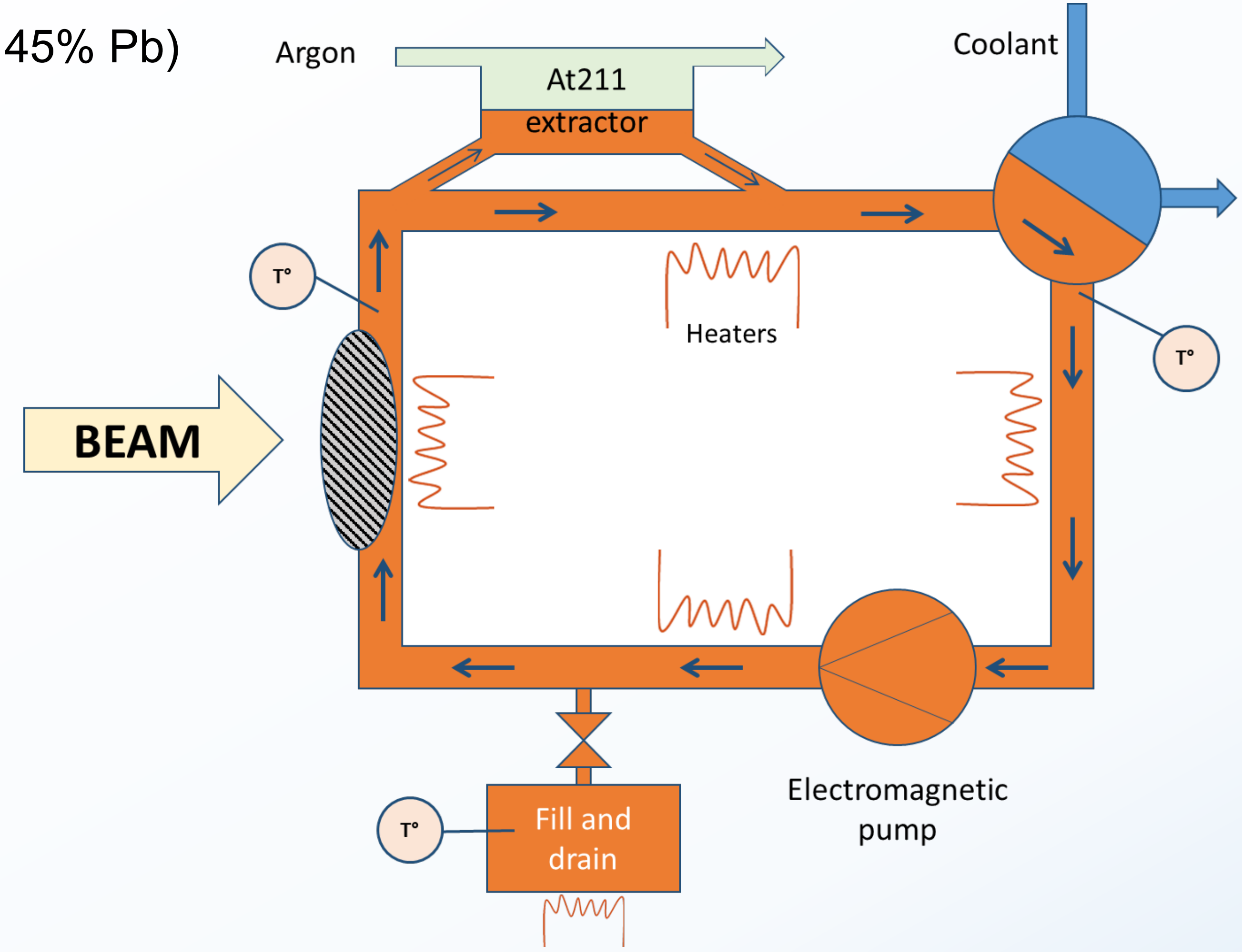
Case	Radius	Window thickness	Power	Temperature	Safety factor	Energy	Intensity	Production
	mm	µm	W	K	-	MeV	**µAe**	**GBq-1h**
Niobium (flat)	15	700	2450	1269	1.05	76.7	**63.9**	**1.37**
Havar (flat)	20	200	2500	1145	1.09	47.5	**105.3**	**2.26**
Niobium (curved)	20	100	4350	1322	1.06	37.7	**230.9**	**4.95**
AISI 316 (curved)	20	100	2030	981	1.03	38.5	**105.5**	**2.26**
Arronax target	*Reference (in service target at the ARRONAX cyclotron)*					28.6	**20**	**0.43**

FLUID LOOP CONCEPT

LBE circuit concept

- Components:
  - Flowing Lead Bismuth Eutectic (55% Bi + 45% Pb)
    - **Lower fusion T°** (123°C)
    - Technically more **mature** than pure Bi
  - Temperature control
    - Heating resistances
    - Thermal exchanger
    - Window cooled by the LBE flow
  - At211 in line extraction (option)
    - ~600K for evaporation
  - EM pump for the flow

- LBE then acts as:
  - **Target material**
  - **Coolant** (for itself and for the window)
  - **At211 carrier**

