Radionuclide Production

by High Intensity Proton Irradiation

at the INR Linear Accelerator

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Requirements for Large-Scale Radionuclide Production

- **Accelerators** providing necessary proton energy and high intensity beams *(many tens or hundreds μA)*

- **Powerful facilities** for target irradiation

- **Targets** with specific technology of fabrication

- **Chemistry** for recovery of “no-carrier-added” radionuclides from massive targets with a great number of different radionuclidic impurities

Accelerators and Facilities for Radionuclide Production at High Intensity Proton Beam of Intermediate Energy

Institute for Nuclear Research (Troitsk, Russia)	160 MeV, 120 μA
Los Alamos National Laboratory (NM, USA)	100 MeV, 200 μA
Brookhaven National Laboratory (NY, USA)	200 MeV, 90 μA
TRIUMF (Vancouver, Canada)	110 MeV, 500 MeV, 50 μA
iThemba Laboratory (Cape Town, South Africa)	66 MeV, 250 μA
ARRONAX GIP (Nantes, France)	70 MeV, 2 x 100 μA

INR Linear Proton Accelerator

High-current proton linac

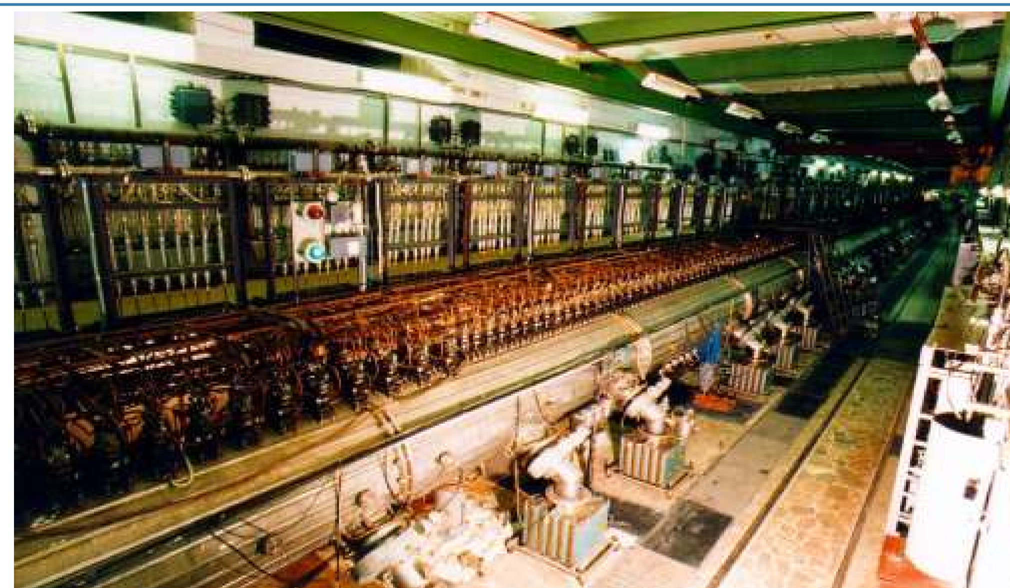
- located in Troitsk (Moscow)

- designed for
H+ and H-, up to 600 MeV,
beam current up to 0.5 mA

Multiple-Discipline Collaboration

- providing a wide range of fundamental and applied research

- including a radionuclide production facility

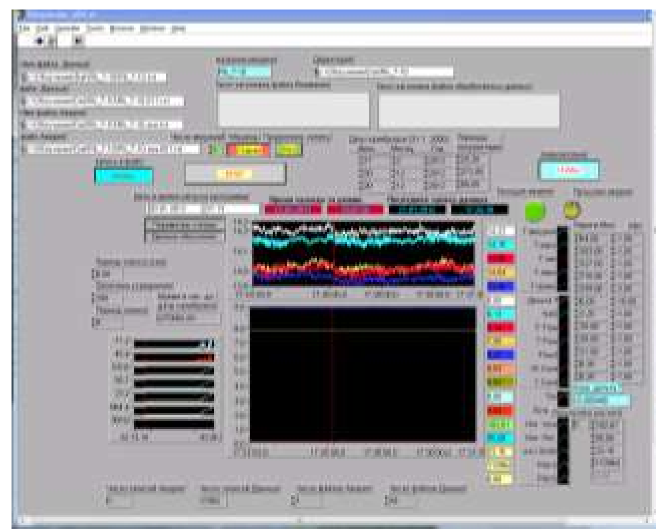


Beam Parameters at Radionuclide Production Facility

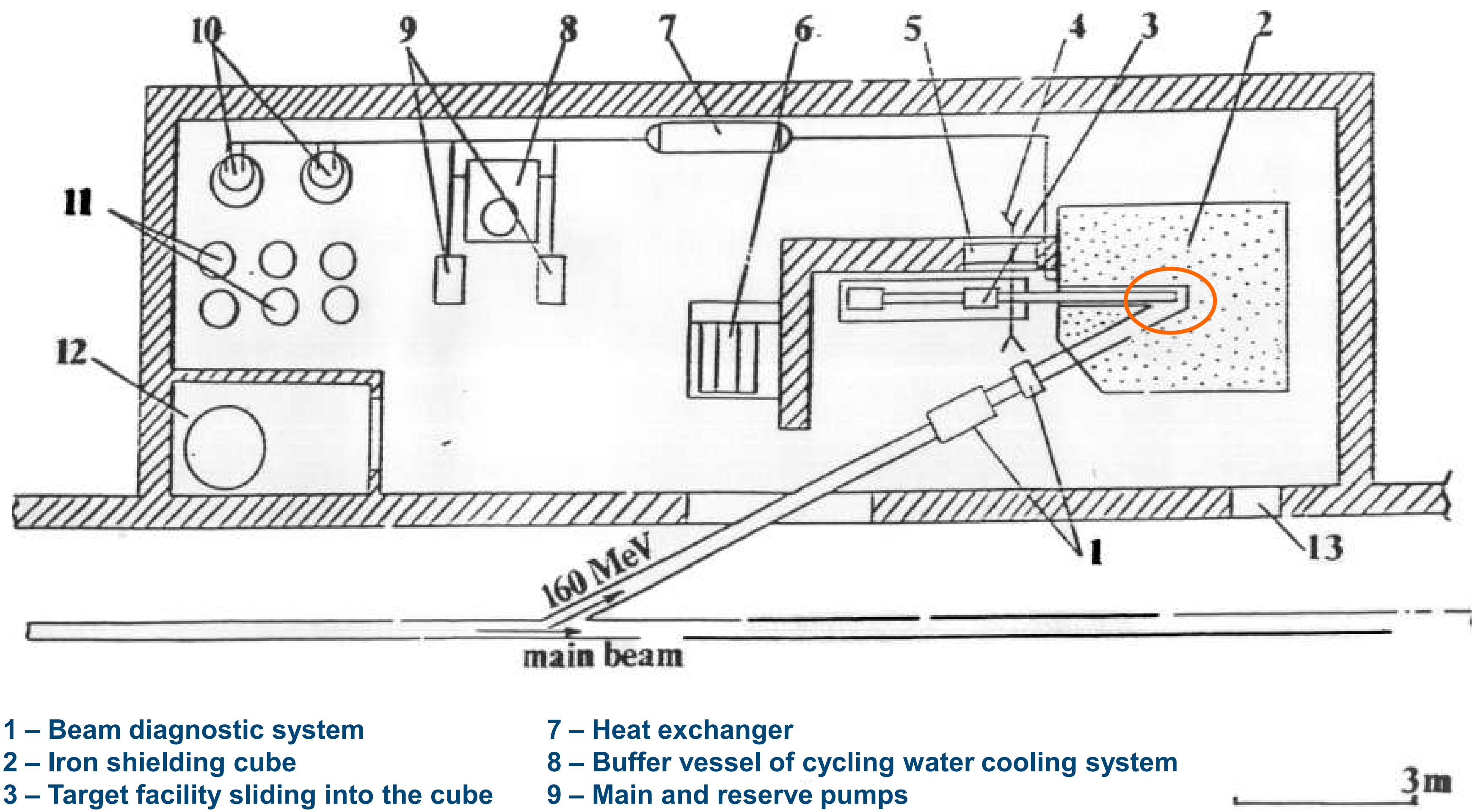
- Proton beam energy at the facility: 158 MeV
  - Options: 143, 127, 113, 100, 94 MeV
- Maximum beam current: 140 μA
- Pulsed beam:
  - Peak beam current 14 mA
  - 50 Hz
  - 200 μsec pulse width

INR Radionuclide Production Facility (*constructed in 1992*)



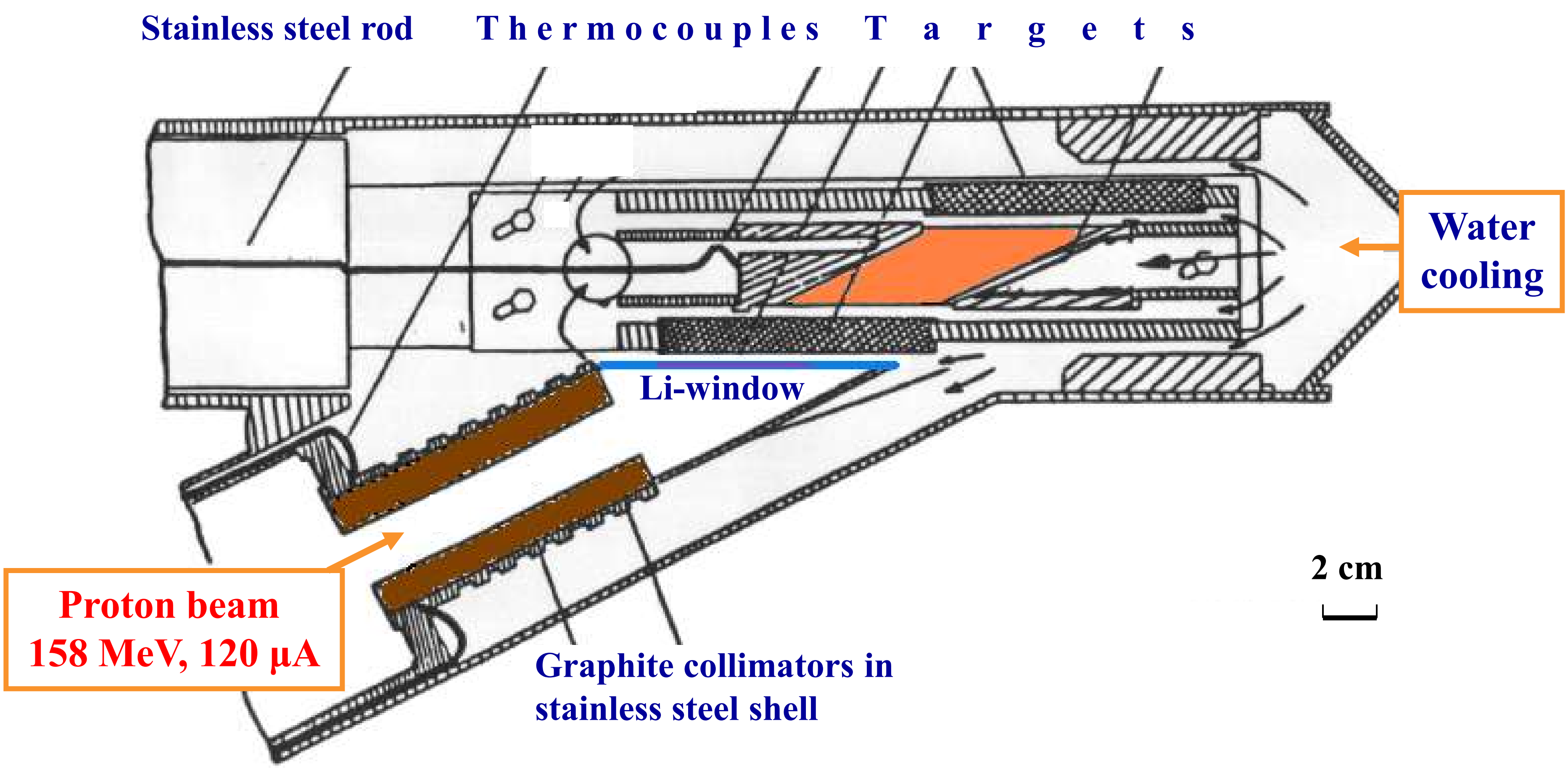



Floor Plan of INR Radionuclide Production Facility



1 – Beam diagnostic system
2 – Iron shielding cube
3 – Target facility sliding into the cube
4 – Manipulator for target handling
5 – Lead window system
6 – Main exit
7 – Heat exchanger
8 – Buffer vessel of cycling water cooling system
9 – Main and reserve pumps
10 – Ion exchange filters
11 – Storage of used radioactive filters
12 – Tambour with hatch for getting radioisotope products off
13 – Reserve exit

