Horia Hulubei National Institute for R&D
in Physics and Nuclear Engineering

IFIN-HH

Consolidating Target Manufacturing Methods for Nuclear Physics Experiments at Accelerators

Drd. Andreea Mitu

GSI

GSI, Darmstadt 2019

Research outlines:

1. Target laboratory in IFIN-HH
2. Methods of interest in target preparation and characterization
3. Specific examples and particularities
4. Conclusions

❖ Where?

Horia Hulubei National Institute for Physics and Nuclear Engineering
IFIN-HH

Bucharest-Magurele, Romania

❖ General information

Brief History

- ✓ 1949: Institute of Physics of the Romanian Academy
- ✓ 1956: Institute of Atomic Physics (IFA)
- ✓ 1977: Central Institute of Physics (ICEFIZ)
- IFIN (nuclear),
- IFTAR (radiation equipment), ω IFTM (materials),
- CFPS (earth),
- IGSS (space),
- ✓ 1990: ICEFIZ became IFA
- ✓ 1996: IFIN-HH

Horia Hulubei (1896-1972)
Founder and First Director of the Institute

Contribution to the study of quantum diffusion of X-rays
(Ph.D. Thesis, Paris, 1933)
Supervisor: Jean Perrin (Nobel Prize)
Chairman: Marie Curie (Nobel Prize)

❖ Research Areas

Basic Physics Research

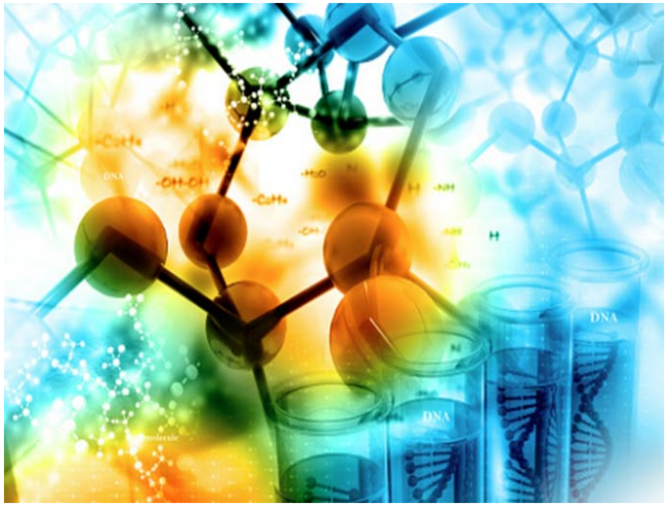
Nuclear Physics and astrophysics

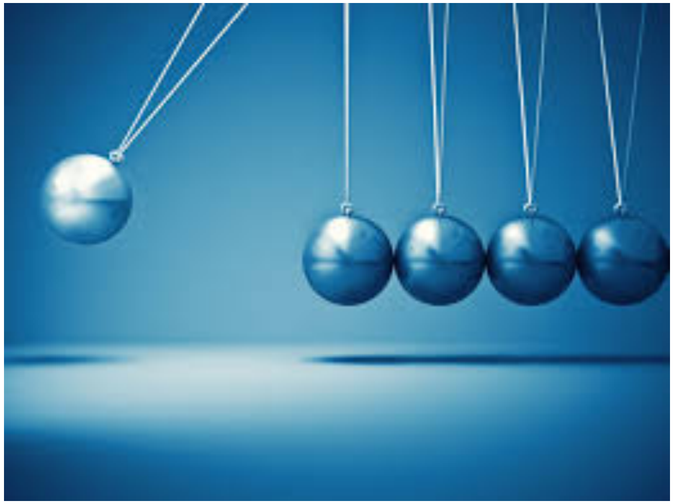
Particle physics and field theory

Atomic physics and condensed matter physics

Mathematical physics and information physics

Life and environmental protection





Applied Physics Research

Advanced detection systems

Nuclear safety, radiation protection and radioactive products

Radioecology and nuclear biomedicine

Nuclear techniques and applications

Advanced communication systems

❖ Research Departments

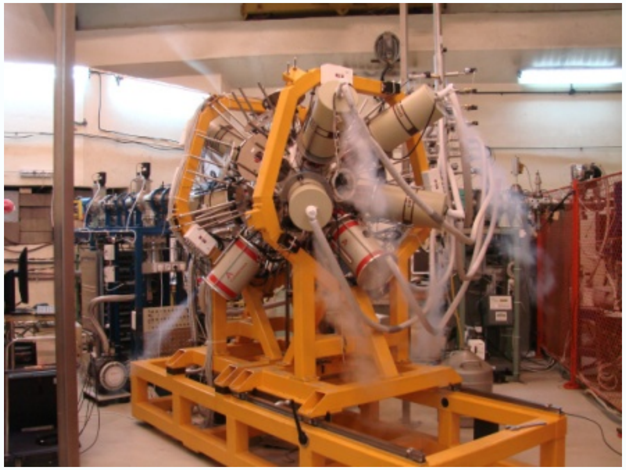
1. ***Theoretical physics***
2. ***Nuclear physics***
3. ***Tandem Accelerators***
4. ***Hadronic Physics***
5. ***Elementary Particles Physics***
6. ***Computational Physics and Informational Technologies***
7. ***Life and Environmental Sciences***
8. ***Radioisotopes and Radiation Metrology***
9. ***Applied Nuclear Physics***
10. ***Multipurpose Irradiation Facility Centre***
11. ***Nuclear Training Centre***
12. ***Reactor Decommissioning***
13. ***Radioactive Waste Management***
14. ***Protection and Prevention Compartment***
15. ***The Centre for Technology Transfer and Marketing***

Details on: http://www.nipne.ro

❖ Experimental Infrastructure in IFIN-HH

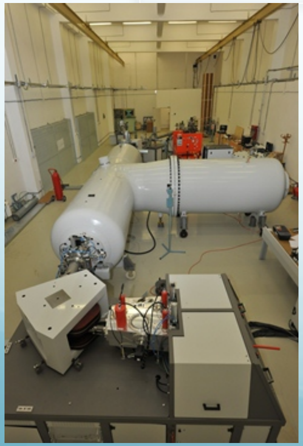
9 MV Tandem

ROSPHERE gamma detectors array



19 MeV Proton Cyclotron for production of radioisotopes

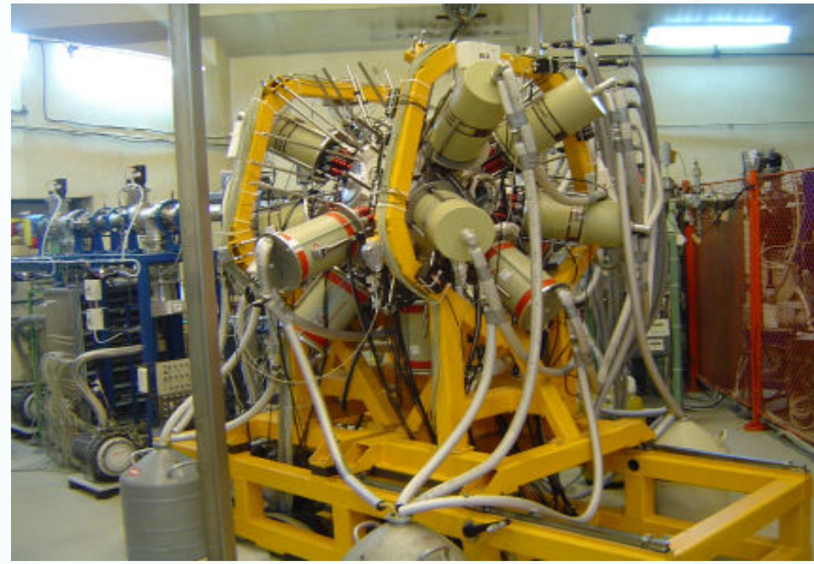
3MV Tandetron



1MV Accelerator Mass Spectrometry

Gamma-ray Irradiator

❖ 9MV TANDEM ACCELERATOR



ROSPHERE - **RO**manian array for γ-**SP**ectroscopy in **HE**avy ion **Re**actions

25 positions on 5 rings (37° ,70°, 90°, 110°, 143°)