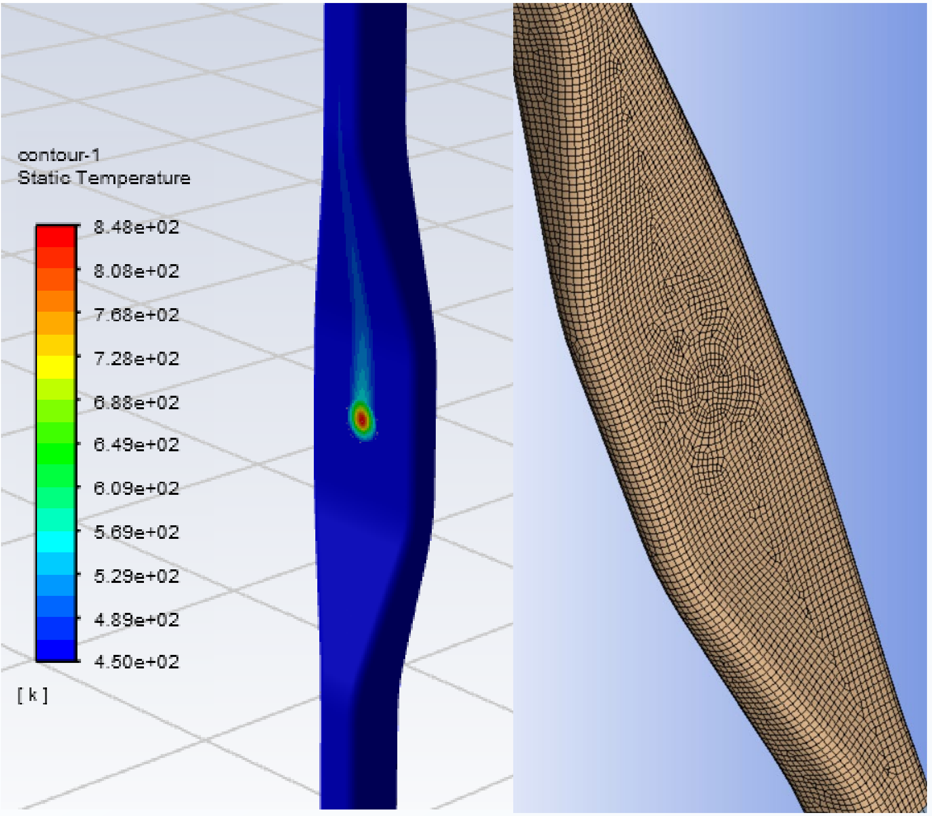


Window sizing

- **Limitations**:
  - Reduced production by 45% compared to pure Bi
  - Sufficient LBE **velocity** to cool the window area => higher pump head pressure
  - Window **thick** enough to withstand pump **pressure**

$\Rightarrow$ **Production: 0.21 GBq/1h**

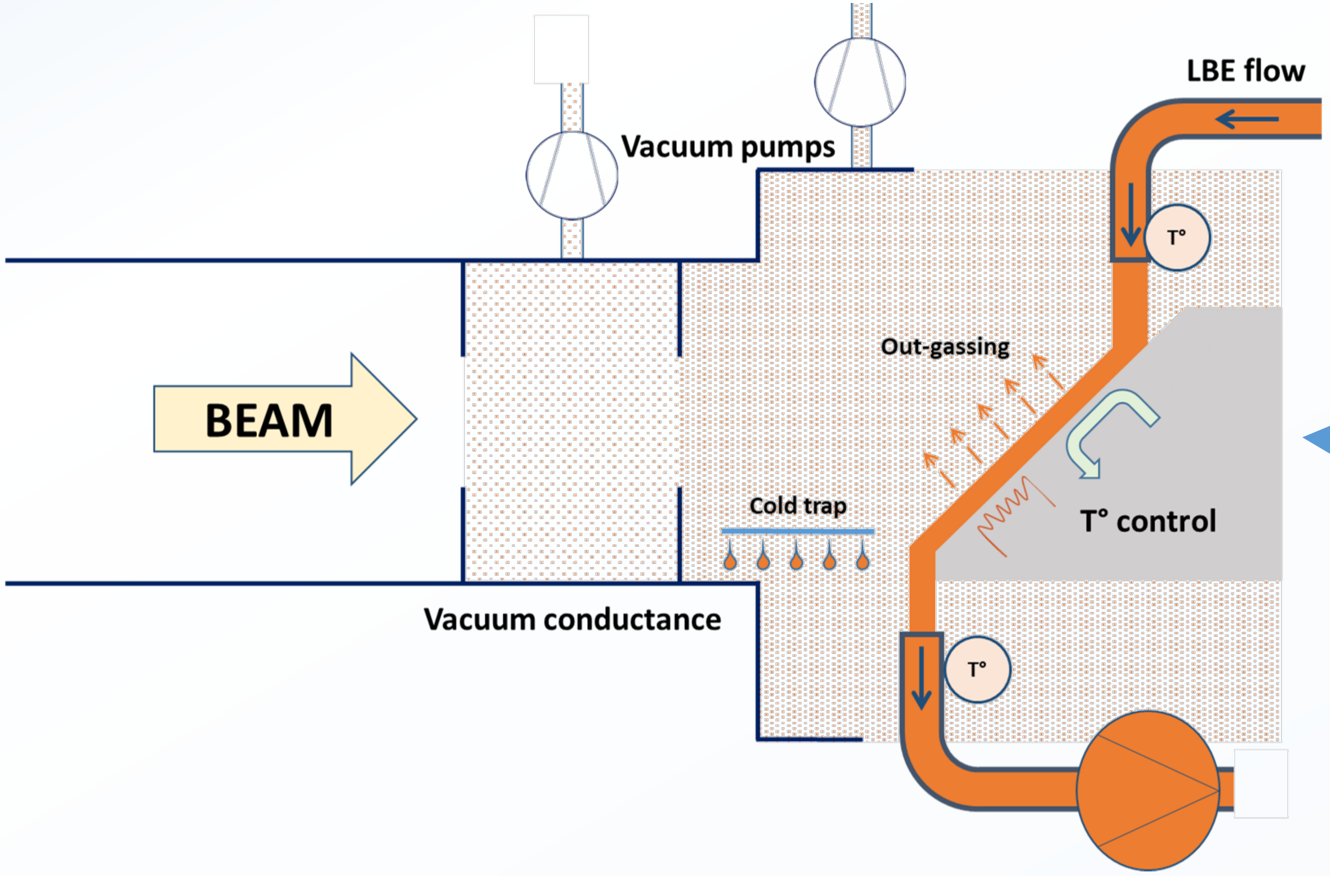
Window Diameter (mm)	13
Flow rate (kg/s)	2.5
Window thickness (µm)	500
Power (W)	593.5
Energy (MeV)	65.7
Intensity (µAe)	18.1
Production (GBq - 1h)	0.21