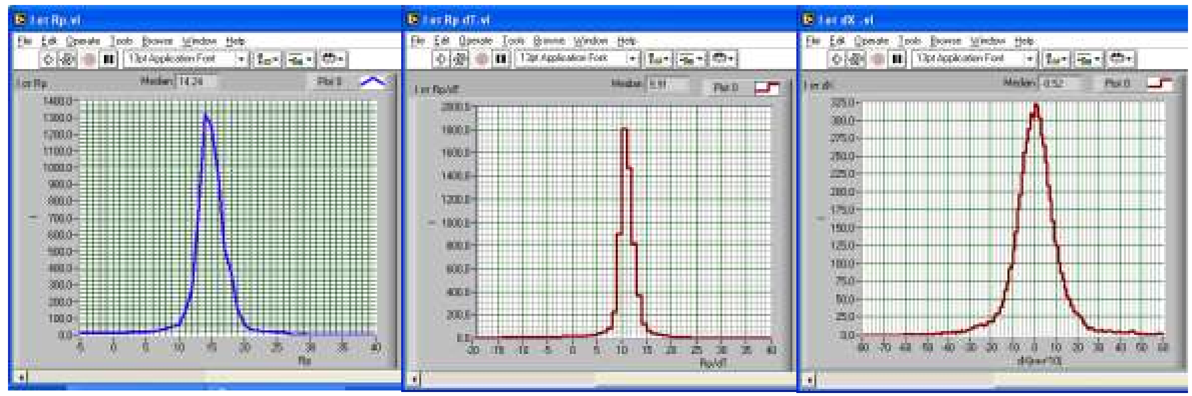
Target Holder at INR Facility



Monitoring High Intensive Irradiation Conditions

❑ Beam Parameters

- Beam current
- Beam shift (X and Y)
- Beam width
- Beam density (safety)
- Beam losses on the collimators



❑ Cooling Water Parameters

-Flow
- Pressure
- Temperature
- Electroconductivity
- Concentration of hydrogen released

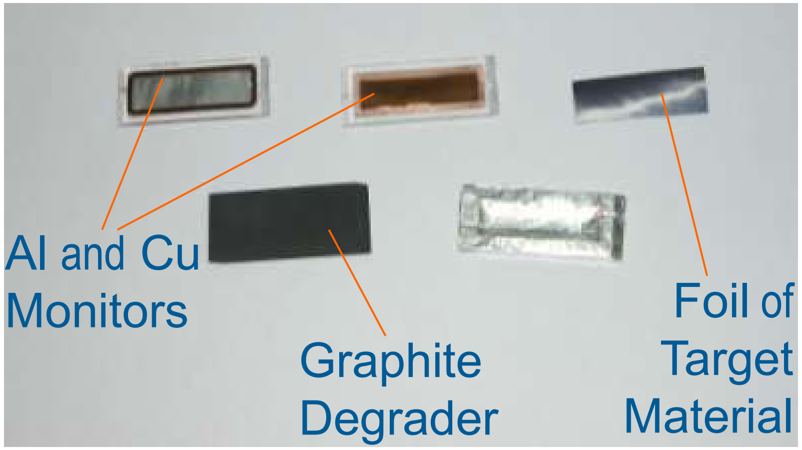
❑ Dose Rate Level

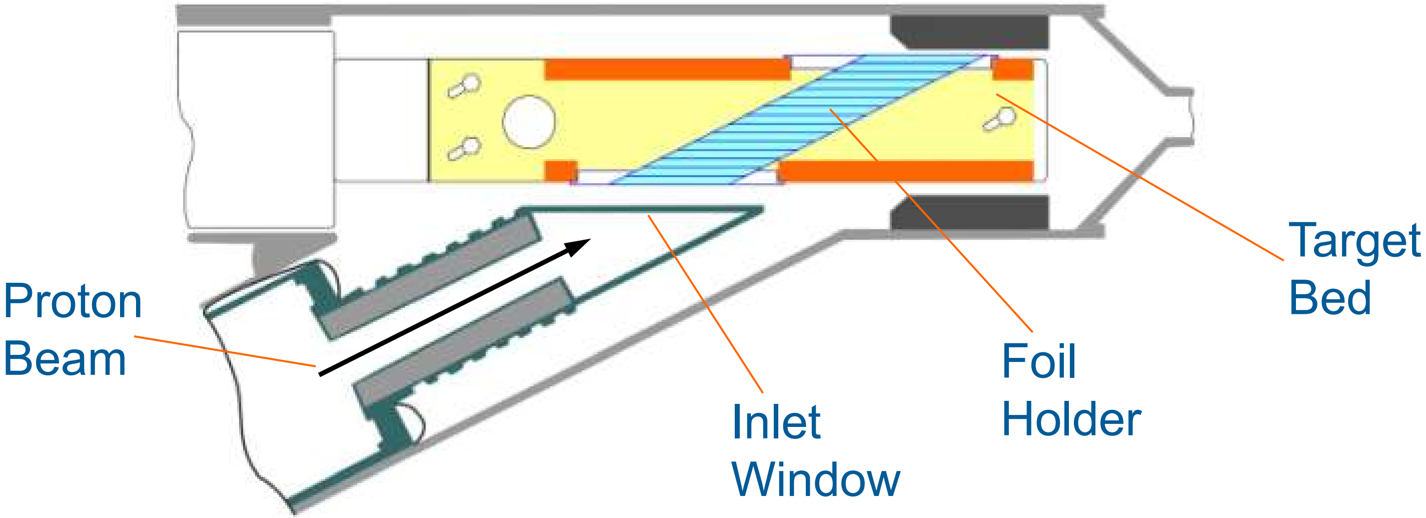
Targetry

Cross-Section measurement

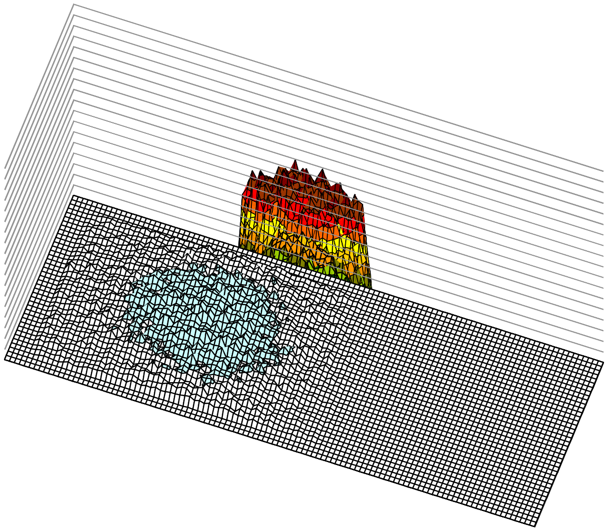
- Optimal proton energy range and target thickness
- Activity of the main radionuclide
- Radionuclide impurities