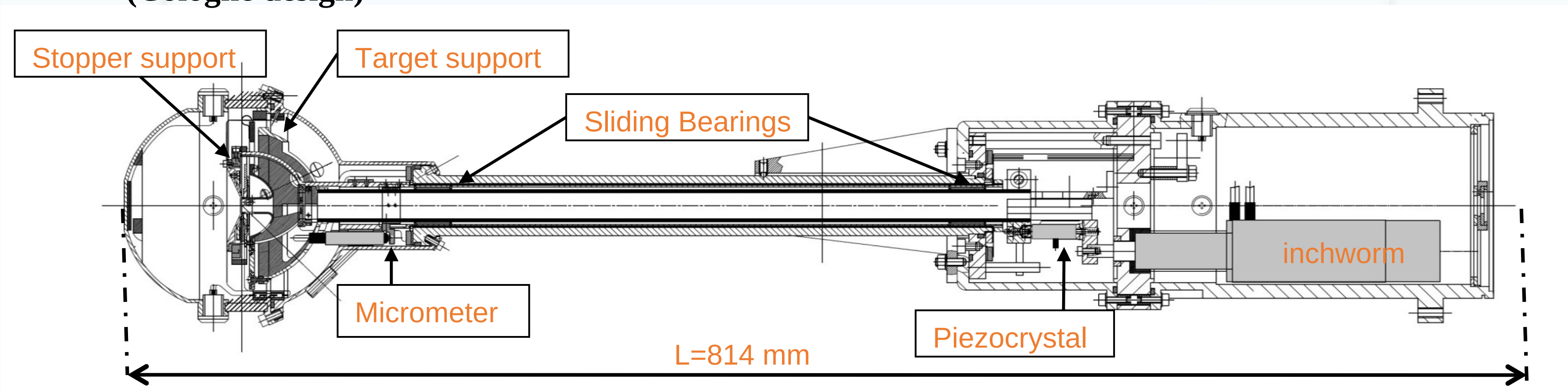
2 configurations : - 15 50% HPGe detectors with BGO shields and 10 to 20 $LaBr_3$(Ce) scintilators

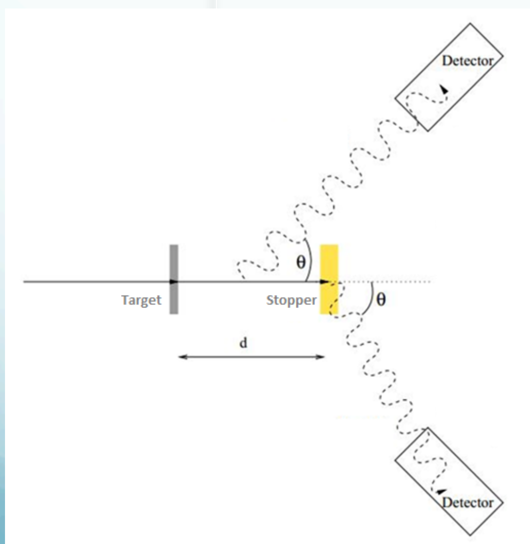
- 25 50% HPGe detectors

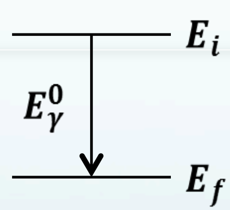
❖ Types of experiments performed at 9MV tandem accelerator

PLUNGER reaction chamber (Cologne design)

Recoil Distance Doppler Shift method (RDDS)

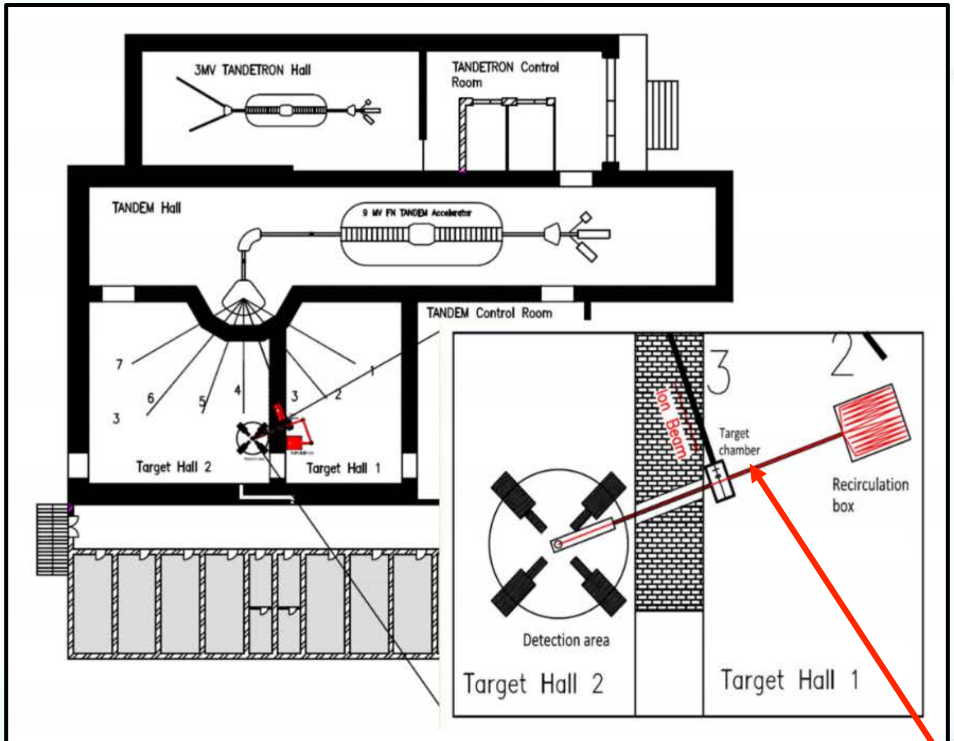






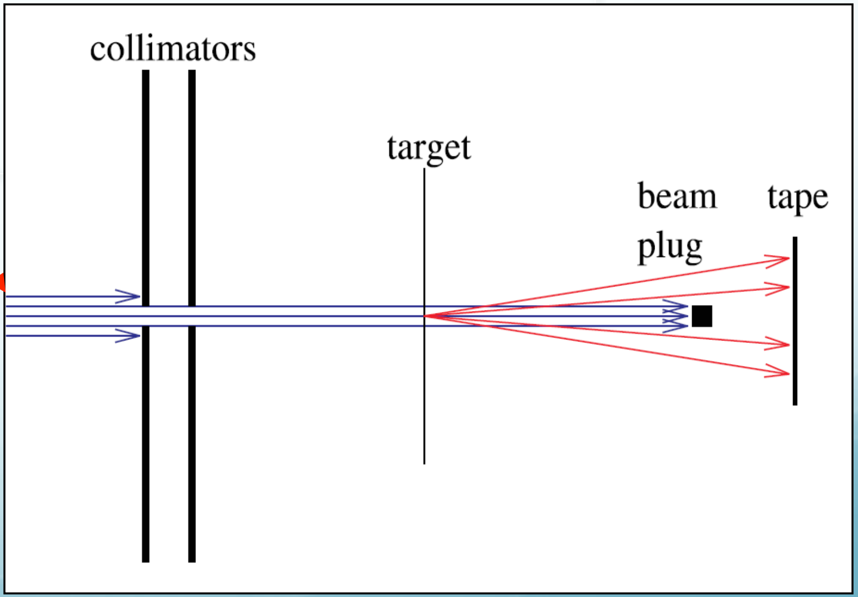
$$E_\gamma^0 = E_i - E_f$$

❖ Types of experiments performed at 9MV tandem accelerator



➢ Tape Station used for gamma-ray spectroscopy:

- Kapton tape transport station in closed loop
- Flexible time cycle control using in-house electronics
- 3x HPGe clovers (Eff ~ 1%) w/ BGO suppression
- Angular correlation table
- Digital data acquisition system



Target requirements

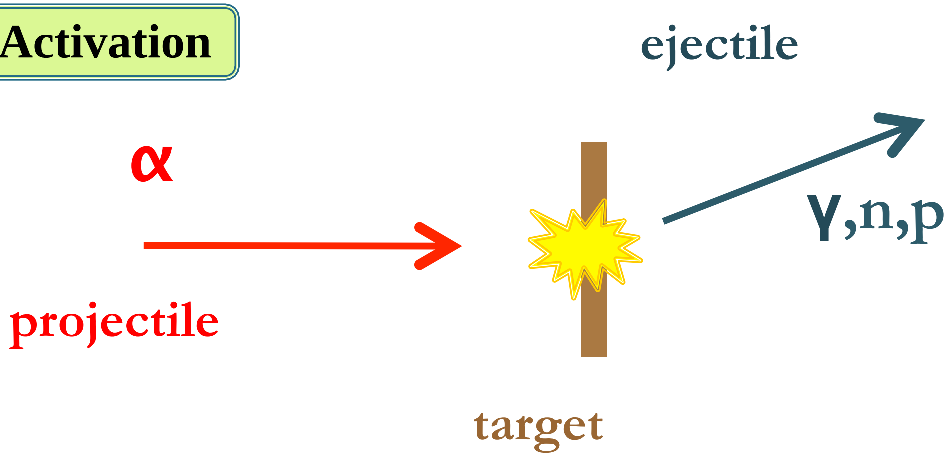
✓ *thin* (<2 mg/cm^2)

✓ *self-supported* targets that allow the recoil products to scatter onto the kapton tape

❖ Types of experiments performed at 9MV tandem accelerator

IFIN-HH

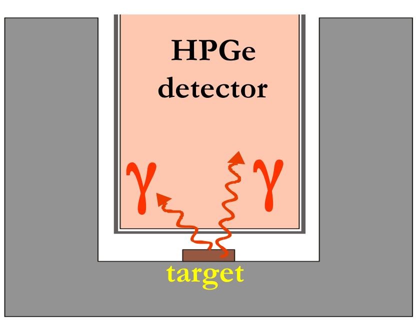
I. Activation



The activation technique consists in bombarding the target with α-particles to produce radioactive nuclei followed by the measurement of their specific activities once the irradiation has stopped.