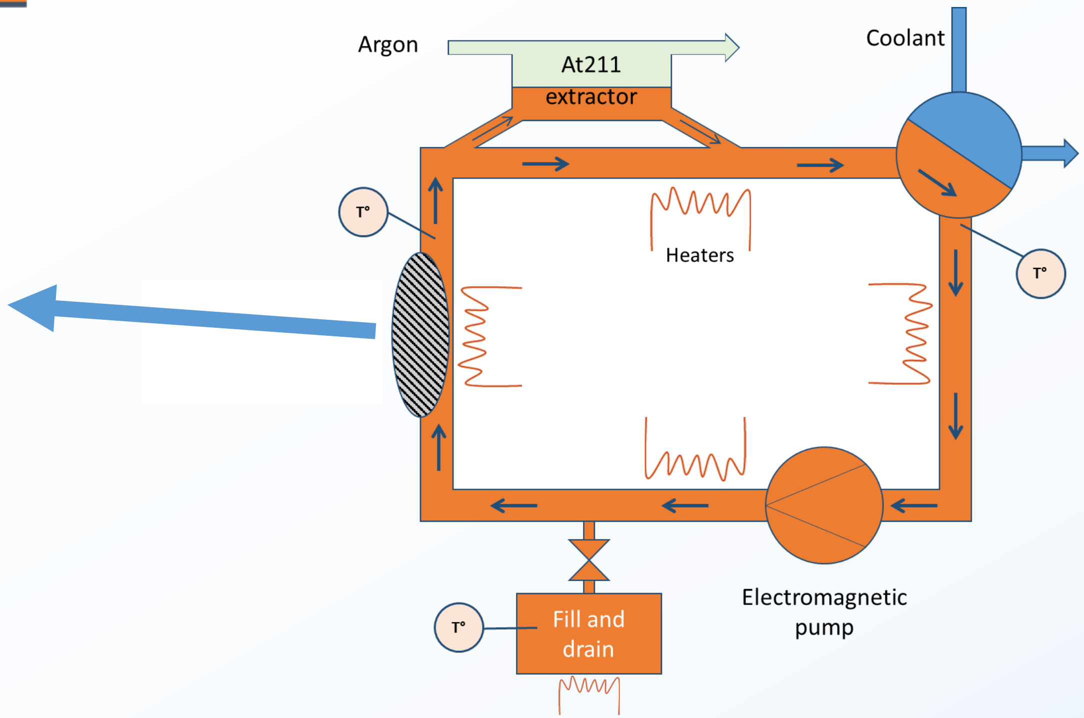


CFD computation overview

WINDOWLESS FLUID LOOP CONCEPT

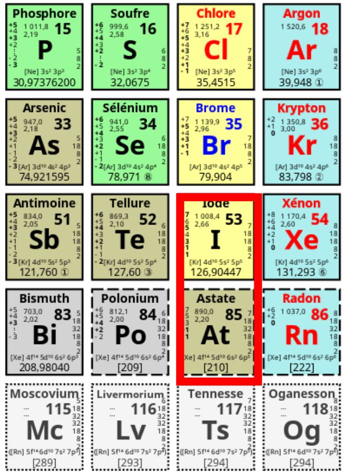
Windowless LBE circuit





Risk assessment

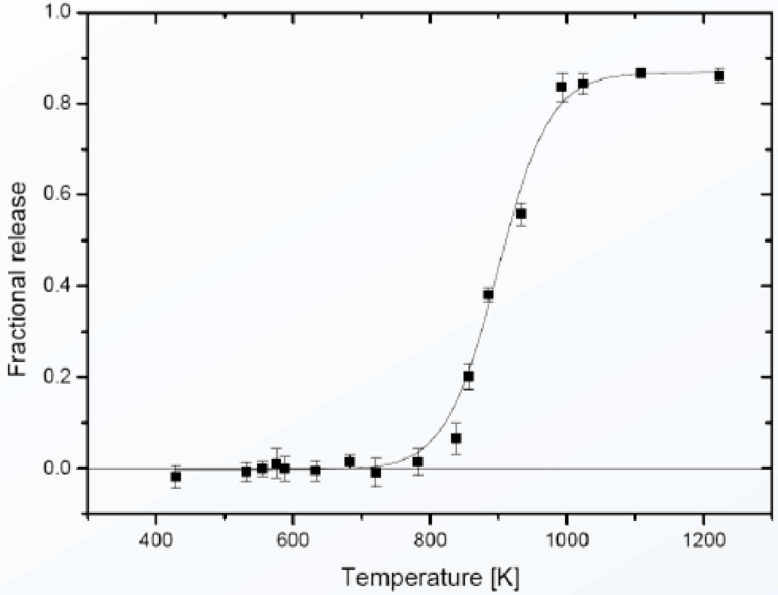
- **Beam line contamination** (LBE evaporation/ebullition)
- **At211 losses through evaporation** in the beam line
- **At211 losses** through fixation to **metallic structures**



	Estimated total activity [Bq]	Predicted activity[a] [Bq]	% of predicted amount	% of estimated total activity
Bulk	295 ± 18	8560	3.4 ± 0.2	7
LBE/cover gas interface	1.2 ± 0.1		$(14 \pm 1) \times 10^{-3}$	0.03
LBE/steel interface	$(37 \pm 20) \times 10^{2}$		43 ± 23	93
Absorber	$(38 \pm 2) \times 10^{-2}$		$(44 \pm 2) \times 10^{-4}$	0.01
Sum	3997 ± 2018		47 ± 24	

[a] Average of two calculations using different nuclear models.

Summary of 129I activity distribution over the different types of samples.
(B. Hammer-Rotzler et al., Radiochemical determination of 129I and 36Cl in MEGAPIE)



Evaporation of iodine from very dilute solution in LBE
(Handbook on LBE Alloy and Lead Properties, Materials Compatibility, Thermalhydraulics and Technologies)