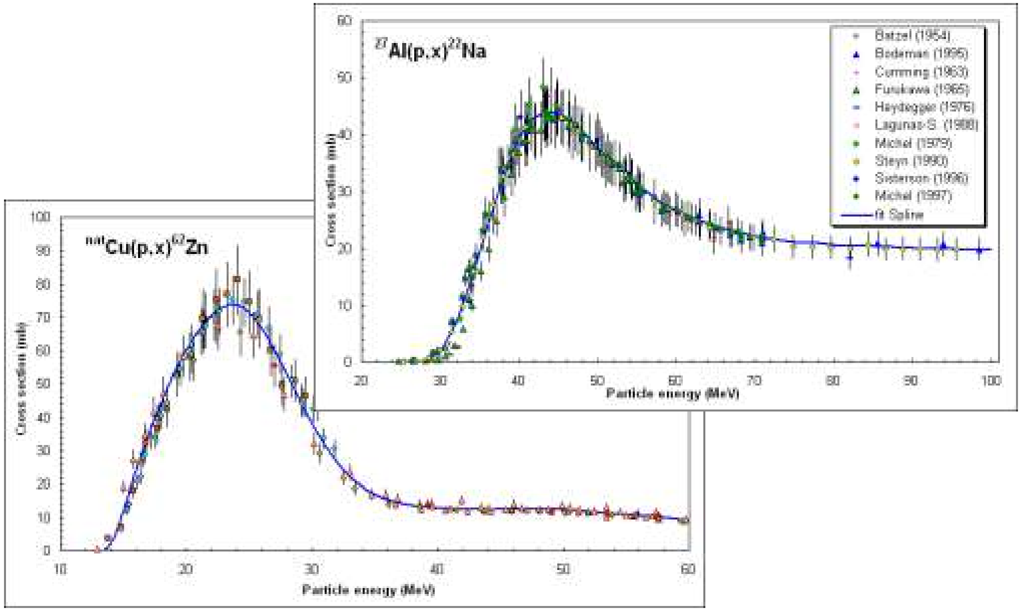
Cross Section Measurement

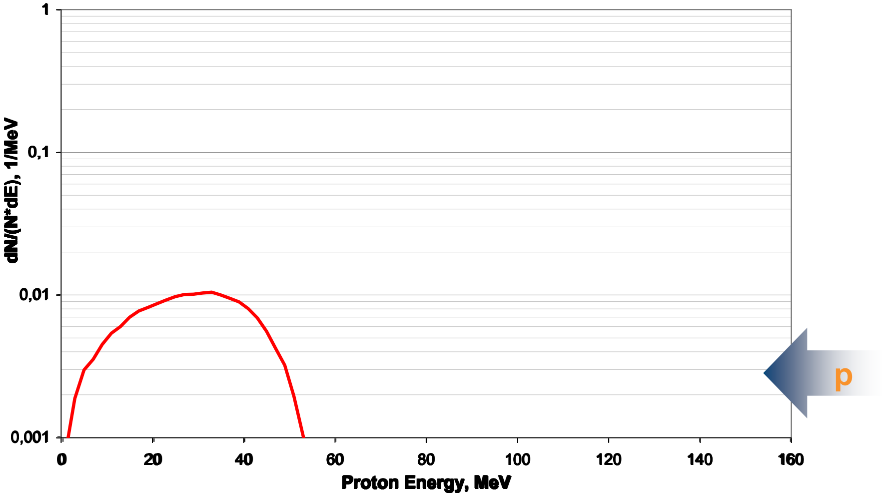


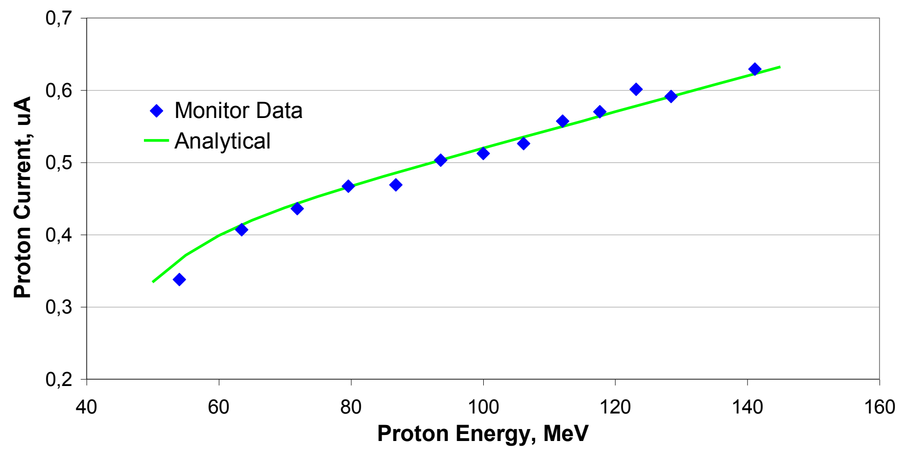


Monitor Data: taking into account proton straggling and scattering



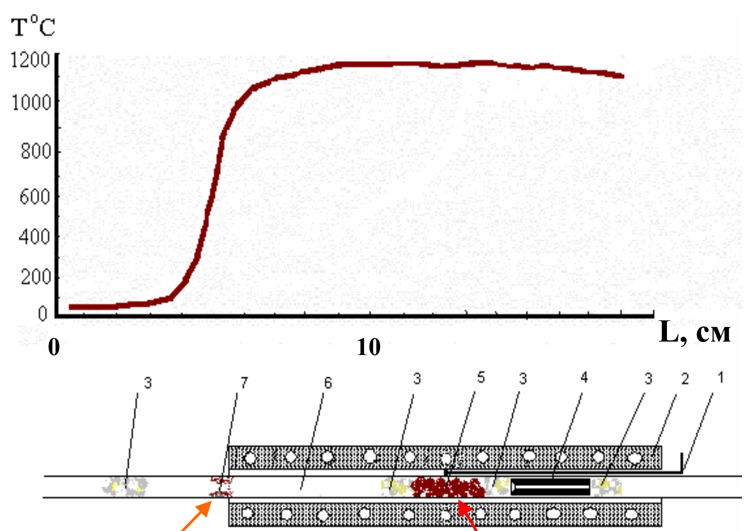






^{117m}Sn Cross Section Determination:

Gas Chemical Separation of Radiotin from Antimony and Tellurium Radionuclides

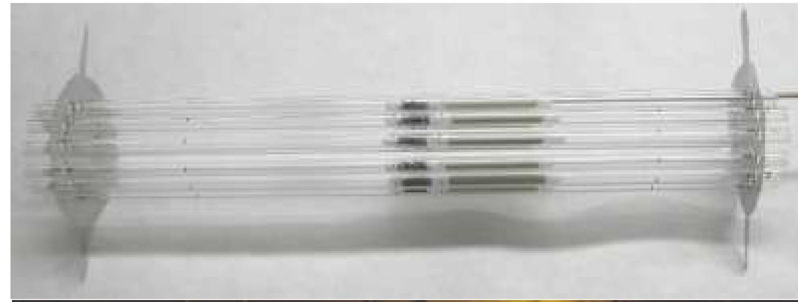


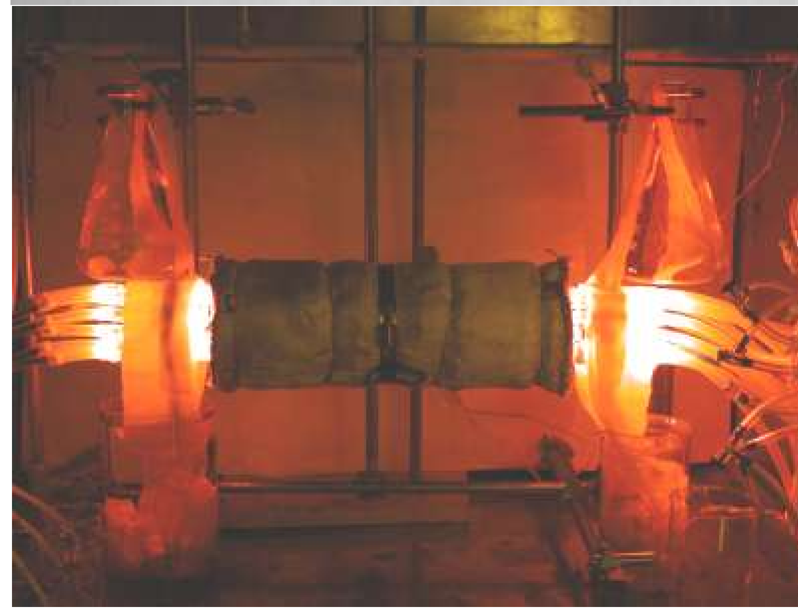
Air

or Helium

^{122}Sb, ^{120m}Sb, ^{124}Sb
^{123m}Te, ^{118}Te, ^{119m}Te

^{117m}Sn, ^{113}Sn

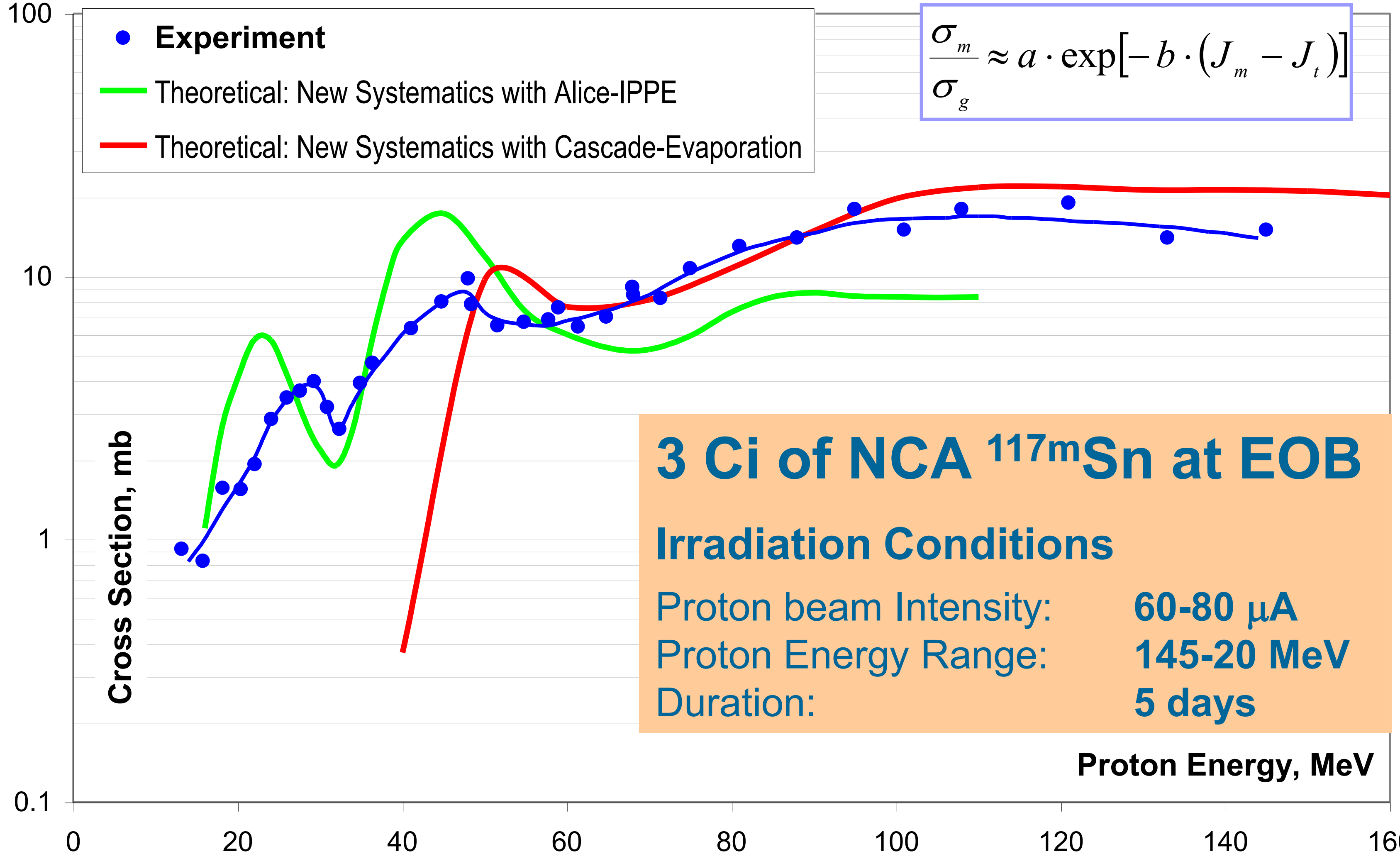




^{117m}Sn: Estimation of Activity at EOB

New Systematics for Isomeric Ratio [1]

$$\frac{\sigma_m}{\sigma_g} \approx a \cdot \exp\left[-b \cdot \left(J_m - J_t\right)\right]$$



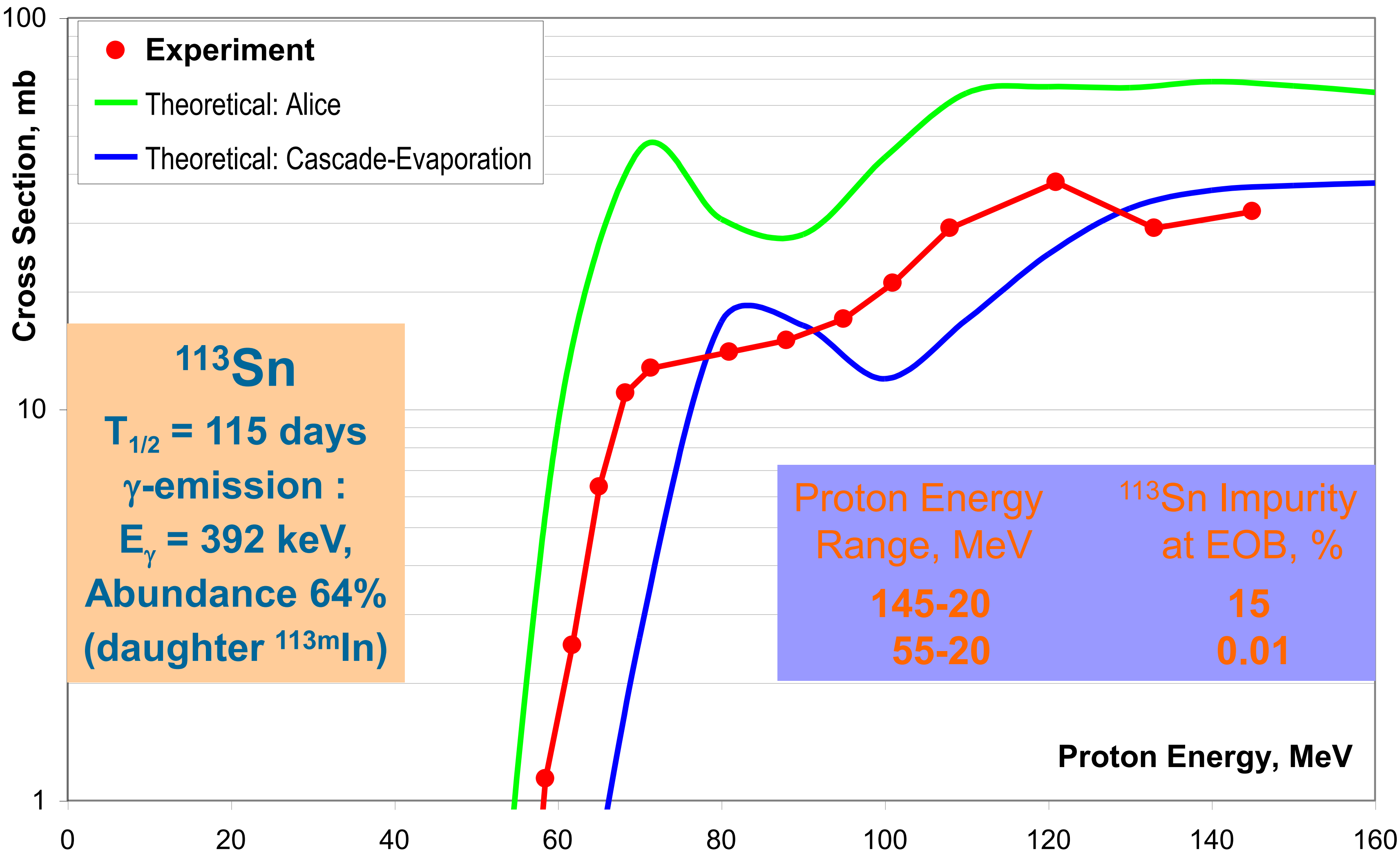
3 Ci of NCA ^{117m}Sn at EOB

Irradiation Conditions

Proton beam Intensity:	**60-80 μA**
Proton Energy Range:	**145-20 MeV**
Duration:	**5 days**

[1] B.L. Zhuikov, A.S. Iljinov, *Physics of Atomic Nuclei*, 2006, **69**, p.739.