II. Counting

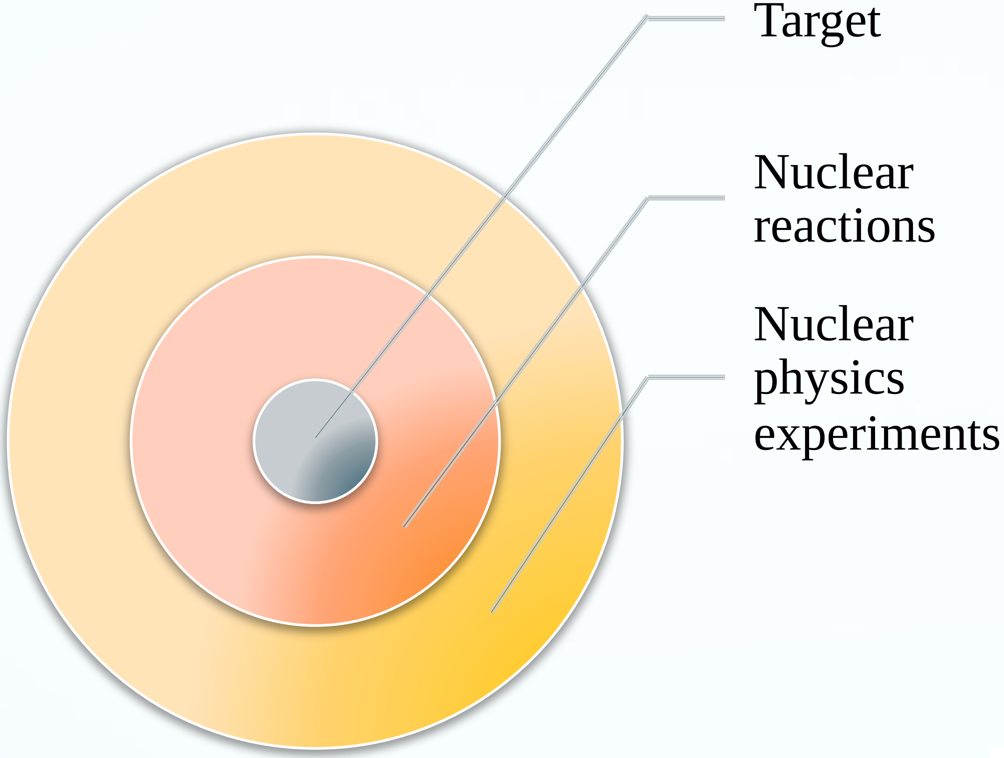
Counting setup in Bucharest, IFIN-HH, Romania



Pb walls and Cu and Al plates on the inside

- 2 HPGe detectors (relative efficiency of 55 % and 100%)
- Passive lead shielding
- Close detection geometry

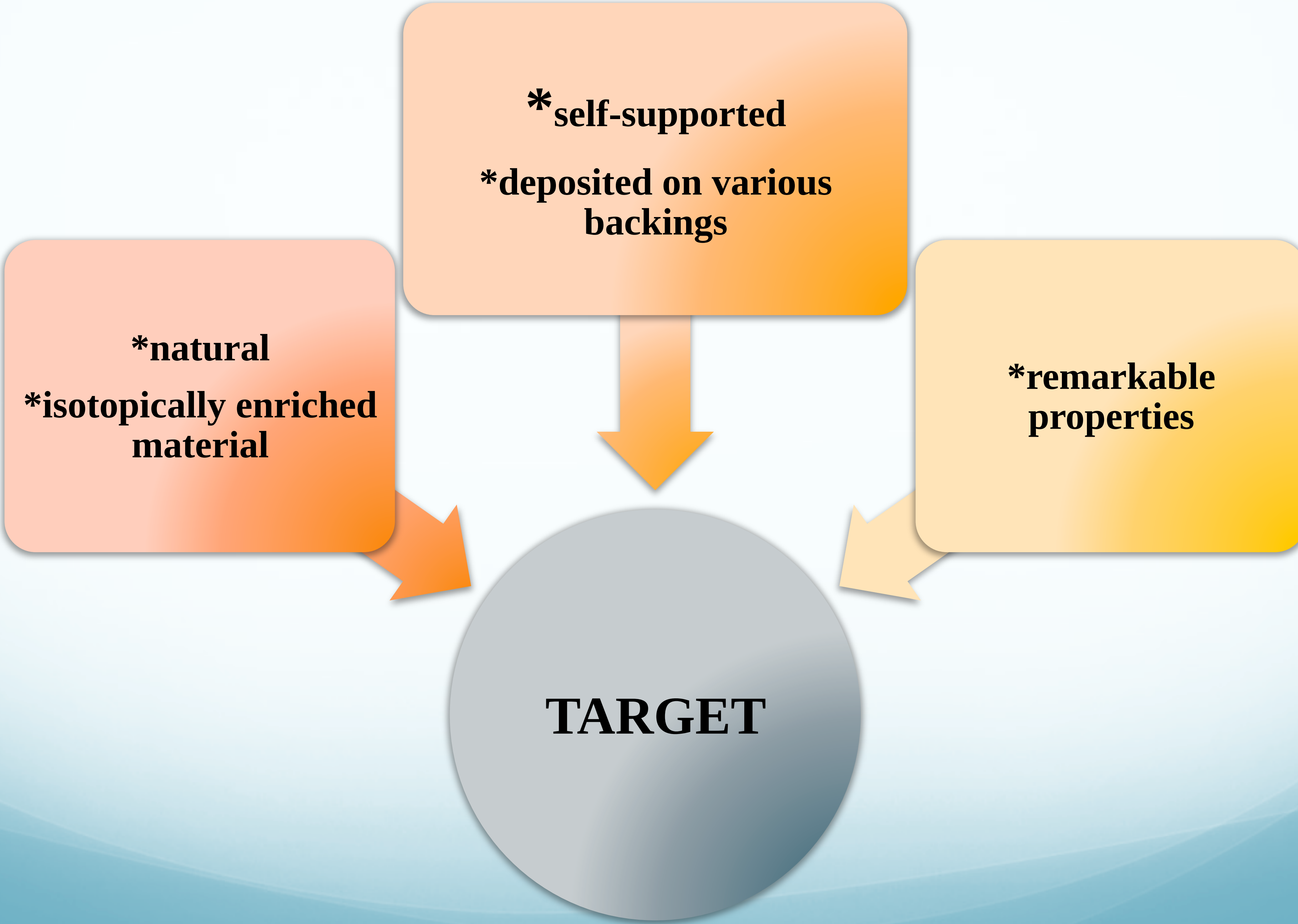
-> the summing corrections were performed using the Monte Carlo simulation code GESPECOR



No matter the type of experiment one wishes to perform, a good target is needed!