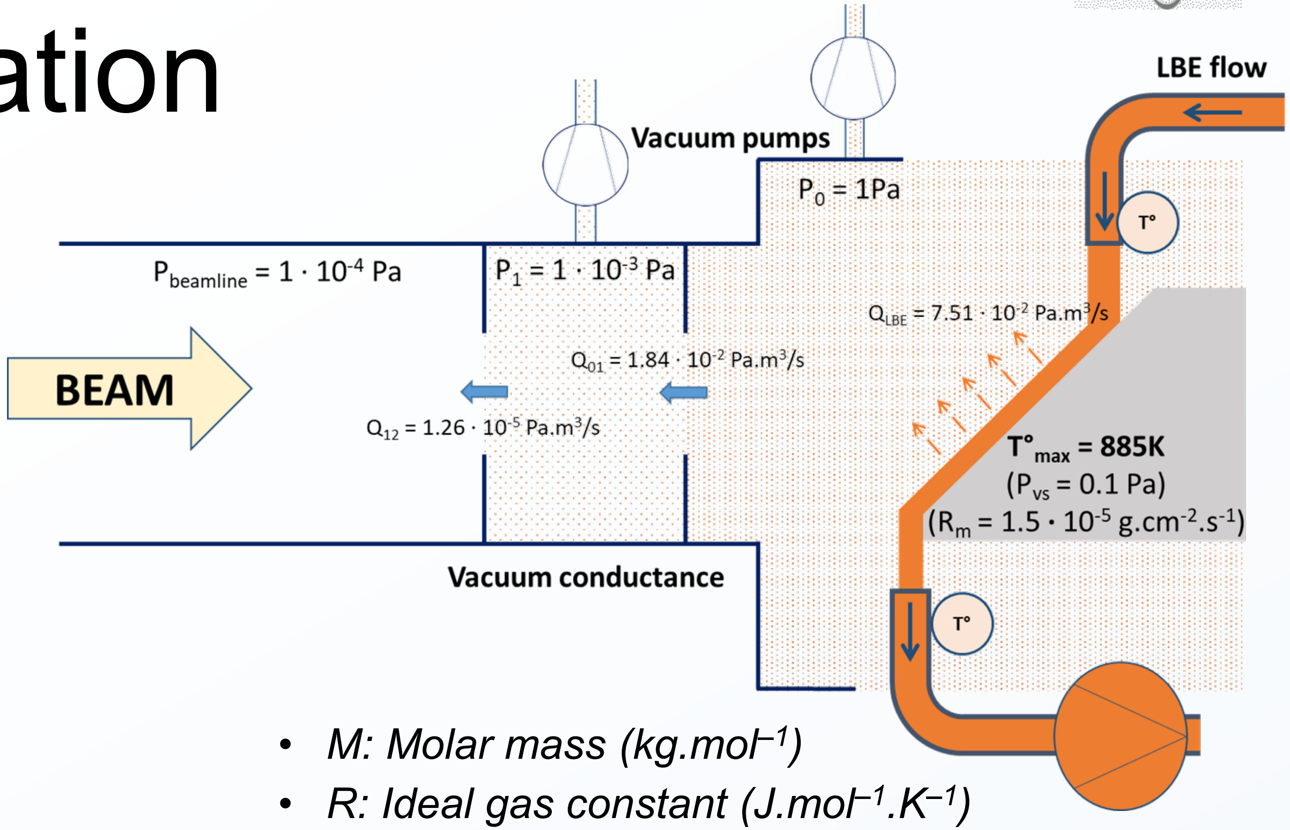
Beam line contamination

- Evaporation rate
  - $R_m = \sqrt{\frac{M}{2\pi RT}} P_s$
  - *Rm (885K) = 1.5 · 10^{-5} g.cm^{-2}.s^{-1}**

⇒ **Capability to maintain low pressure**

- Vapor pressure
  - $P_{s(LBE)}[Pa] = 1.22 * 10^{10} * \exp\left(-\frac{22552}{T[K]}\right)$
  - **$T_{boiling}$ (0.1 Pa) = 885K**

⇒ **Maximal production: 11.5 GBq / 1h**
(14kW, 979µAe)



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- **Beam line contamination** (LBE evaporation/ebullition)
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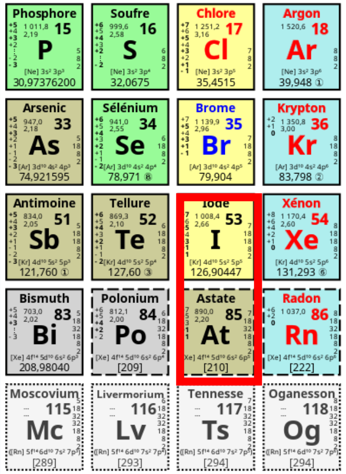

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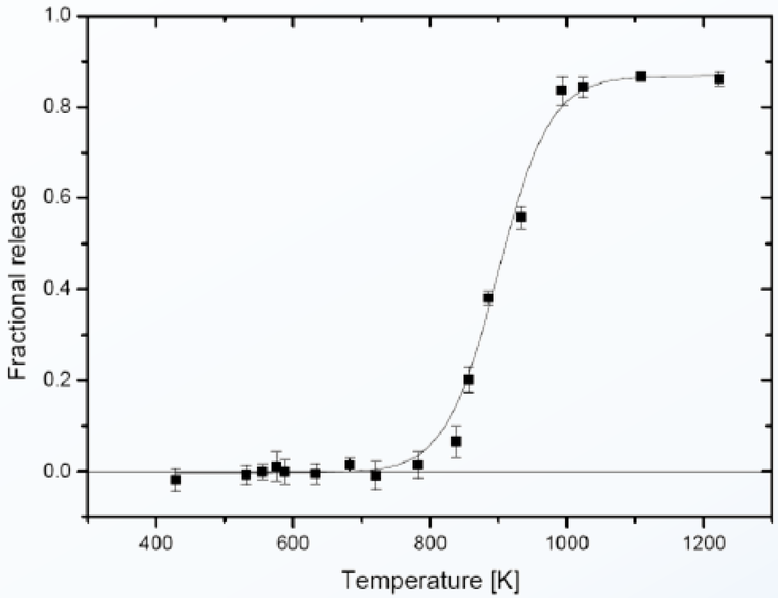

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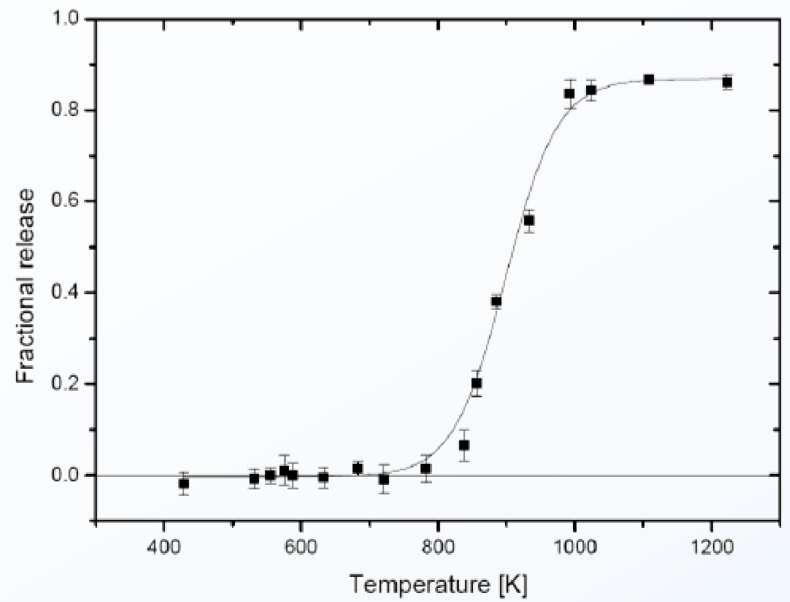