^{113}Sn Impurity: The Choice of Proton Energy Range

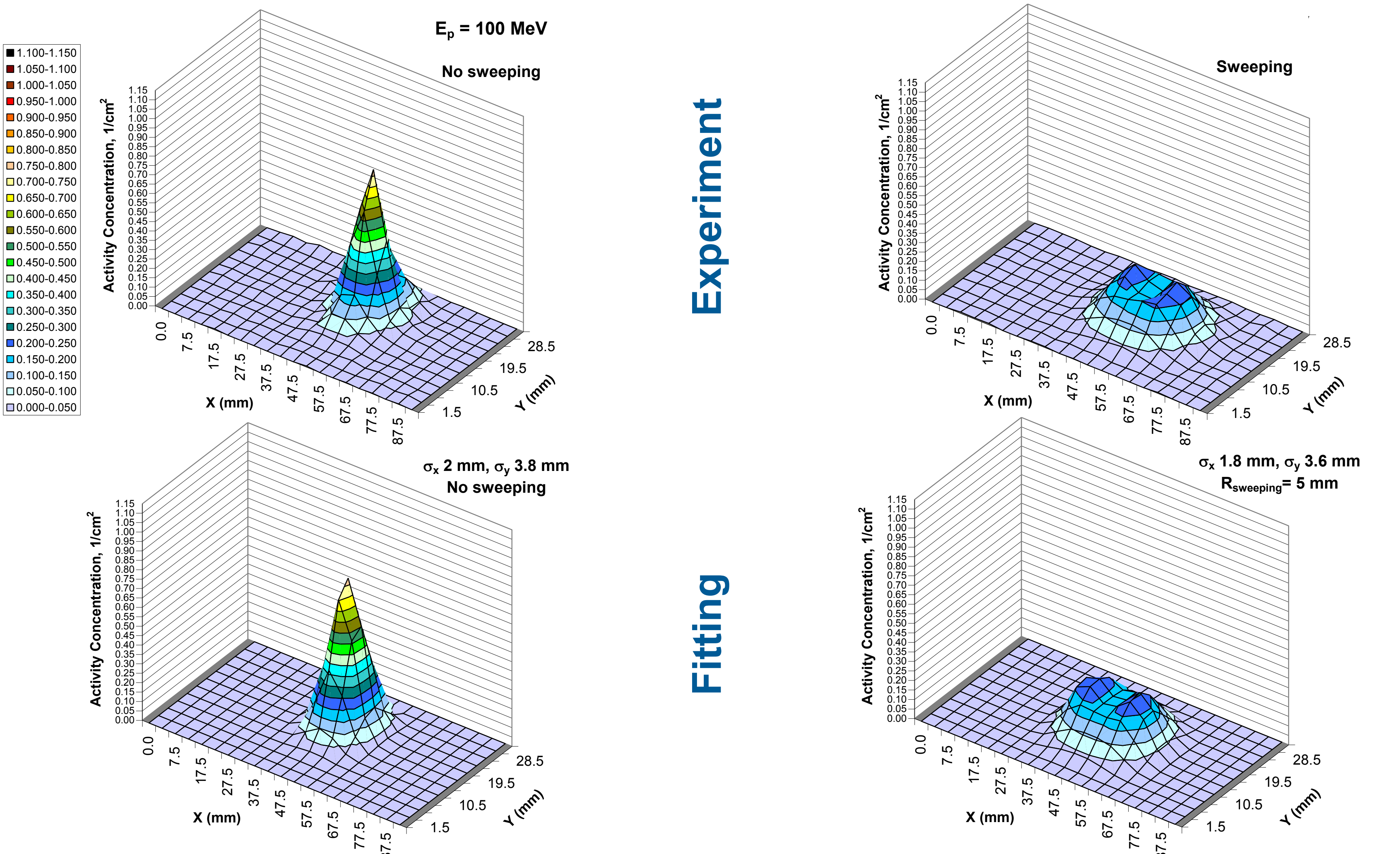


Targetry

Development of target design

- Testing beam shape and positioning
- Choice of materials for target and target shell
- Geometric parameters of target and estimation of beam losses
- Temperature generated in target and estimation of the most possible current of proton beam

Beam Shape and Positioning: Irradiation and Scanning Al Foils



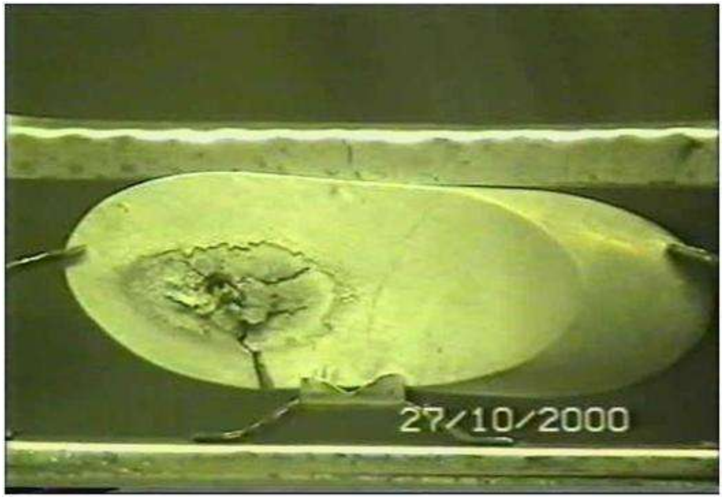
General Requirements for Target Material at High Intensity Beam

- High cross-section to form the desired radionuclide in the given particle energy range
- High abundance of the main producing material
- Availability and acceptable price
  (low energy protons or heavy ions – enriched stable isotopes,
  middle energy protons – natural mixture of isotopes)
- Known and acceptable non-radioactive impurities
  (especially, impurity of stable isotopes of the obtained element)
- High temperature stability
- High heat conductivity
- High radiation stability
- Low vapor pressure
- Low reaction ability with available material of shell, OR
- Low reaction ability with cooling water and its products of radiation decomposition
- Low toxicity of the main material and impurities
- Acceptability for radiochemical processing

A compromise is needed!

What Happens if the Target Requirements are not Fulfilled?

Not only thermal but also radiation impact



Pure Aluminum

Graphite

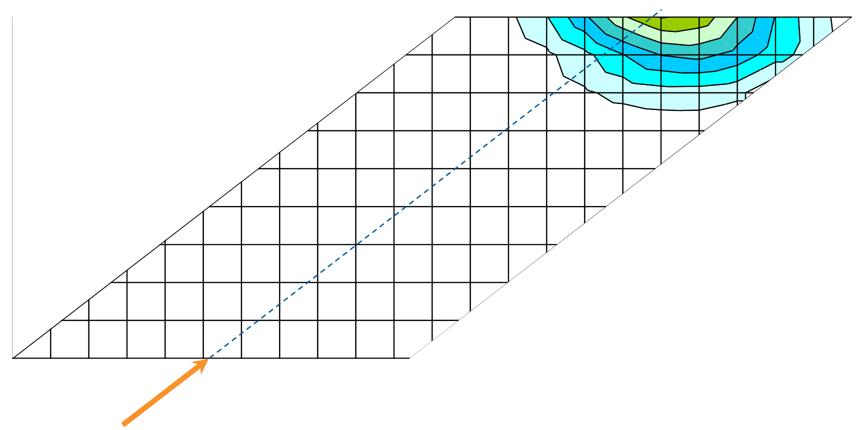
Molybdenum

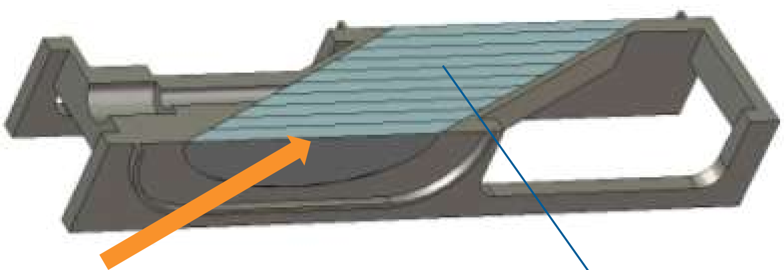
Target Geometry: distribution of protons absorbed inside the target

Rb Target ($^{nat}Rb\ (p,x)\ ^{82}Sr$):