Target laboratory in IFIN-HH

- ***Evaporation techniques***
- ***Mechanical methods***

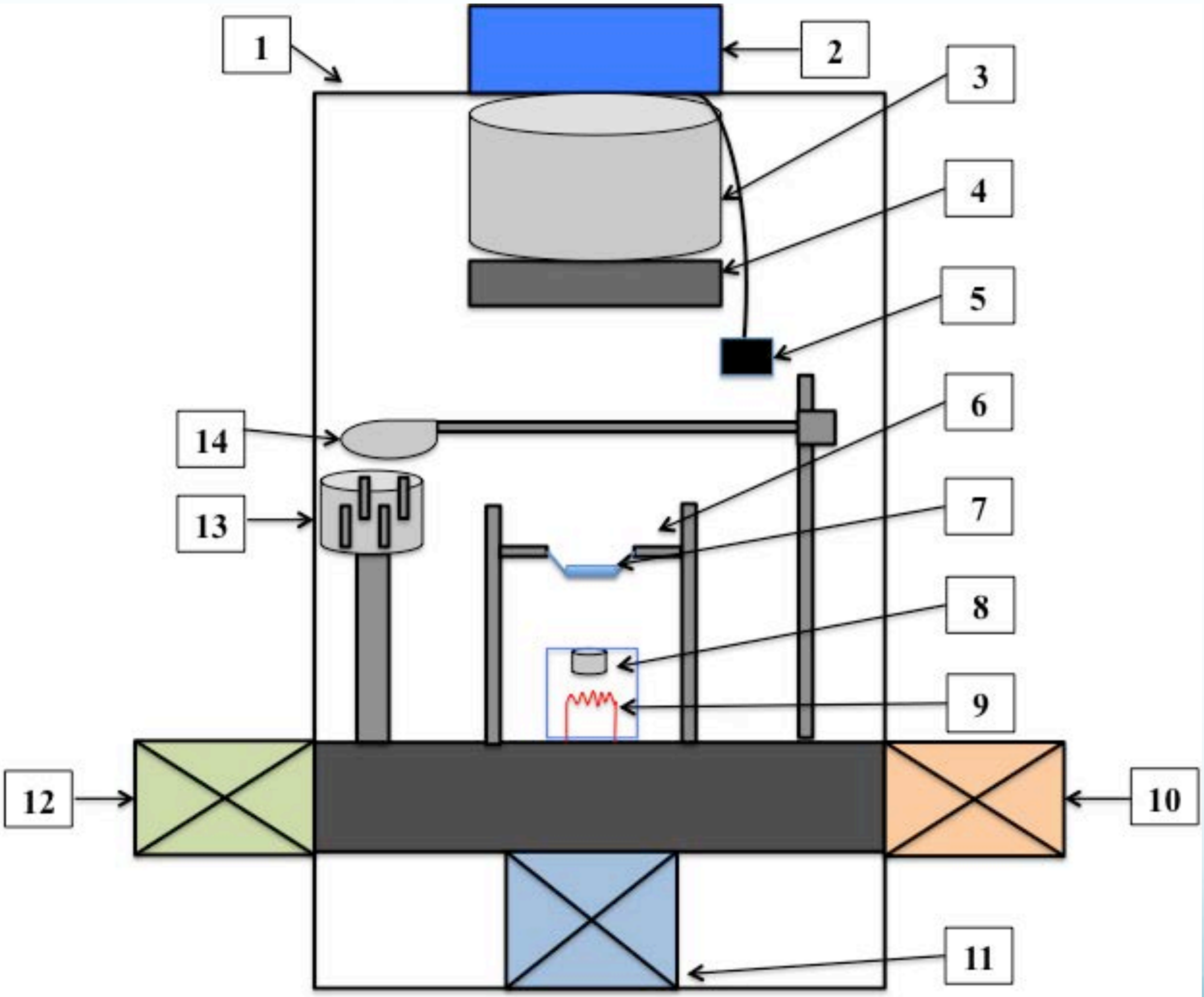

*self-supported
*deposited on various backings
*natural
*isotopically enriched material
*remarkable properties
TARGET

Equipment for target preparation

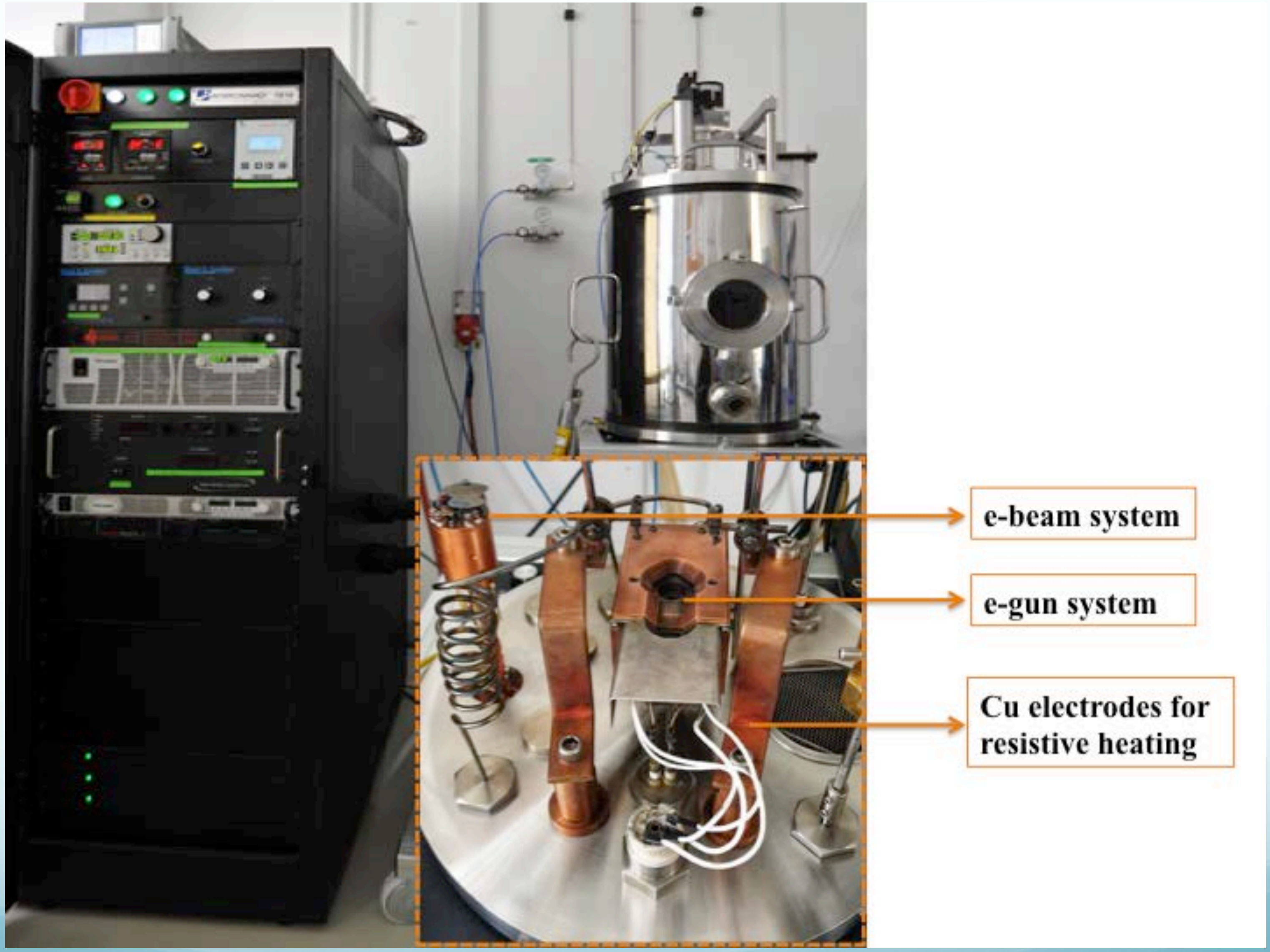
HIGH VACUUM DEPOSITION SYSTEM produced by Intercovamex Company

Vacuum chamber
- stainless steel
- cylindrical
- 45 cm diameter
- 53 cm high

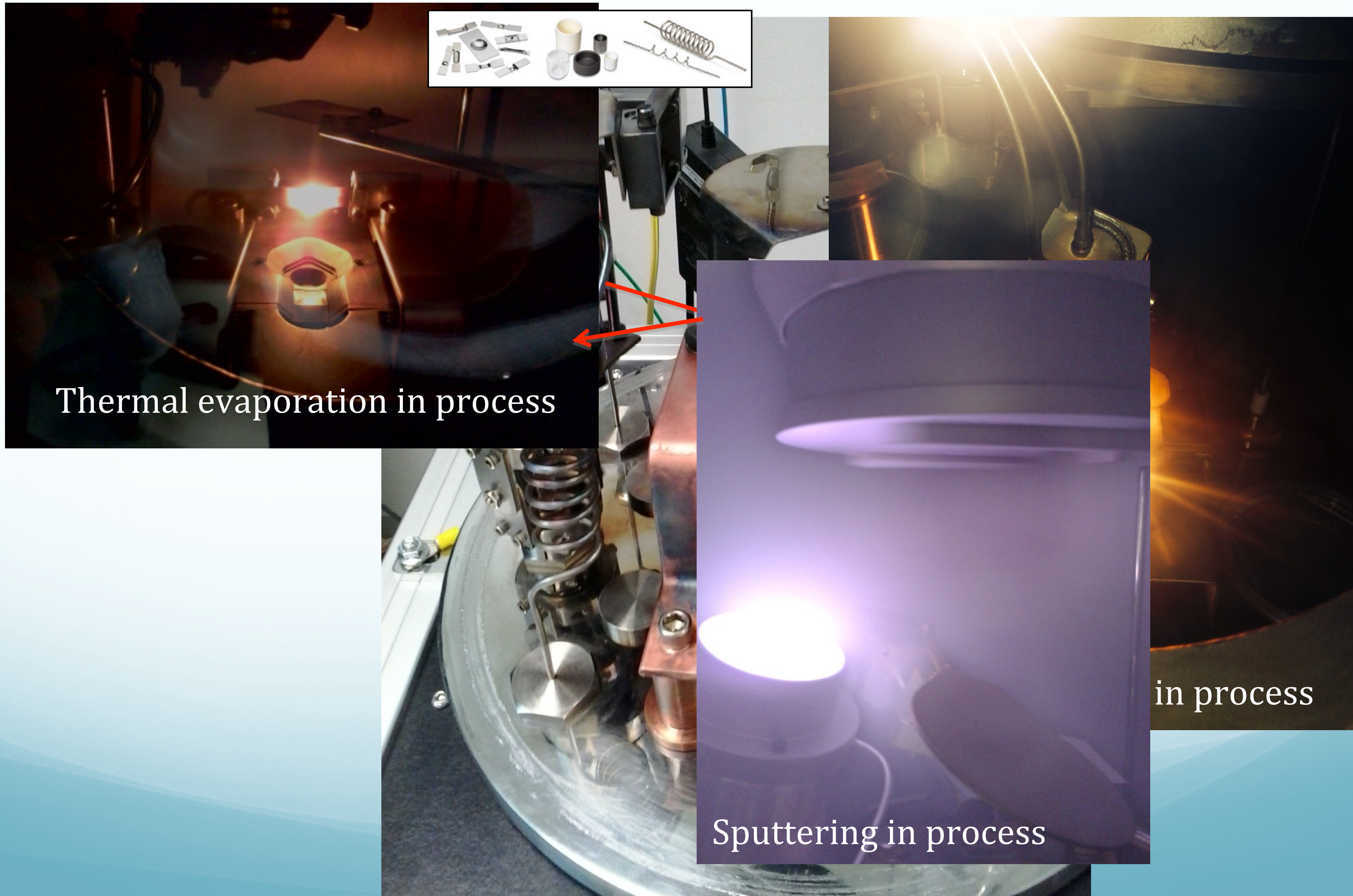
- used for creating self-supported, backed or sandwiched targets by:
  - **thermal evaporation**
  - **electron-gun evaporation**
  - **sputtering deposition methods**