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  - Mitigation : lower tolerable T°
- **At211 losses** through fixation to **metallic structures**
  - Experts are **pessimistic**
  - Iodine bibliography is **pessimistic**
  - **Mitigation: ?**

⇒ **Experiment planned to investigate this last issue**

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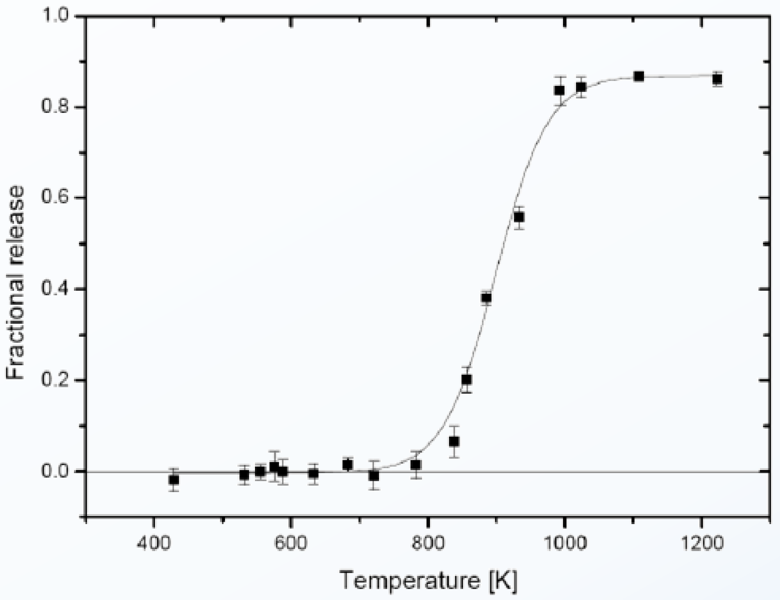

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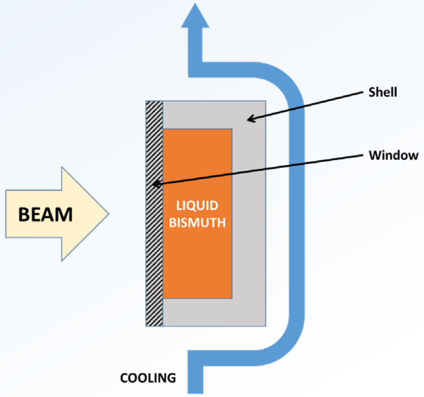

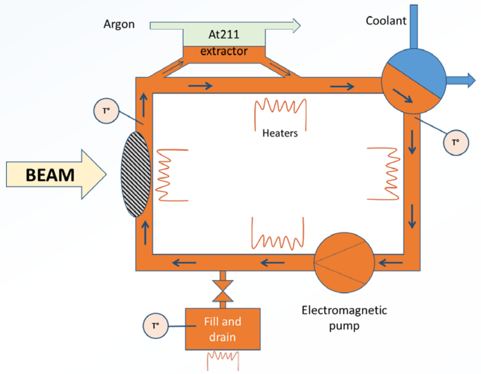

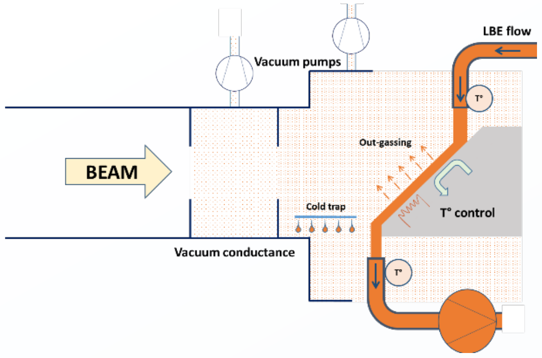

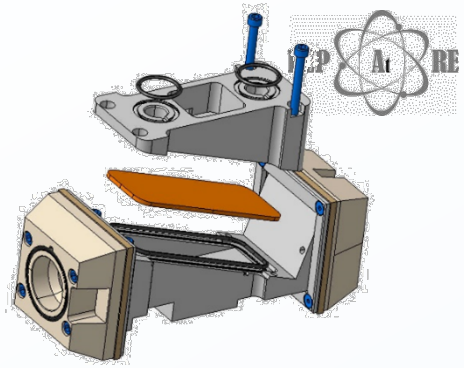

Criteria	Bismuth Capsule	LBE loop	Windowless LBE loop	ARRONAX
Production	** 4.9 GBq - 1h	* 0.21 GBq - 1h	*** 11.5 GBq - 1h *(pending losses evaluation)*	* ~0.43 GBq – 1h TBC
Maturity	*** In service for other targets. Curved window not demonstrated.	** Feedback from MEGAPIE.	* Lack of experience on liquid LBE in the vacuum.	***** In service.
Exploitation	*** Manual extraction, easier transport.	** In line extraction as an option. Important volume of LBE.	* In line extraction. Important volume of LBE. Beam line losses.	*** Manual extraction, easier transport.
Cost	**** Simpler system.	** Pump, pipe, exchanger…	* Pump, pipe, exchanger, beam line modifications…	**** Simpler system.
Integration	**** Simpler system.	** Pump, pipe, exchanger…	* Pump, pipe, exchanger, beam line modifications…	**** Simpler system.