Metallic Rubidium in a Stainless Steel Shell

Diameter	25 mm
Thickness	30 mm
Angle	26°

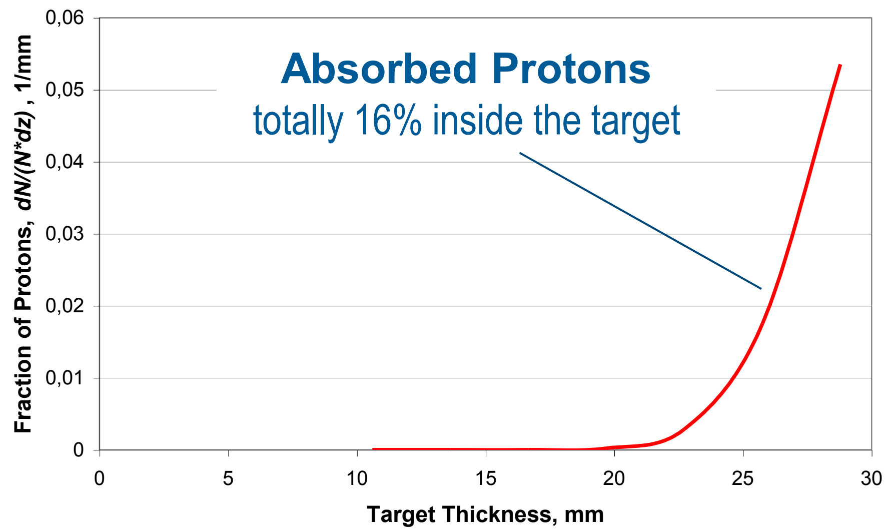




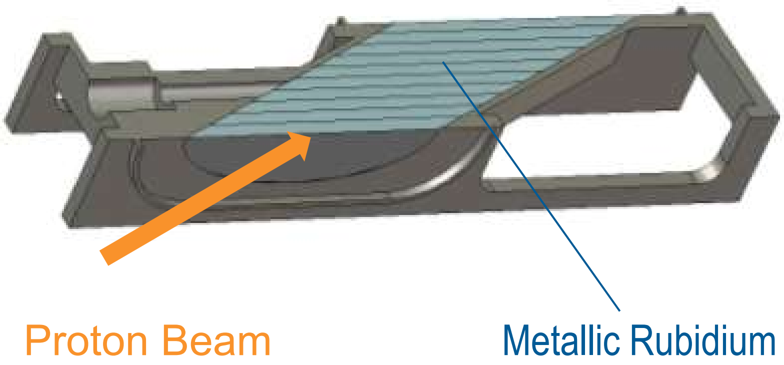
Proton Beam

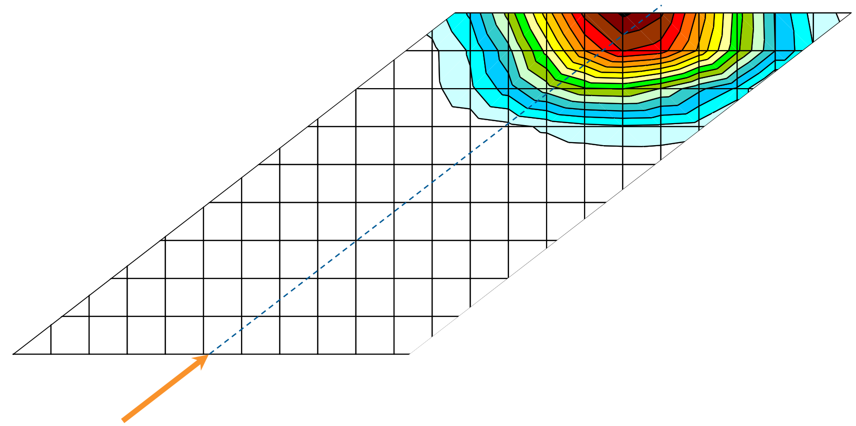
Metallic Rubidium

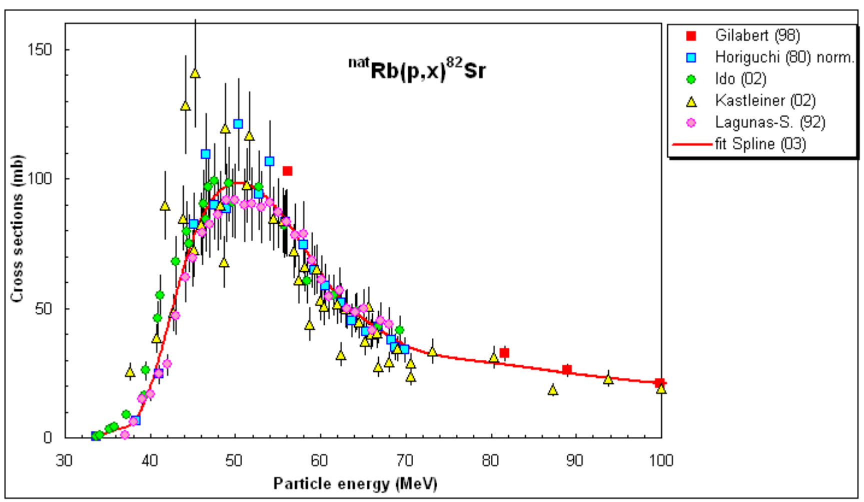
E_p	**143 MeV**
E_{inlet}	**89 MeV**
E_{outlet}	**26 MeV**