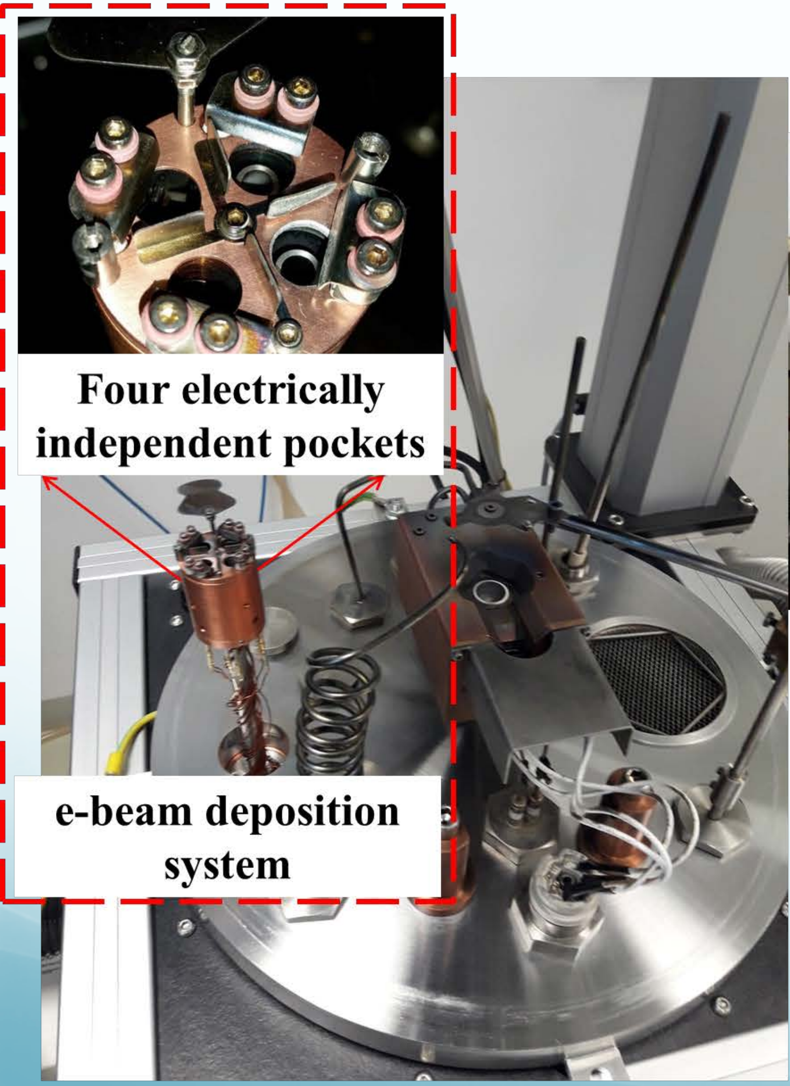
(1) vacuum chamber;
(2) water cooling system;
(3) substrate heater;
(4) substrate holder;
(5) quartz crystal monitor;
(6) copper electrodes;
(7) boat for resistive heating;
(8) e-gun crucible;
(9) W filament;
(10) oil diffusion pump;
(11) turbo molecular pump;
(12) rotary pump for preliminary vacuum;
(13) e-beam system with 4 pockets;
(14) shutter


e-beam system
e-gun system
Cu electrodes for
resistive heating

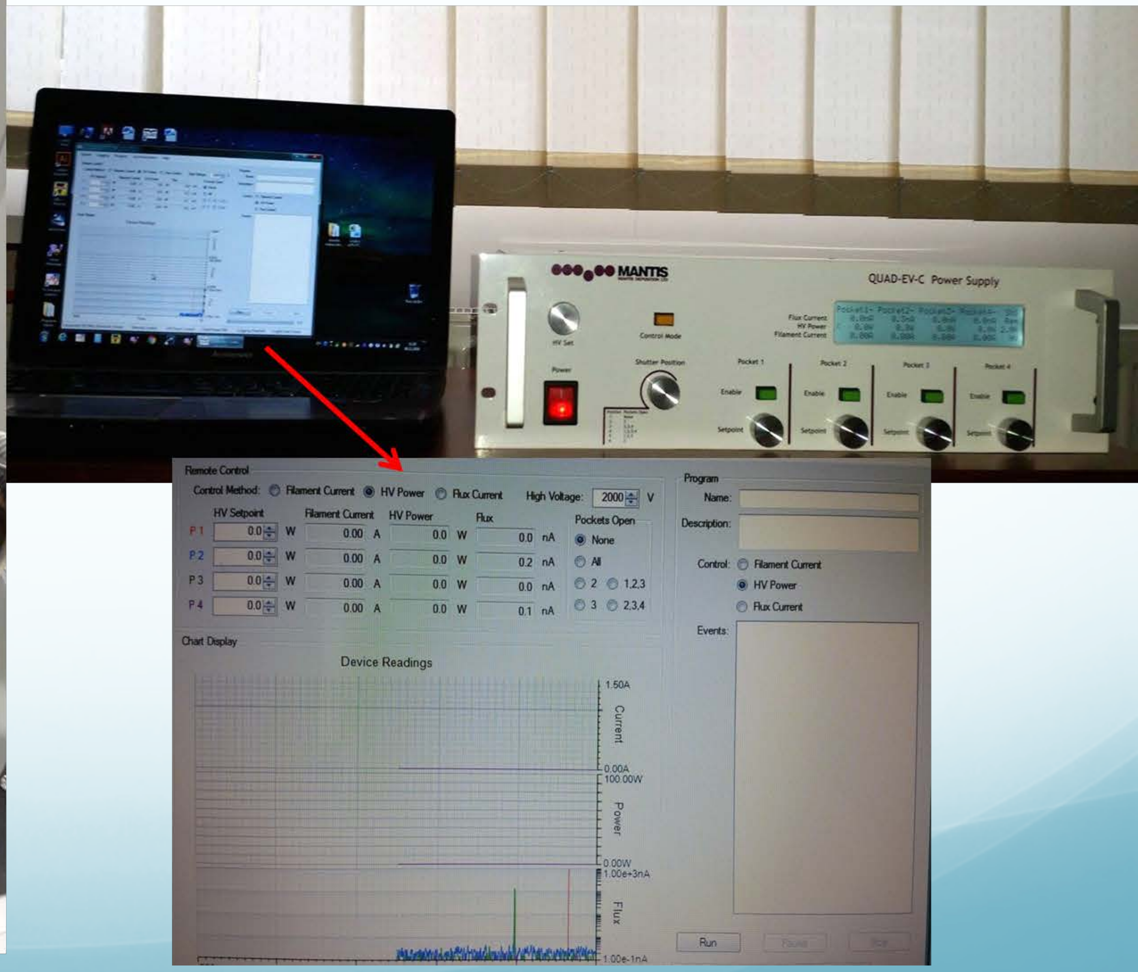
Evaporation techniques:



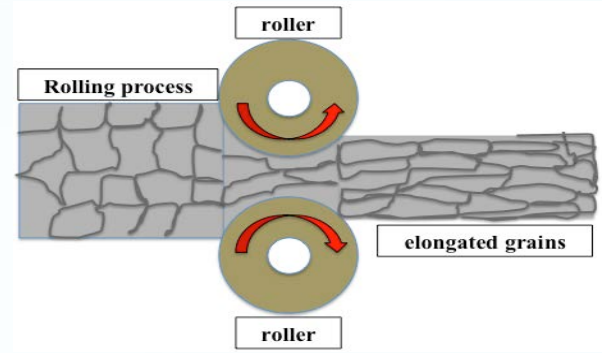
Mantis deposition system



PC-operated control software of power supplies



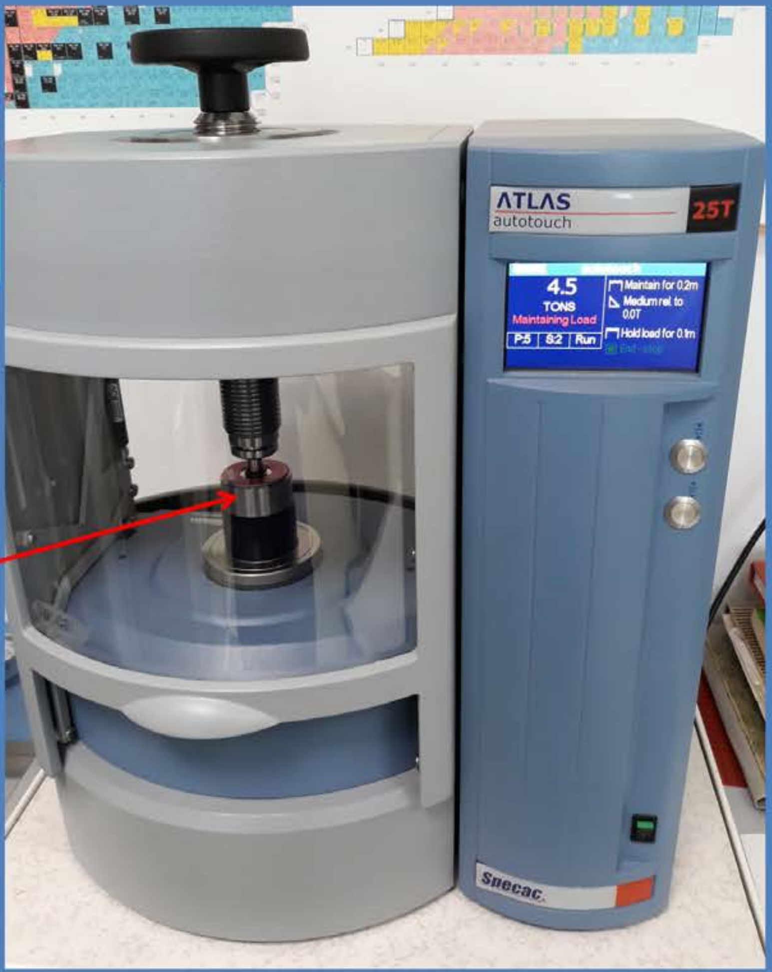
Mechanical methods



Rolling method: an extremely efficient method for self-supported metallic foil production using small amounts of starting material

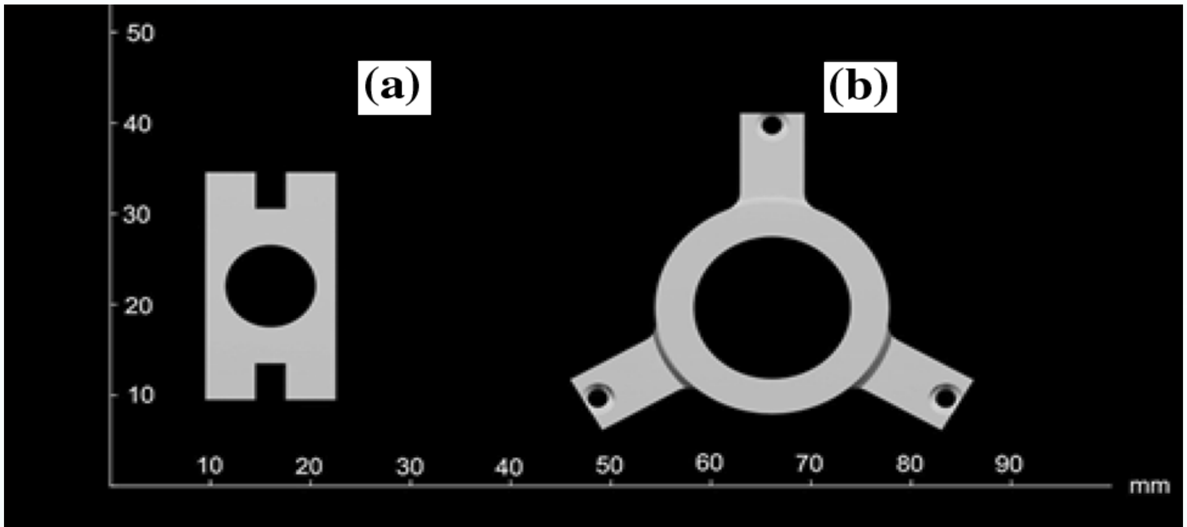
Hydraulic press mainly for producing pellets which can be used in some types of experiments as thick targets

EVACUABLE PELLET DIES



TARGET CLASSIFICATION

I. TARGET FRAME

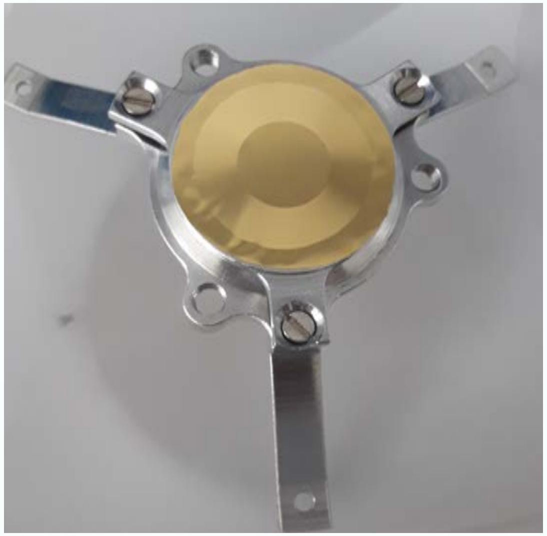


- **surface**
- **shape**

(a) regular frame for thin/thick targets for nuclear structure experiments

(b) frame for Recoil Distance Doppler Shift Method (Plunger)