Conclusion and way forward

- **Physical limits** to At211 production through liquid bismuth target
  - **Windows** strongly limit the production rate: beam absorption and mechanical stress
  - **Window removal** compromises At211 retrieval
  - Bismuth **metallic loops** compromise At211 retrieval (?)
- **High power liquid target** dedicated installation?
  - Current concepts are showing physical limits
  - Only **30%** (loop) **to 2%** (flat capsule) of SPIRAL II's **3mAe** are used
  - Smaller local production units more adequate?
- Small scale **experiment** (capsule) to:
  - Study At211 migration risks
  - Crosscheck computation
  - Demonstrate capsule concept's feasibility

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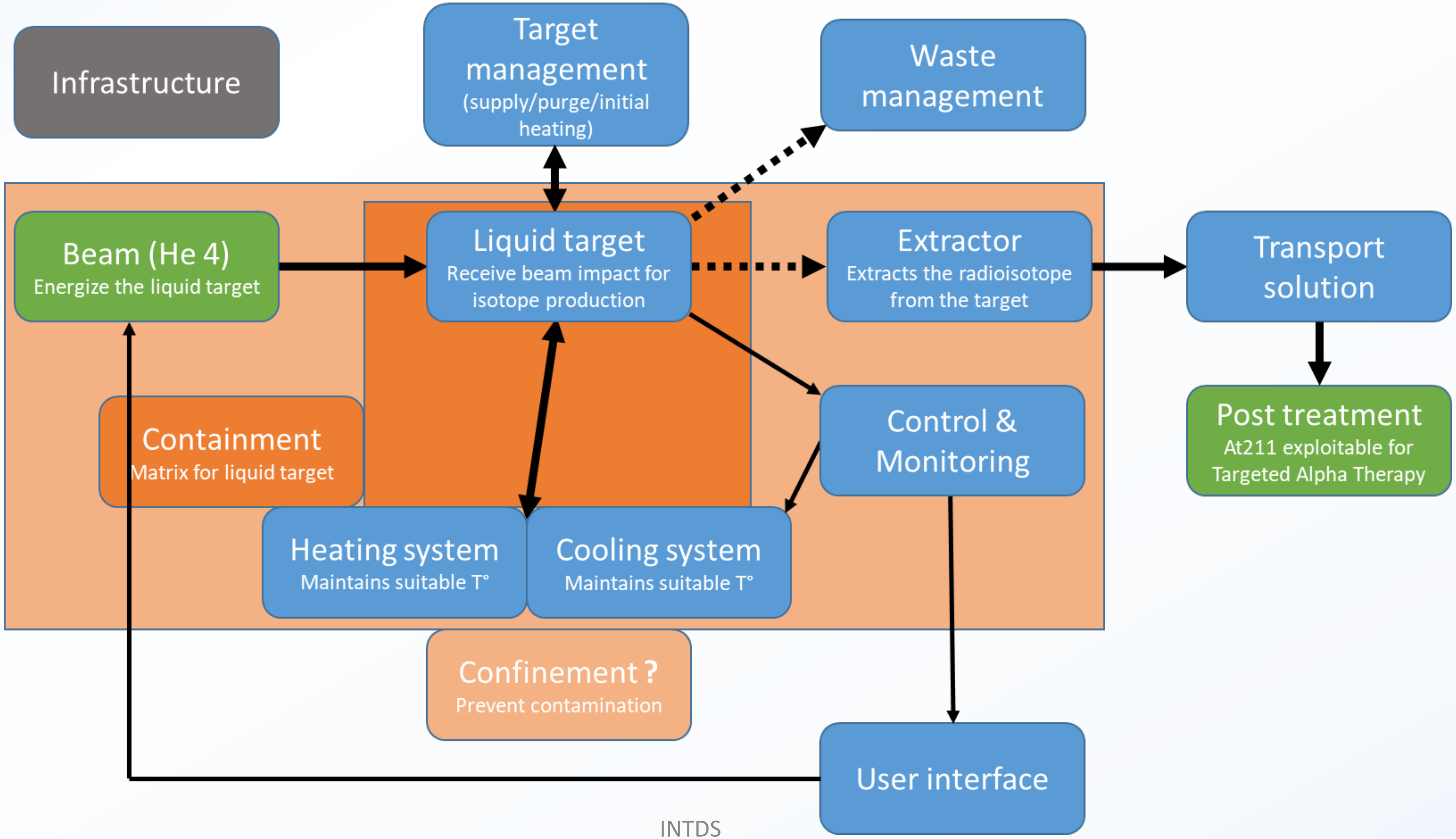
Production	**	*	***	*
Maturity	***	**	*	*****
Exploitation	***	**	*	***
Cost	****	**	*	****
Integration	****	**	*	****

Schematic view of the problematic



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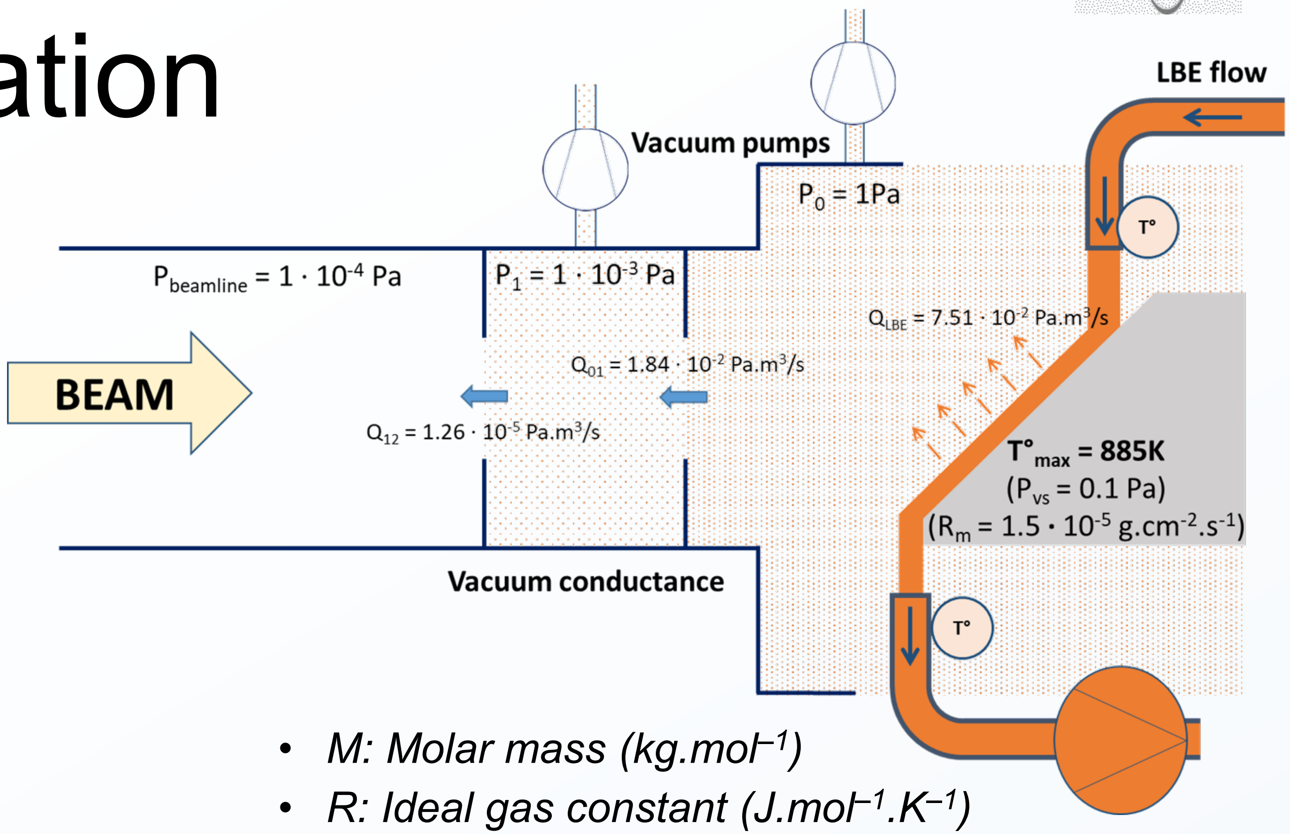