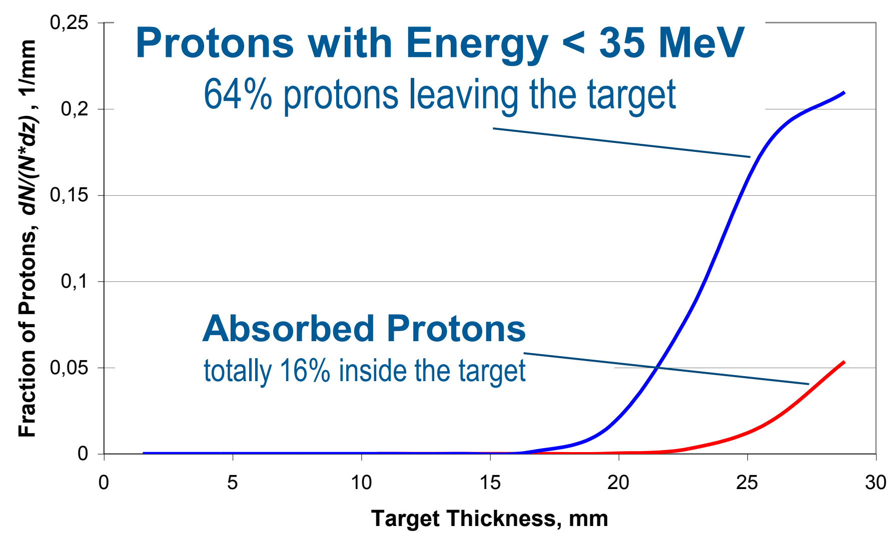


Target Geometry: distribution of “useless” protons inside the target

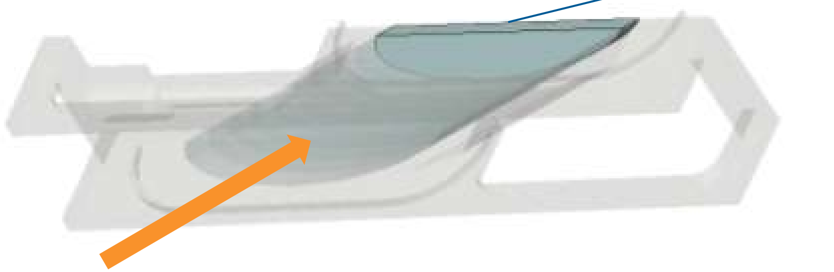








Target Geometry: beam losses through lateral surface of the target



Diameter 25 mm
Thickness 30 mm

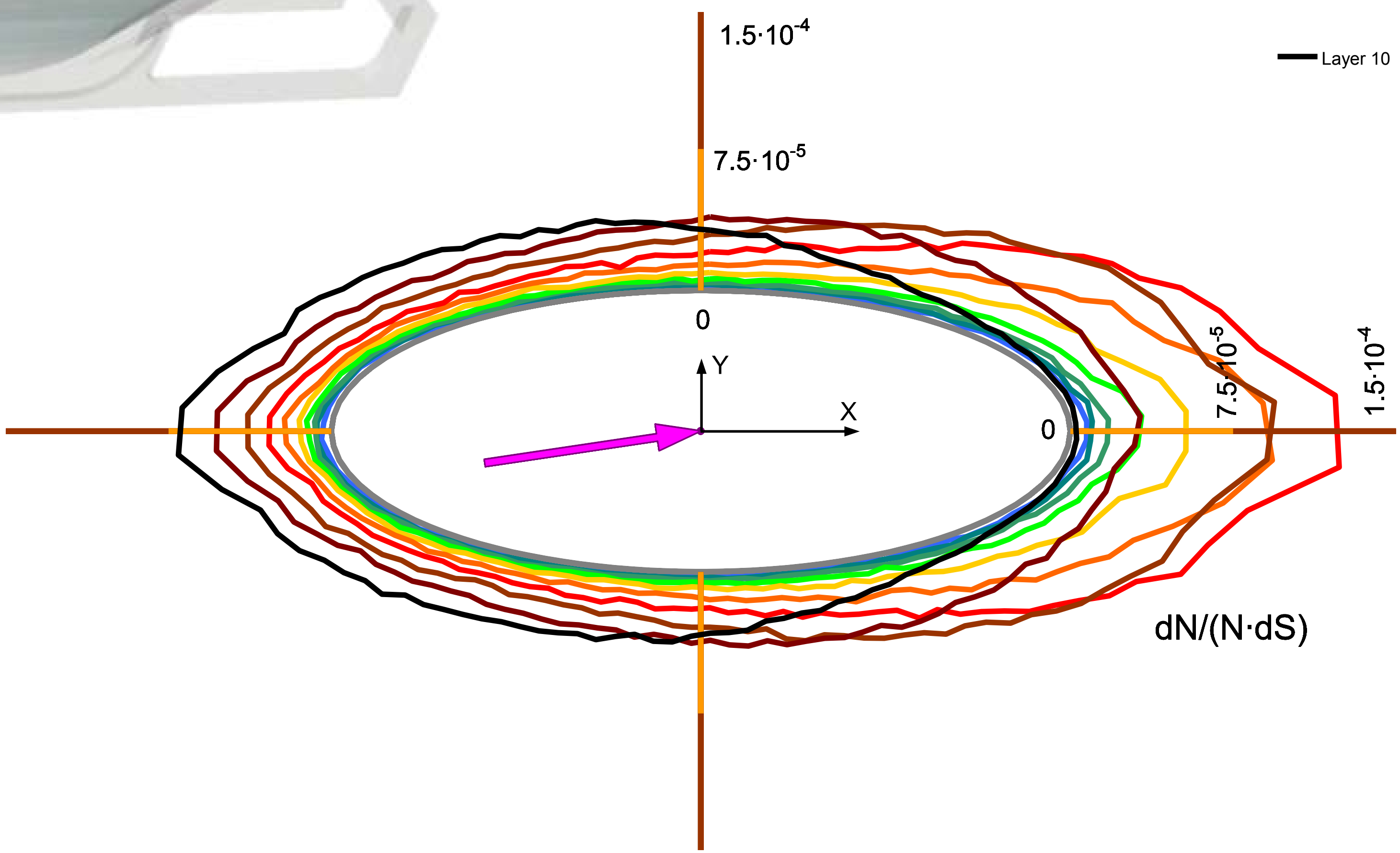
Proton Beam

Gaussian Form

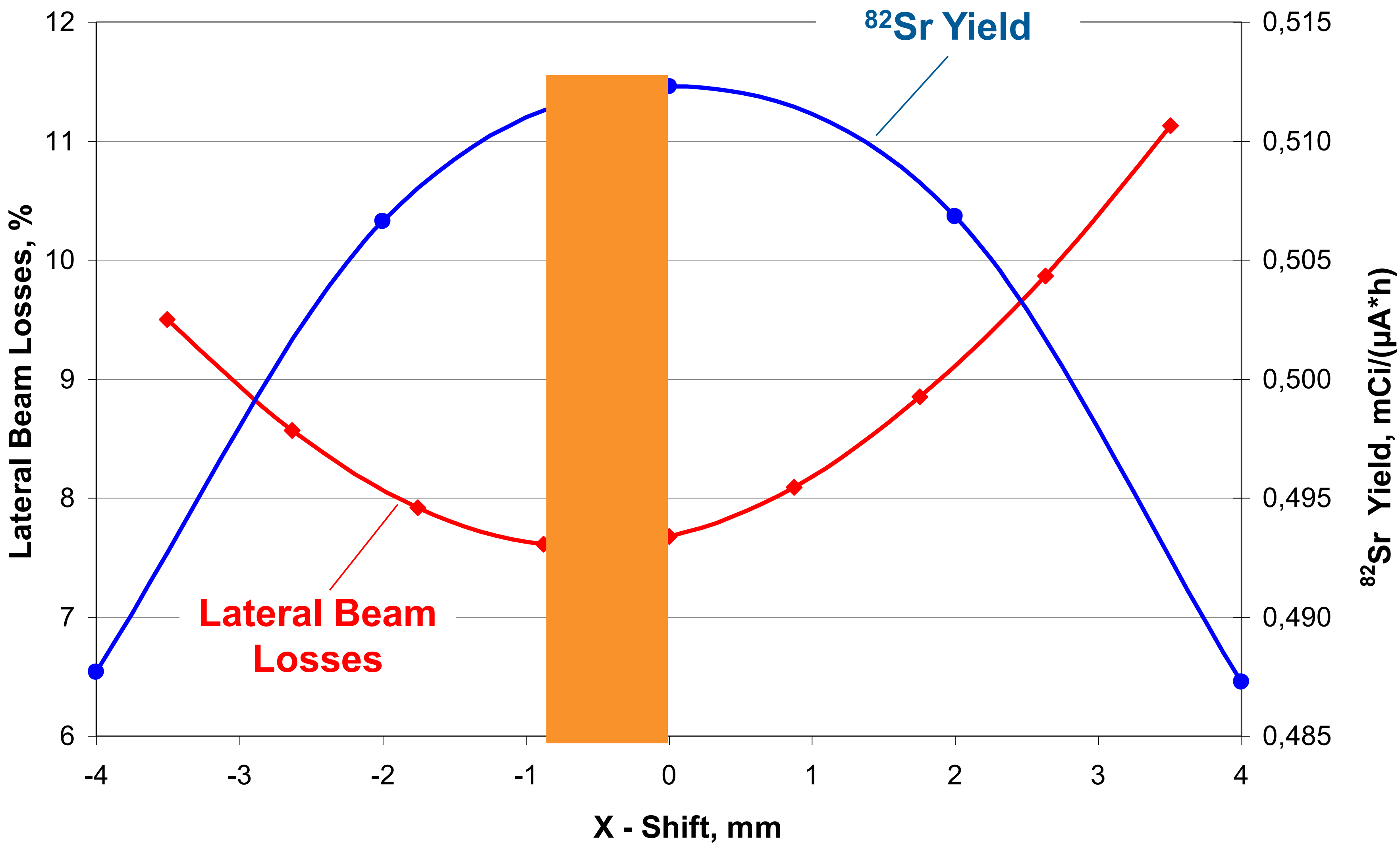
σ 4 mm

Angle 26°

X-shift 0 mm

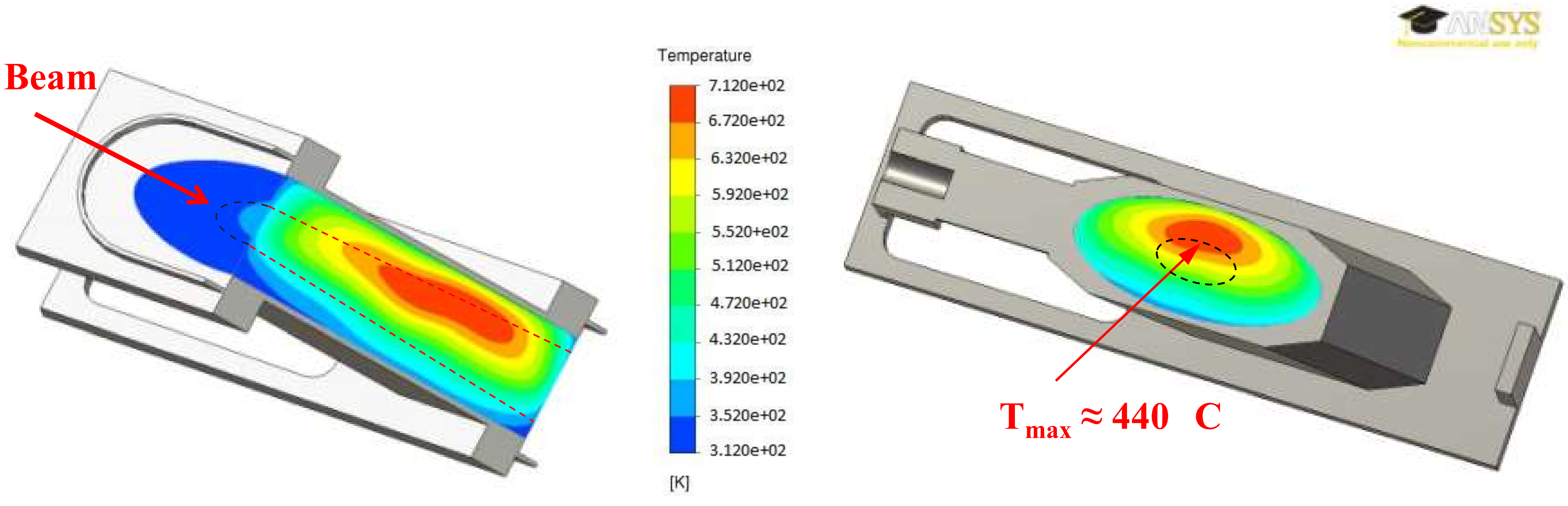


Target Geometry: influence of X-shift of proton beam

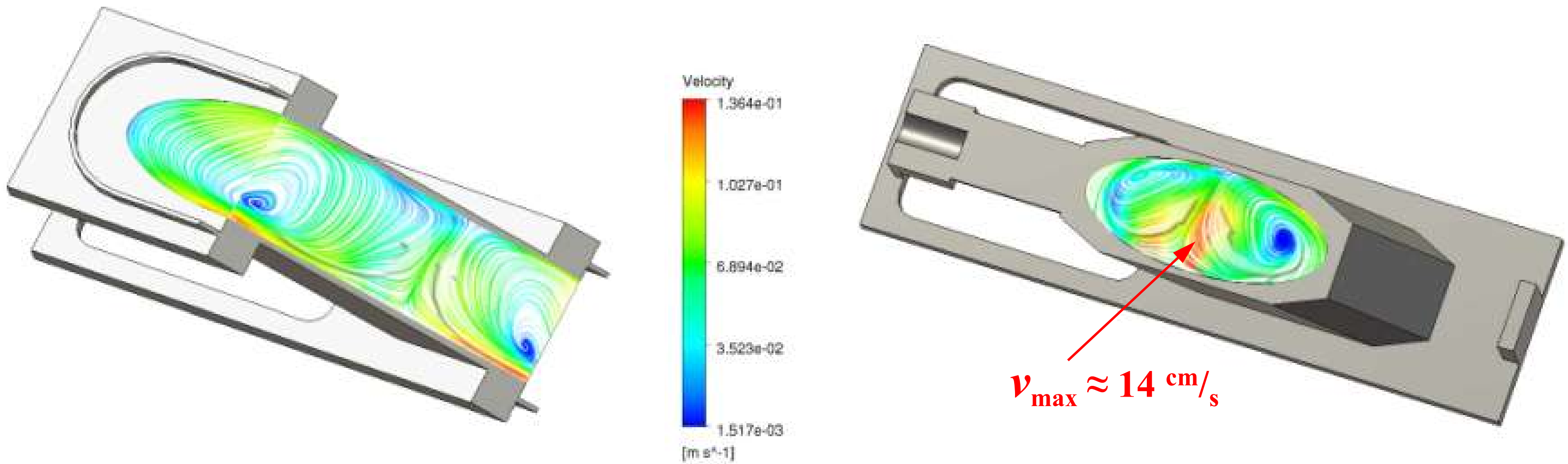


Temperature and **Velocity Distributions** in Liquid Rb (100 μA, σ = 4 mm)

Temperature Distribution

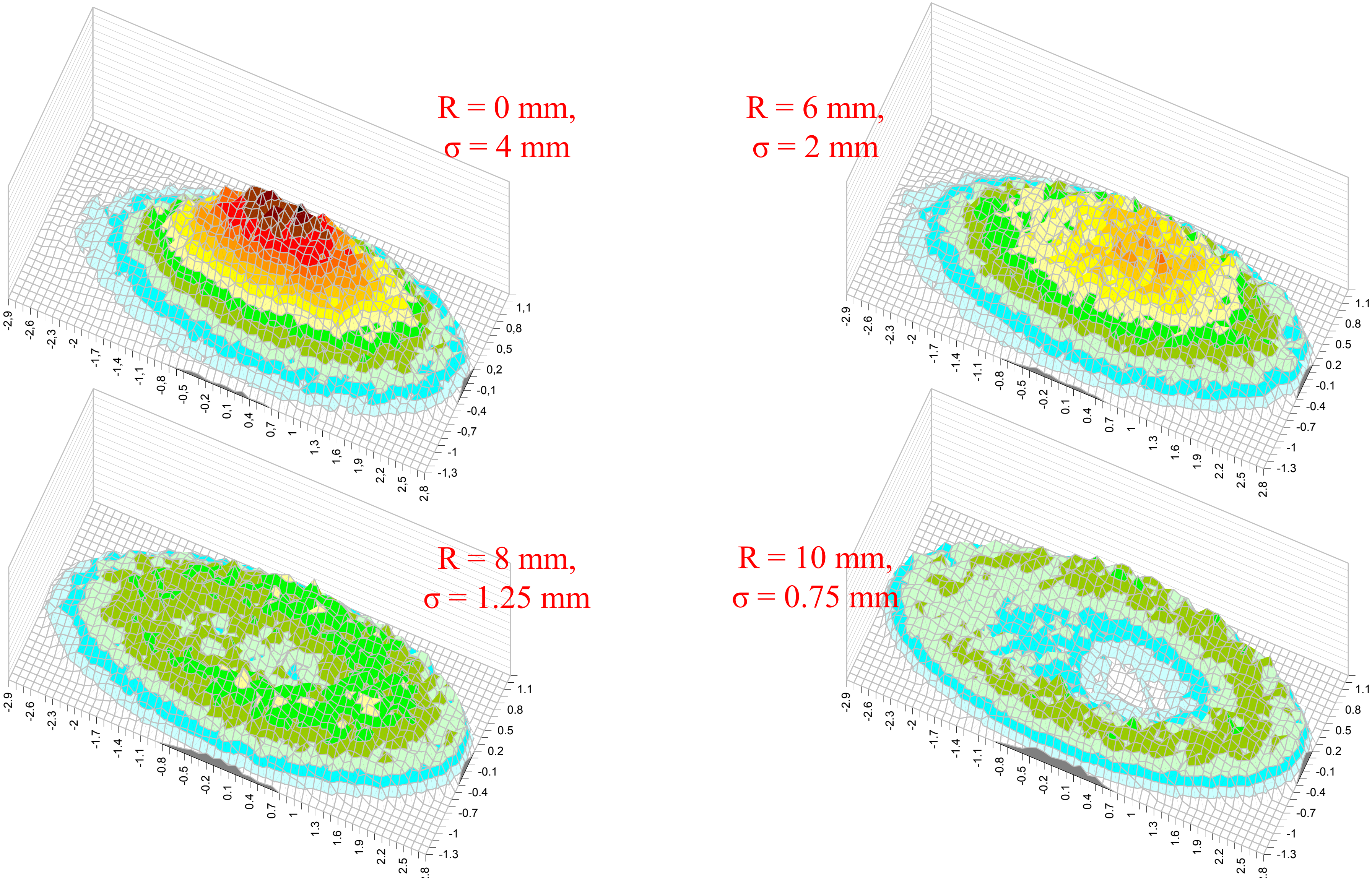


Velocity Distribution



Heat Release in the Middle of Rb Target

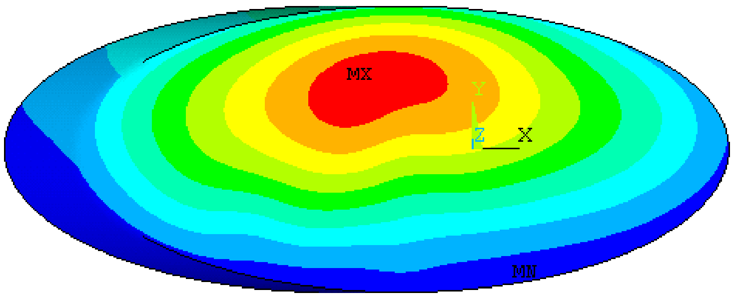
at different sweeping parameters (R – sweeping amplitude)