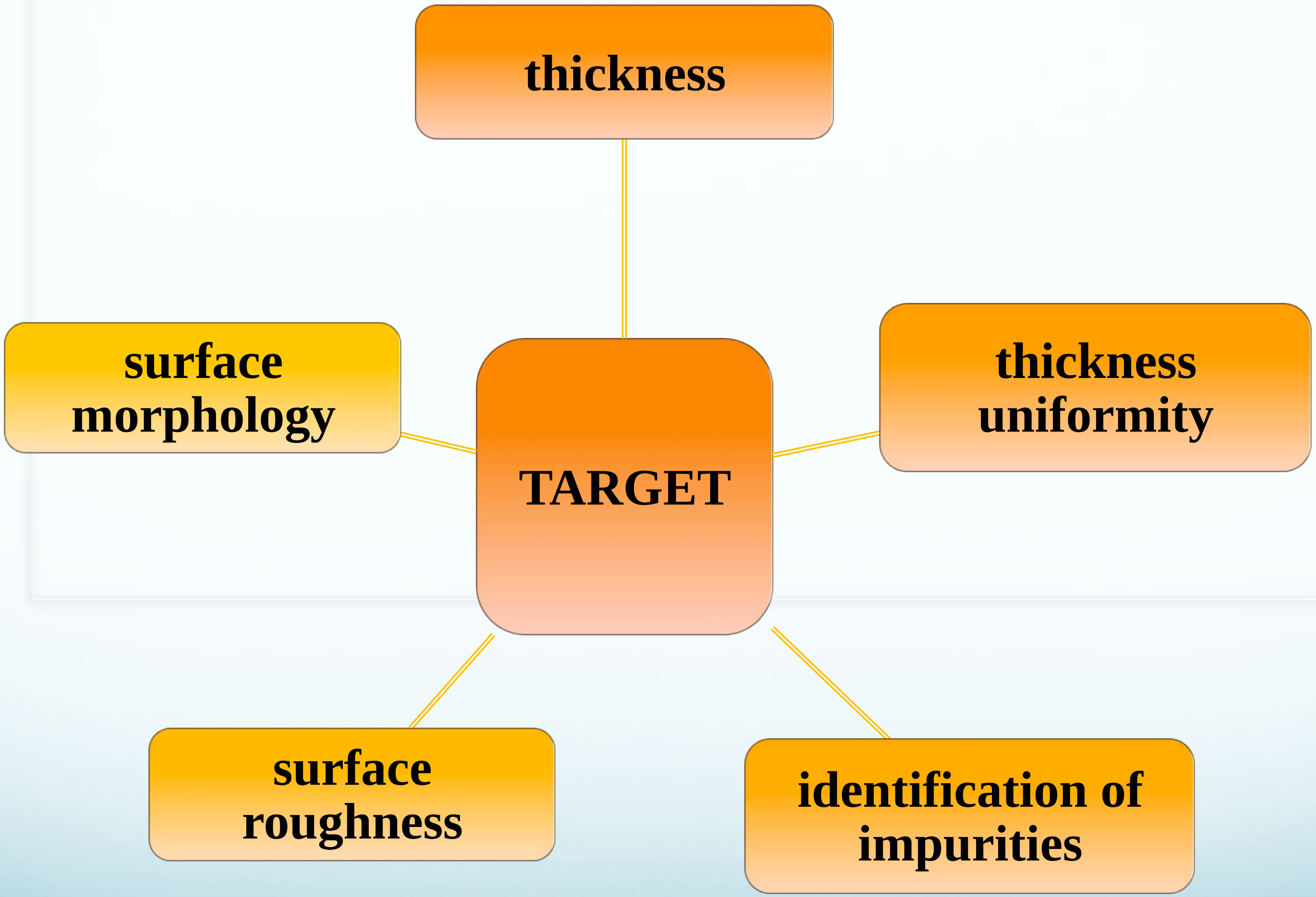
II. BACKING or SELF-SUPPORTED



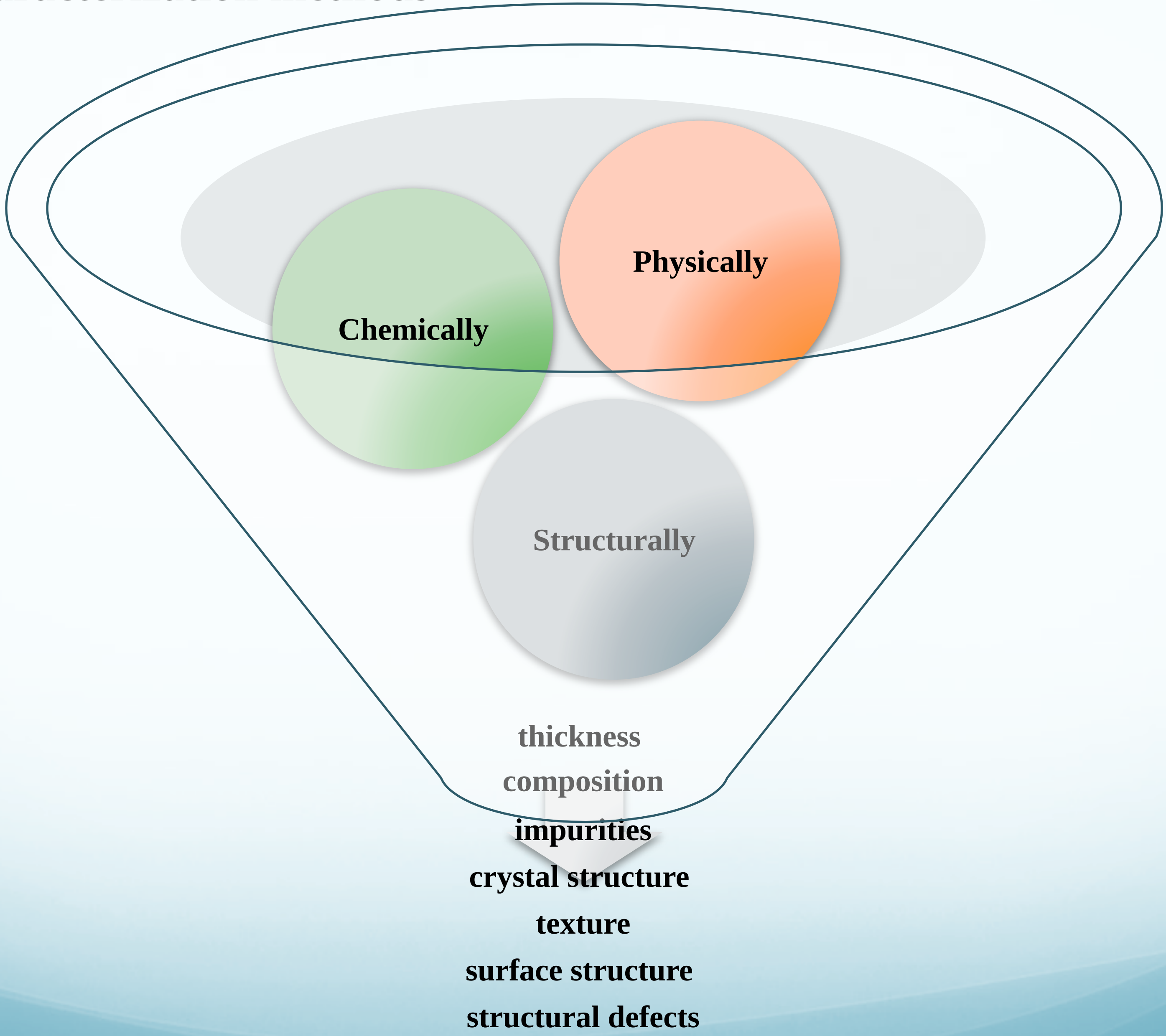
How to choose the preparation method?

- target characteristics (thickness, surface, shape)
- material cost and availability (metallic form, compound)
- availability of the method in the target preparation laboratory
- method effectiveness
- avoiding contamination of the target

Target characteristics



Target characterization methods



Target characterization

-morphology, composition, structure and thickness determination-

AFM *(Atomic Force Microscopy)*

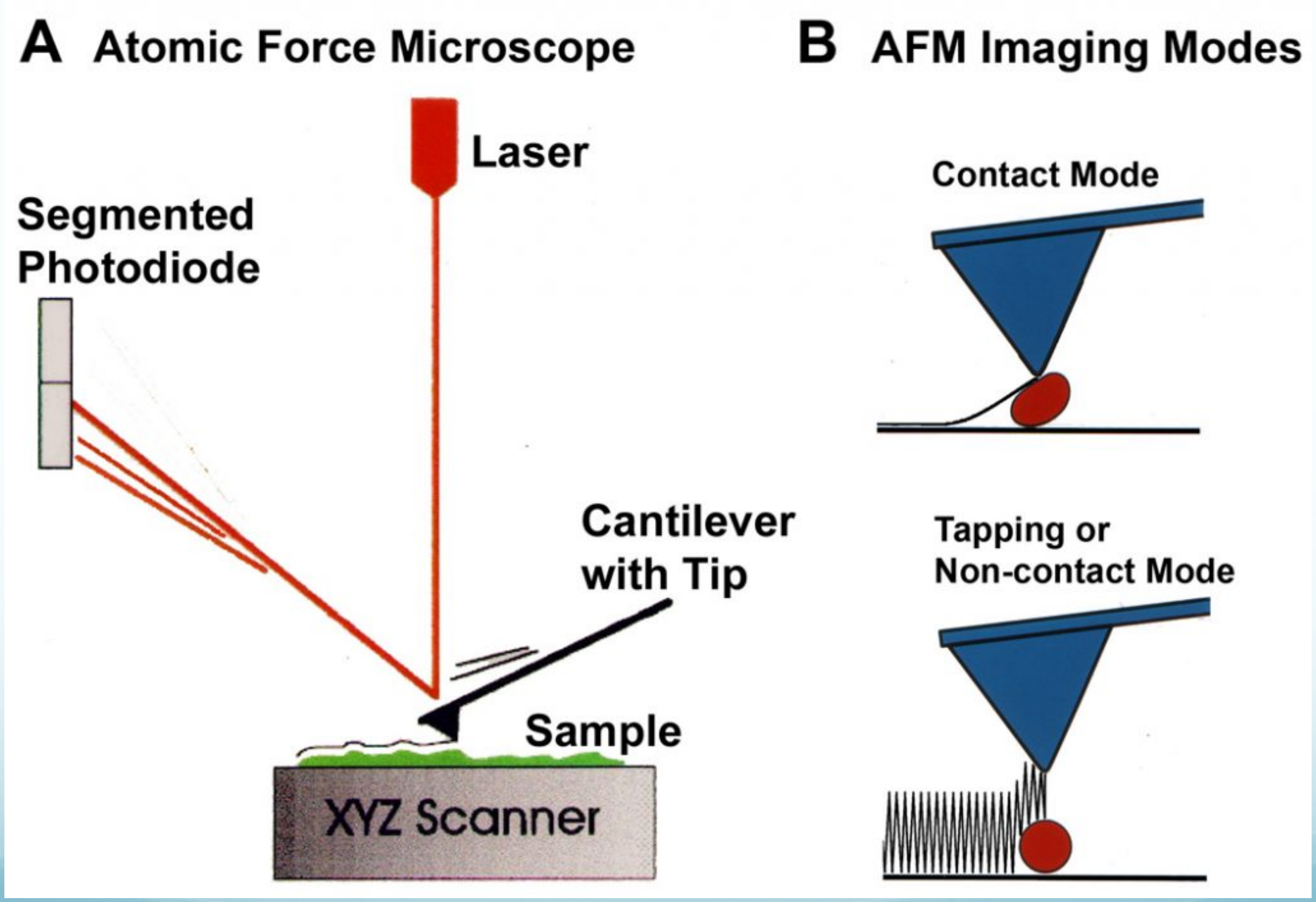
SEM/EDX *(Scanning Electron Microscopy/Energy Dispersive X-ray Spectroscopy)*