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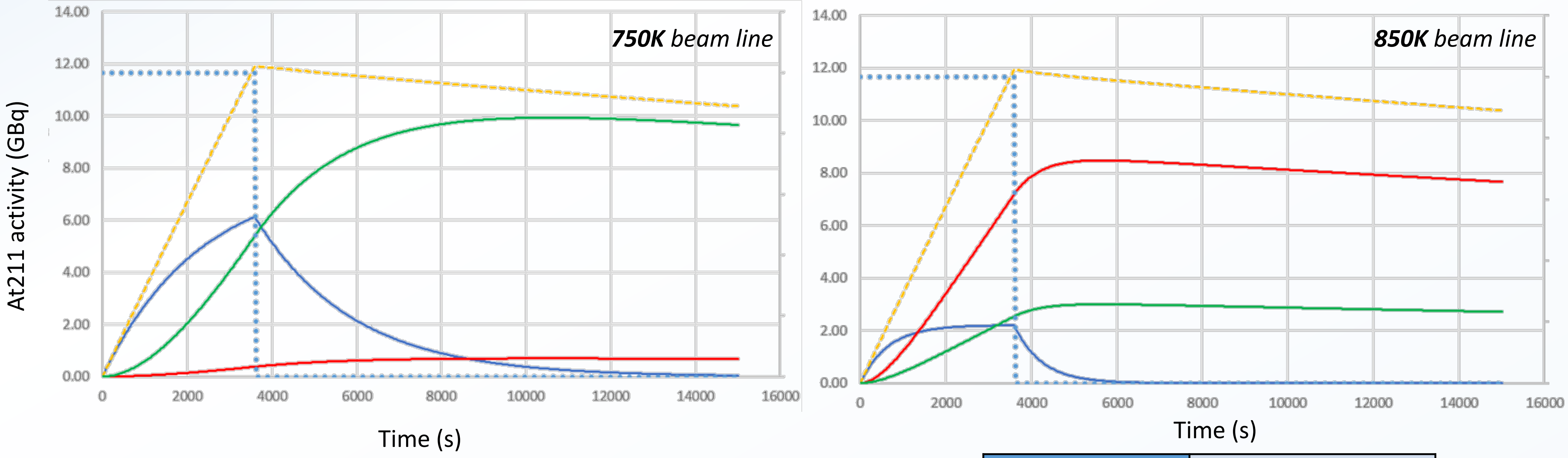
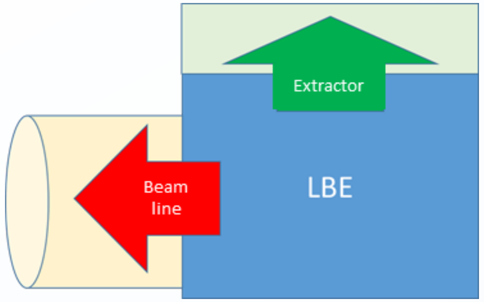
At211 losses through evaporation

Astatine production and distribution as a function of time





Irradiation time	3600	s
Intensity	979	µA
At211 Radioactive constant	1.16E-05	s-1
LBE mass	200	kg
Extractor T°	800	K
Beam line T°	750 ; 850	K
Extractor surface	1000	cm²
Beam line surface	500	cm²

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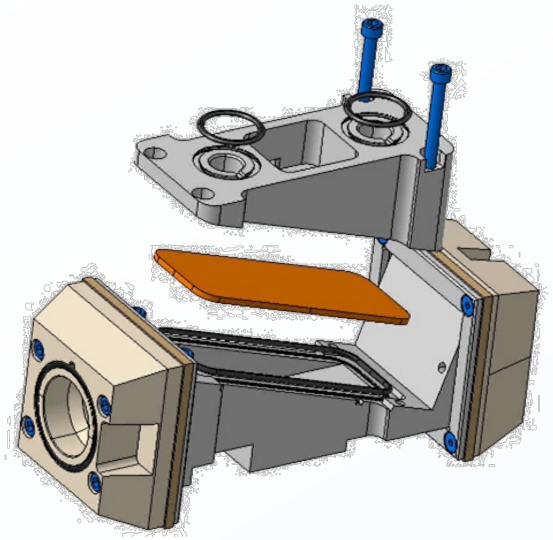
Production order of magnitude

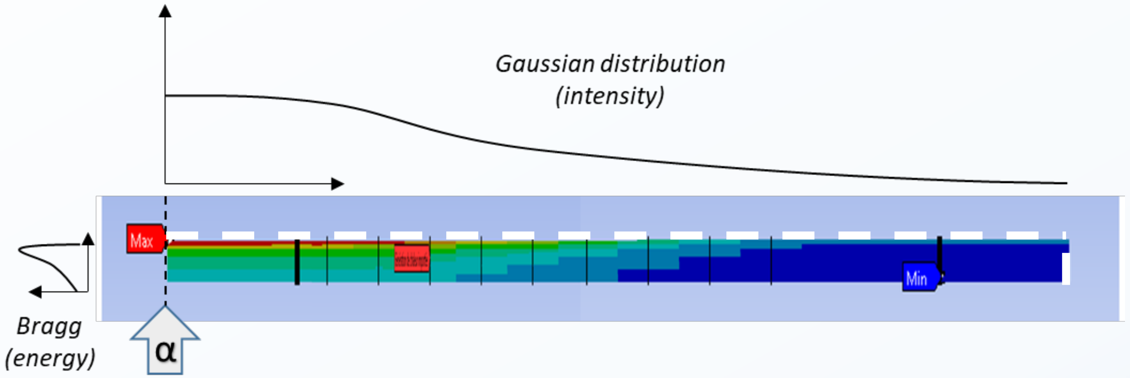
Two main references were considered to assess the efficiency of the targets we considered.