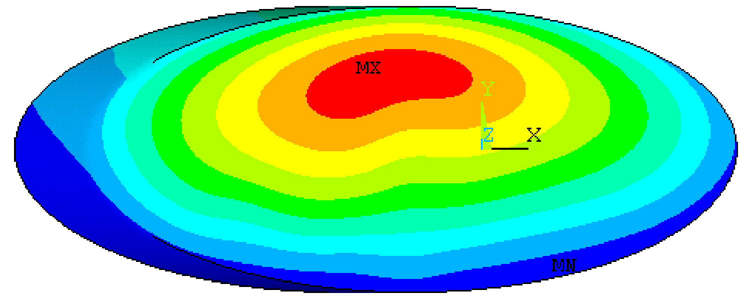
Temperature Profiles Inside Rb Target

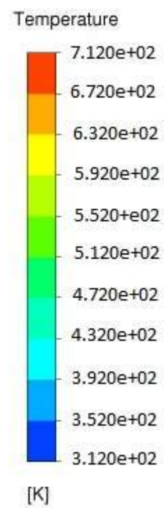
as a function of different sweeping parameters

R = 0 mm, σ = 4 mm, **I = 100 μA**

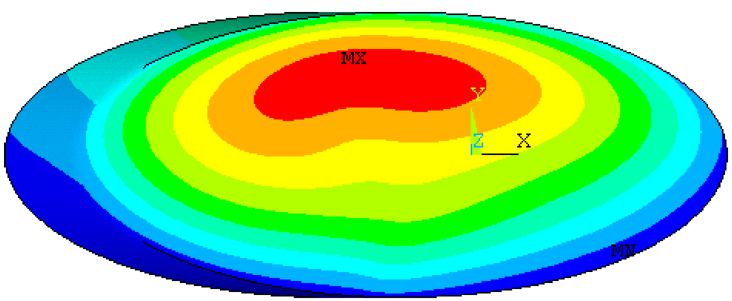


R = 6 mm, σ = 2 mm, **I = 110 μA**

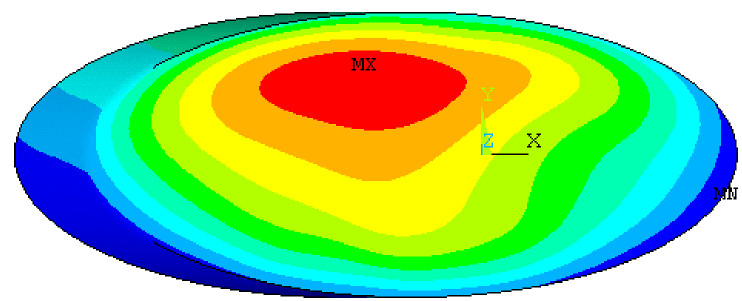




R = 8 mm, σ = 1.25 mm, **I = 145 μA**



R = 10 mm, σ = 0.75 mm, **I = 190 μA**



Targetry

Upgrade to Large-Scale Production

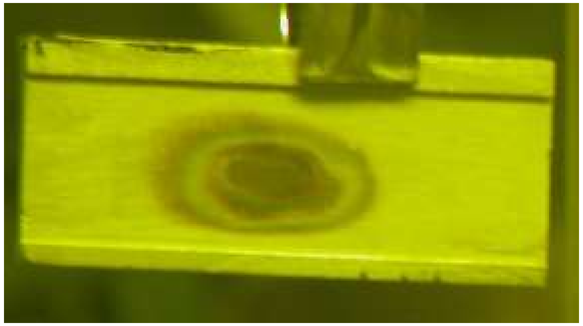
- Fabrication and irradiation of experimental targets
- Control of target integrity
- Optimization of target design for routine production

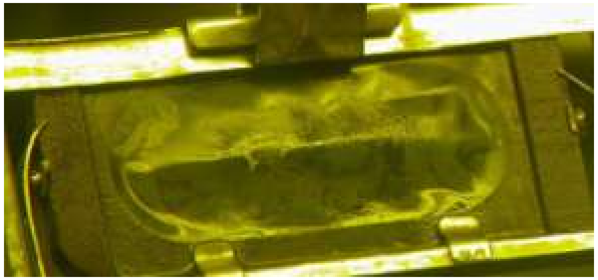
Experimental Sb Targets for ^{117m}Sn Production

Stainless Steel Shell	Graphite Shell Ni-electroplated	Molybdenum Shell

before irradiation

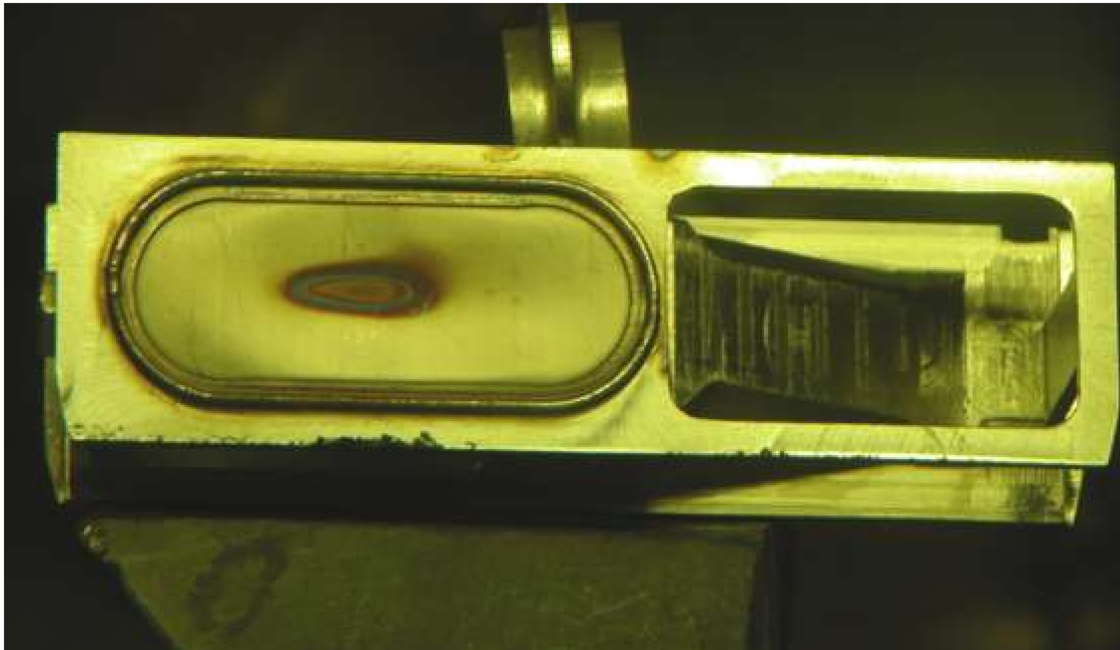
after irradiation



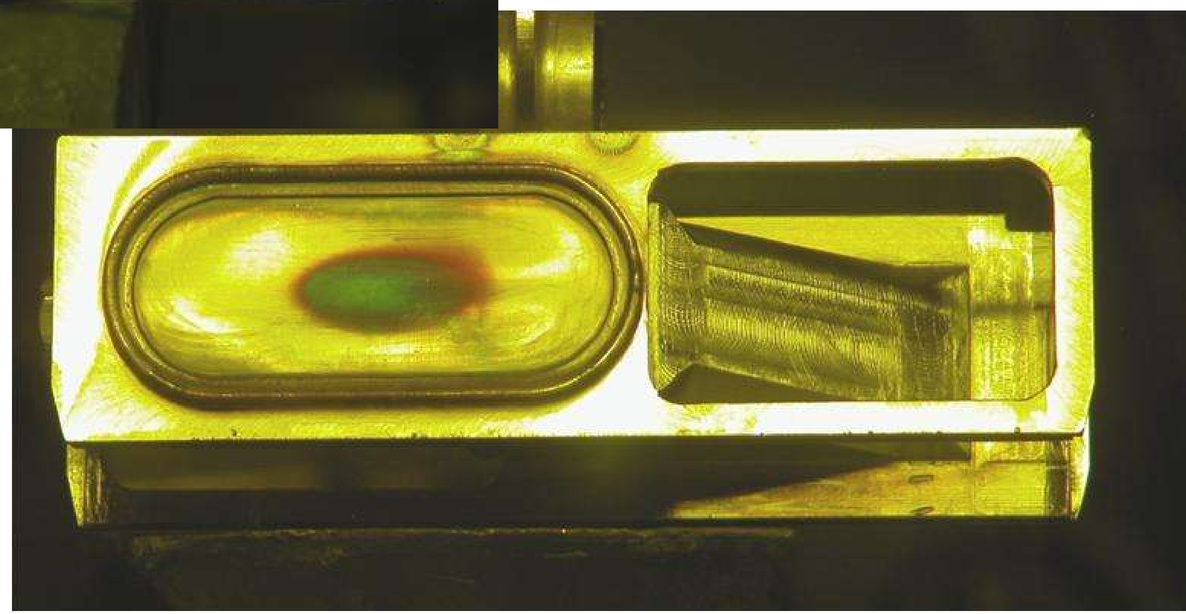


Influence of Beam Sweeping on Rb Target Irradiation

Inlet Window



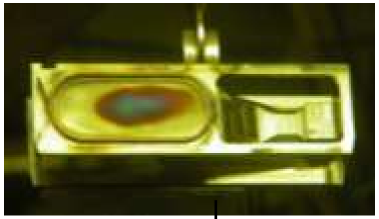
Not sweeping beam
8,000 μA·h
90 μA



Sweeping beam
R ~ 2mm
10,000 μA·h
110 μA

Targets Developed and Irradiated at INR

Rb



^{82}Sr

- -Los Alamos, USA
- -IPPE, Obninsk

Ag

^{103}Pd

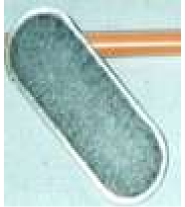
- -Karpov,Obninsk
- -Mayak, Ozersk

Sb

^{117m}Sn

- IPPE, Obninsk
- Brookhaven, USA

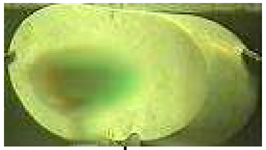
Ga



^{68}Ge

- -Los Alamos, USA

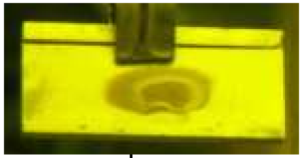
Al



^{22}Na

- -IPPE, Obninsk

Th



^{225}Ac, ^{223}Ra

- - Karpov, Obninsk

Radionuclide Recovery and Application

Radionuclides Produced at INR

Possible Activity for Generation in One Accelerator Run at 120 μA

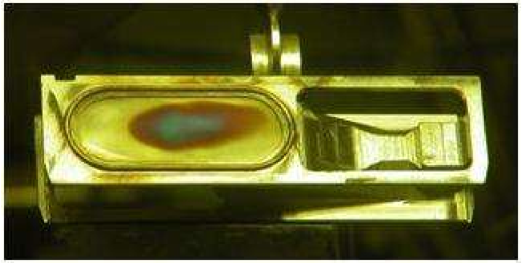
Radionuclide	Half life	Target	Energy range, *MeV*	Bombardment period, *hours*	Activity produced in one run at EOB, *Ci*
^{82}Sr	25.5 d	Rb	100-40	250	5
^{22}Na	2.6 y	Mg, Al	150-35	250	2
^{109}Cd	453 d	In	150-80	250	2
^{103}Pd	17 d	Ag	150-50	250	50
^{68}Ge	288 d	Ga, GaNi	50-15	250	0.5
^{117m}Sn	14 d	Sb, TiSb	150-40	250	3
^{72}Se	8.5 d	GaAs	60-45	250	3
^{67}Cu	62 hr	Zn-68	150-70	100	10
^{64}Cu	12.7 hr	Zn	150-40	15	15
^{225}Ac	10 d	Th	150-40	250	4
^{223}Ra	11.4 d	Th	150-40	250	13