- Current production assessment at **ARRONAX**
  - **0.43 GBq/1h**
  - Intensity limited to 20µAe by the energy degrader used to go from 68MeV to 28.6MeV
- Computations
  - Taking optimistic hypotheses, regardless of technical feasibility
  - **Ideal solid** target production: **4.4 GBq/1h**
    - T° allowable: 271°C (bismuth fusion)
  - **Ideal liquid** target production: **15.3 GBq/1h**
    - T° allowable: 600°C (engineering judgement)

⇒ Significant benefits could be achieved



ARRONAX bismuth target



Heat generation of the ideal target model cross-section
A convection boundary condition (25°C ; 25kW.m^{-1}.K^{-1}) is applied on the back plate, represented here by the dashed line.

Window optimization

- Window radius optimization:
  - ↗ Radius ⇨ ↗ stress
  - ↗ Radius ⇨ ↘ temperature ⇨ ↗ mechanical strength

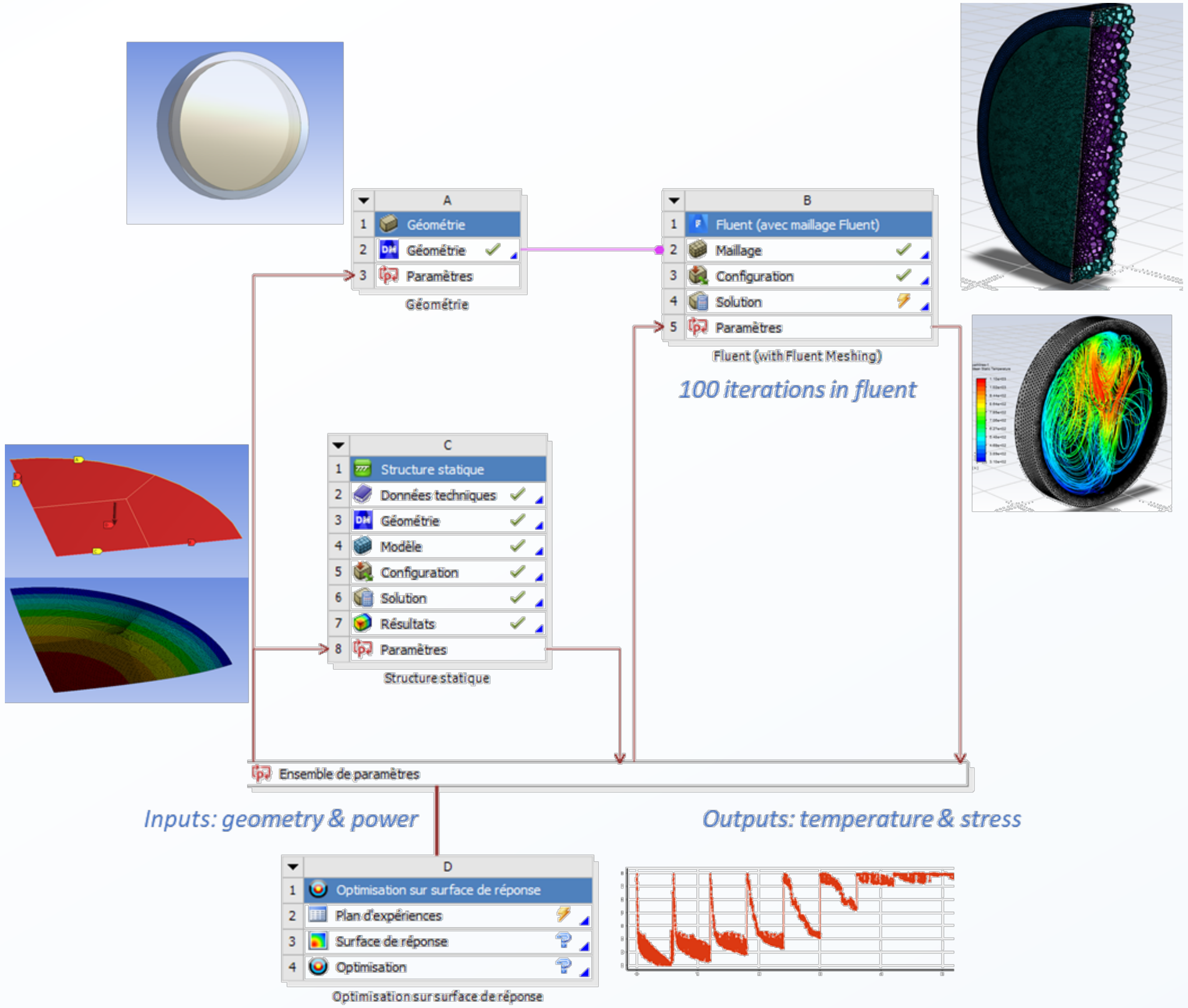
(considering a constant heat generation, the beam is distributed through a bigger area)

- Window thickness optimization:
  - ↗ thickness ⇨ ↘ stress
  - ↗ thickness ⇨ ↗ Temperature ⇨ ↘ mechanical strength

(to keep the same production, it is needed to increase initial energy to reach the bismuth at the adequate energy)

⇒ No obvious optimum

⇒ Run the optimization for various materials



Target safety factor	Power range	Radius range	Window thickness range	Curvature radius range*
1.1 +/- 0.1	1000 to 8000W	10 to 20mm	100 to 700µm	50 to 80mm

*For cylindrical window solution only.