Green – regular mass production

Blue – technology developed, test samples supplied to customers

Red – production method developed, technology under development

Processing INR Rb target and recovery ^{82}Sr at Los Alamos



Traditional chemical approach:
Rb dissolution followed by ion exchange separation

Over 100 targets were irradiated and shipped

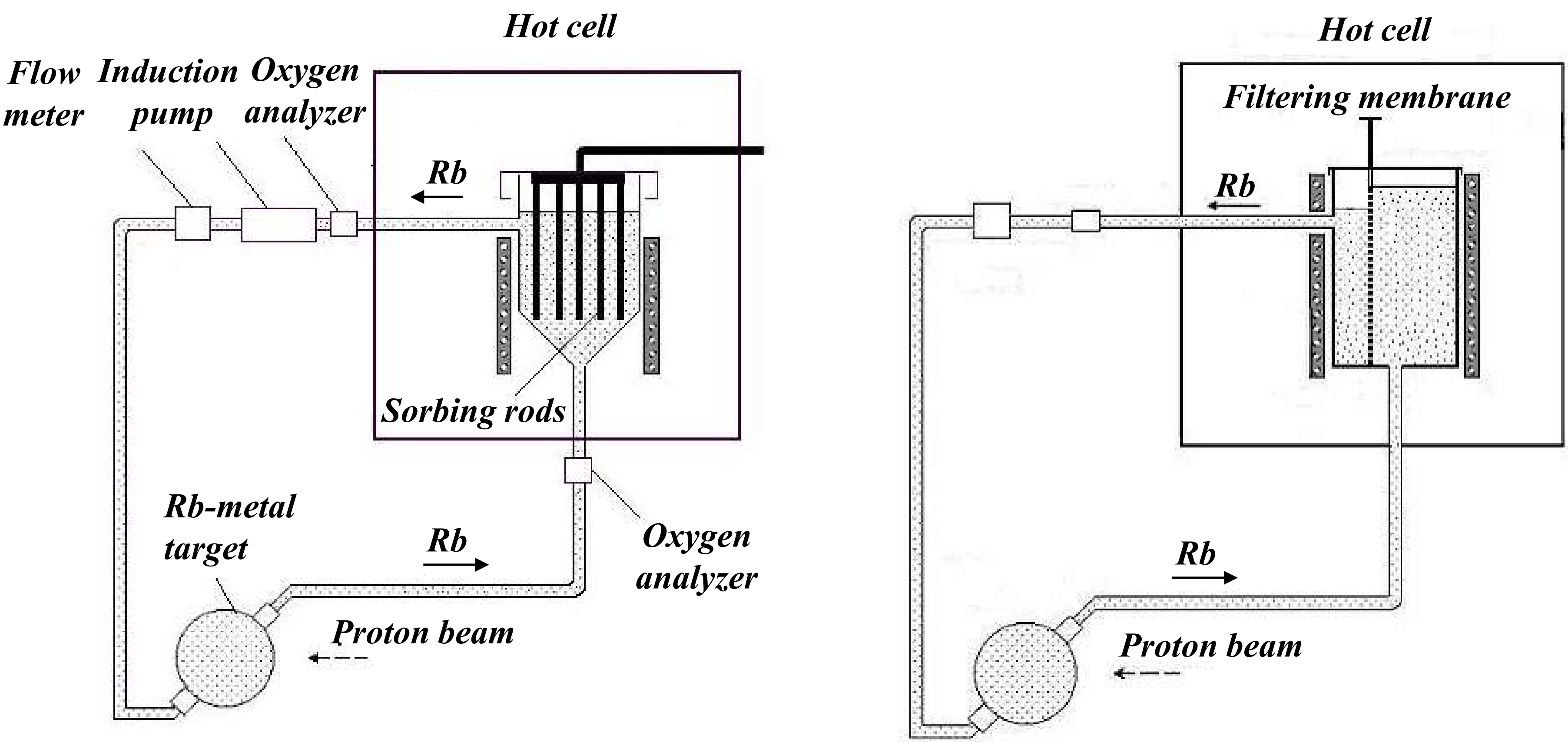
New Technology of ^{82}Sr Recovery Developed by INR

IPPE (Obninsk, RUSSIA)

Direct ^{82}Sr sorption from liquid metallic rubidium containing oxygen (0.1 – 4 wt.%)

ARRONAX (Nantes, FRANCE)

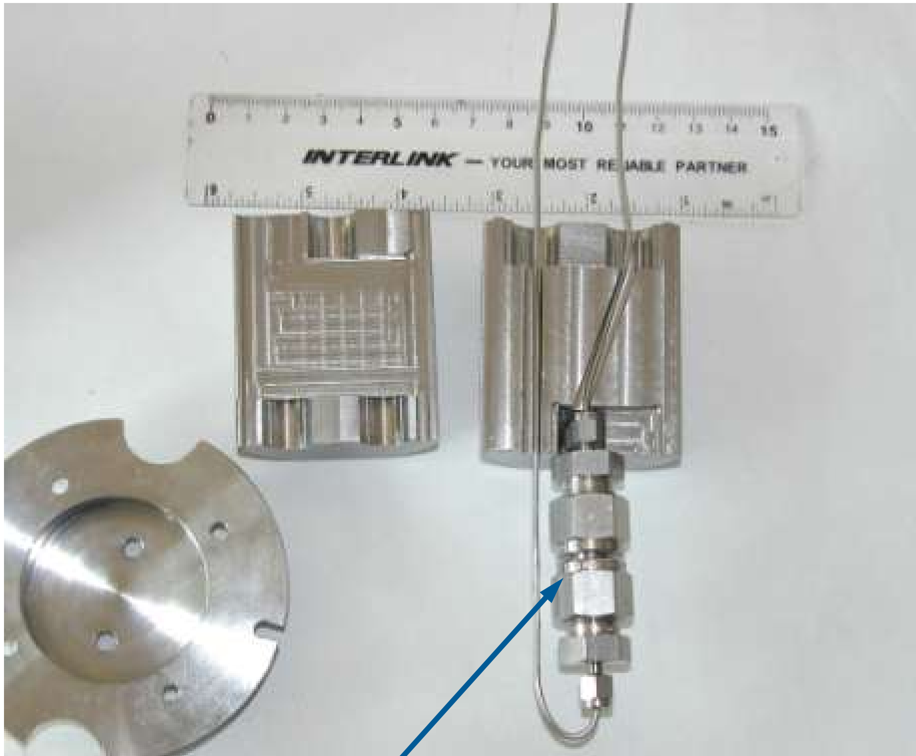
Development of On-Line ^{82}Sr Production



Rb volume	*1.5 L*
Rb flow	*5-10 L/min*
Target diameter	*8 cm*

Beam current	*500 μA*
Proton energy range	*63-36 MeV*
Production capacity ^{82}Sr	*>3 Ci/day at EOB*

Construction of Sr/Rb-82 Generator (GR-01, INR)



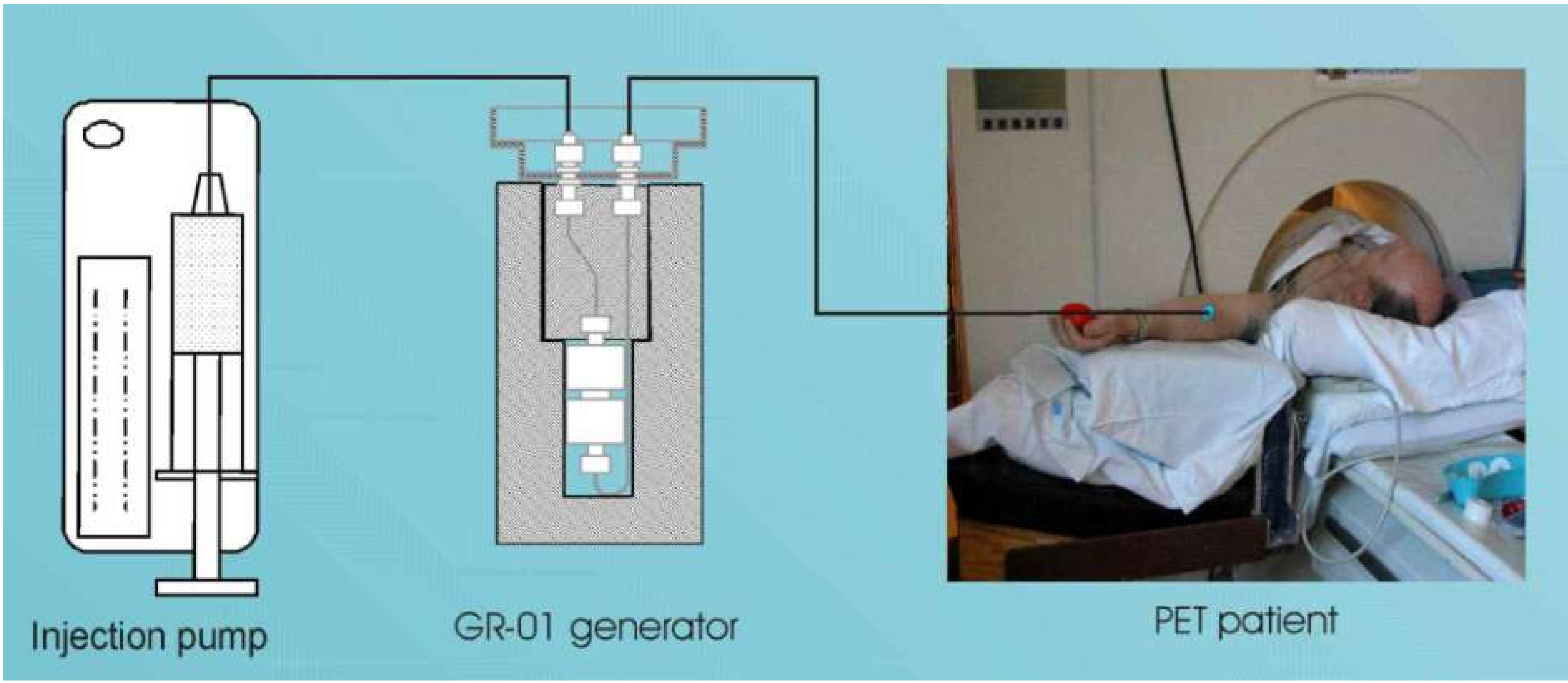
*^{82}**Sr** (25.5 d) → ^{82}**Rb** (1.3 min)*

Sr/Rb-82 generator
in tungsten shielding container

Chromatographic column
filled with α-hydrous tin oxide

PET Diagnostics using INR Sr/Rb-82 Generator

(RRC Radiology and Surgery Technologies, St. Petersburg)



Conclusion

Great interest in medical radionuclides

- ^{82}**Sr,** ^{225}**Ac,** ^{223}**Ra**
- ^{117m}**Sn,** ^{64,67}**Cu**

Growing demand for development of **compact** accelerators providing an **intensive** beam of **medium-energy protons** and proper **targetry**

Cyclotron **ARRONAX** (Nantes, France)
Proton energy **70 MeV**
Beam Intensity **2   375 µA**

Designed and constructed by **IBA** (Belgium), fully operational since January 2011

IBA is going to install several 70 MeV cyclotrons in the nearest years (including one in Russia)

Thank you !