XRD *(X-Ray Diffraction)*

RBS *(Rutherford Back Scattering)*

Atomic Force Microscopy (AFM)

- ***surface topography***
- ***surface roughness***
- ***thickness***

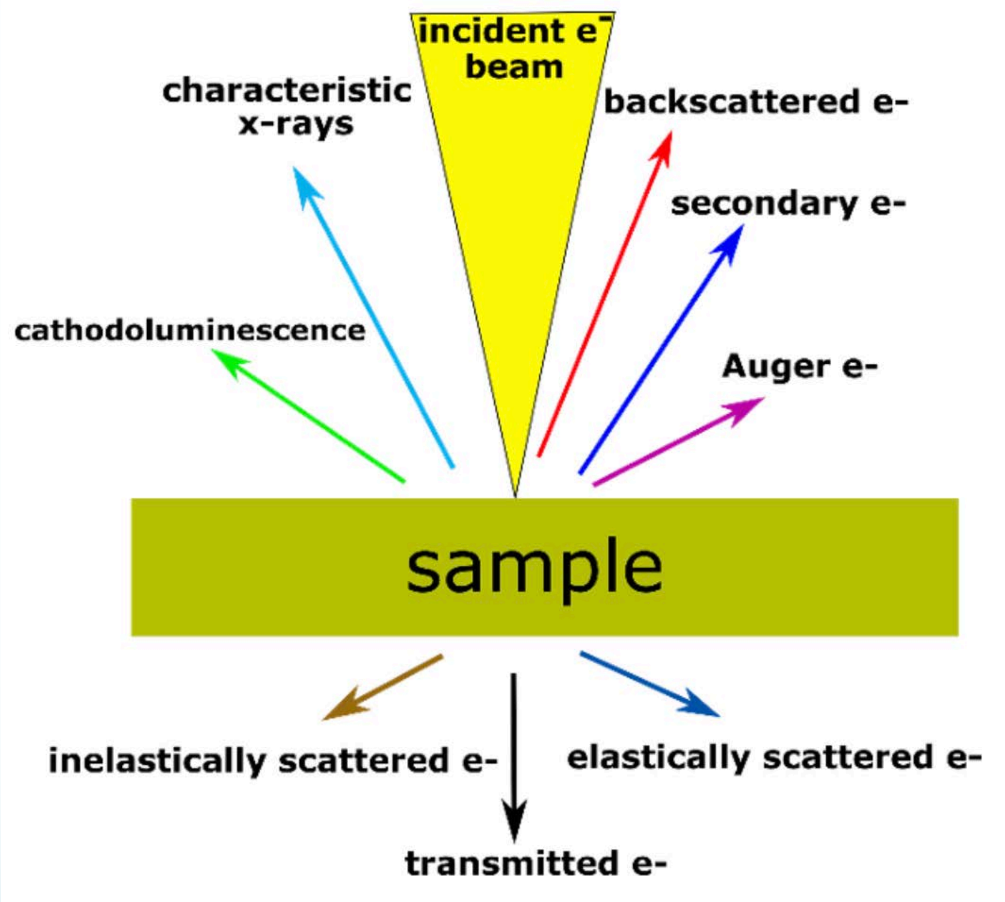


Scanning Electron Microscopy/ Energy Dispersive X-ray (SEM/EDX)

based on the electron-matter interaction, which generates a large variety of signals

- **qualitative-** the atomic number of the element within the analyzed volume

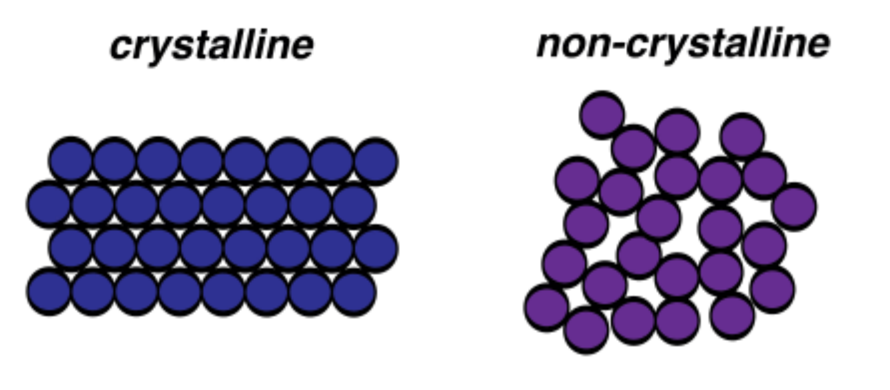
- **quantitative-** the concentration percentage of each element present in the sample

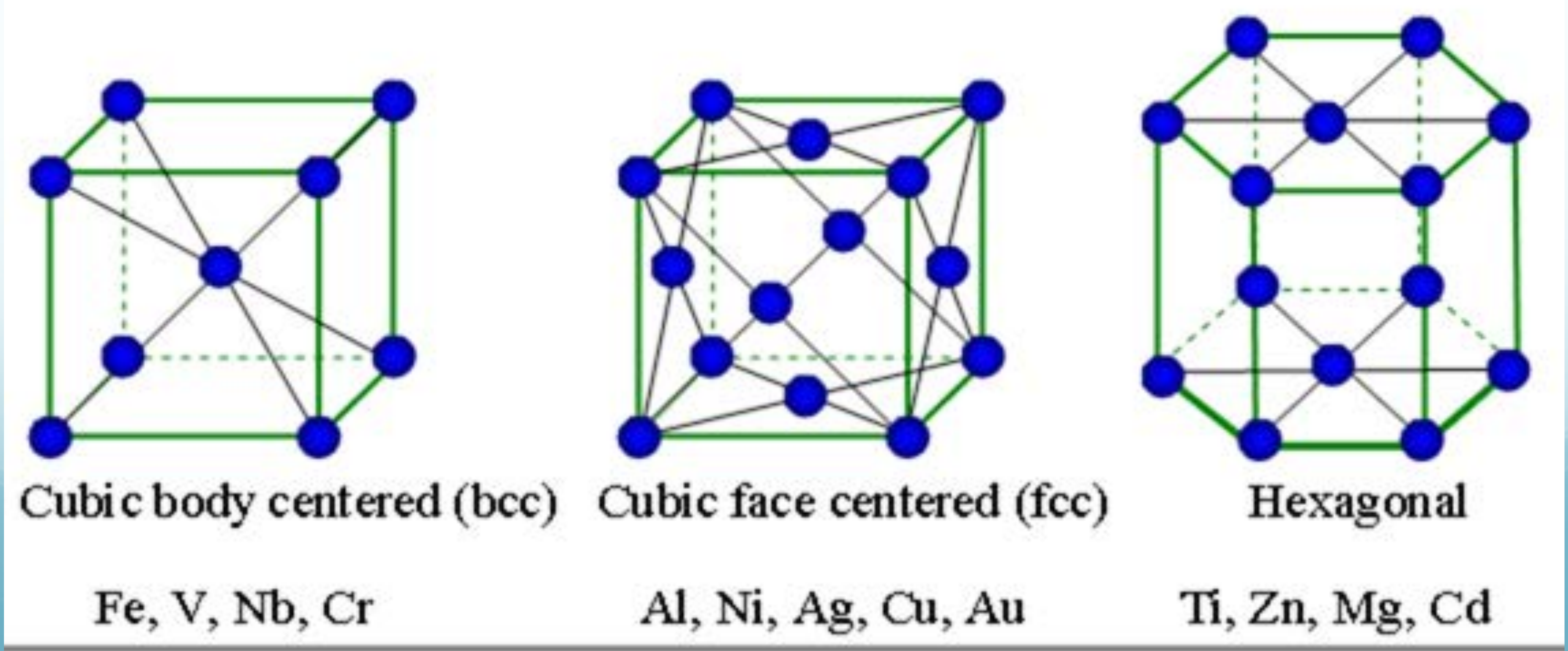


EDX system allows obtaining X-ray elemental maps bringing information on the distribution of the elements within the analyzed sample and surface uniformity.

X-ray Diffraction (XRD)

- *material crystallinity (the arrangement of atoms in a crystal)*
- *chemical composition*
- *purity*
- *stoichiometry*





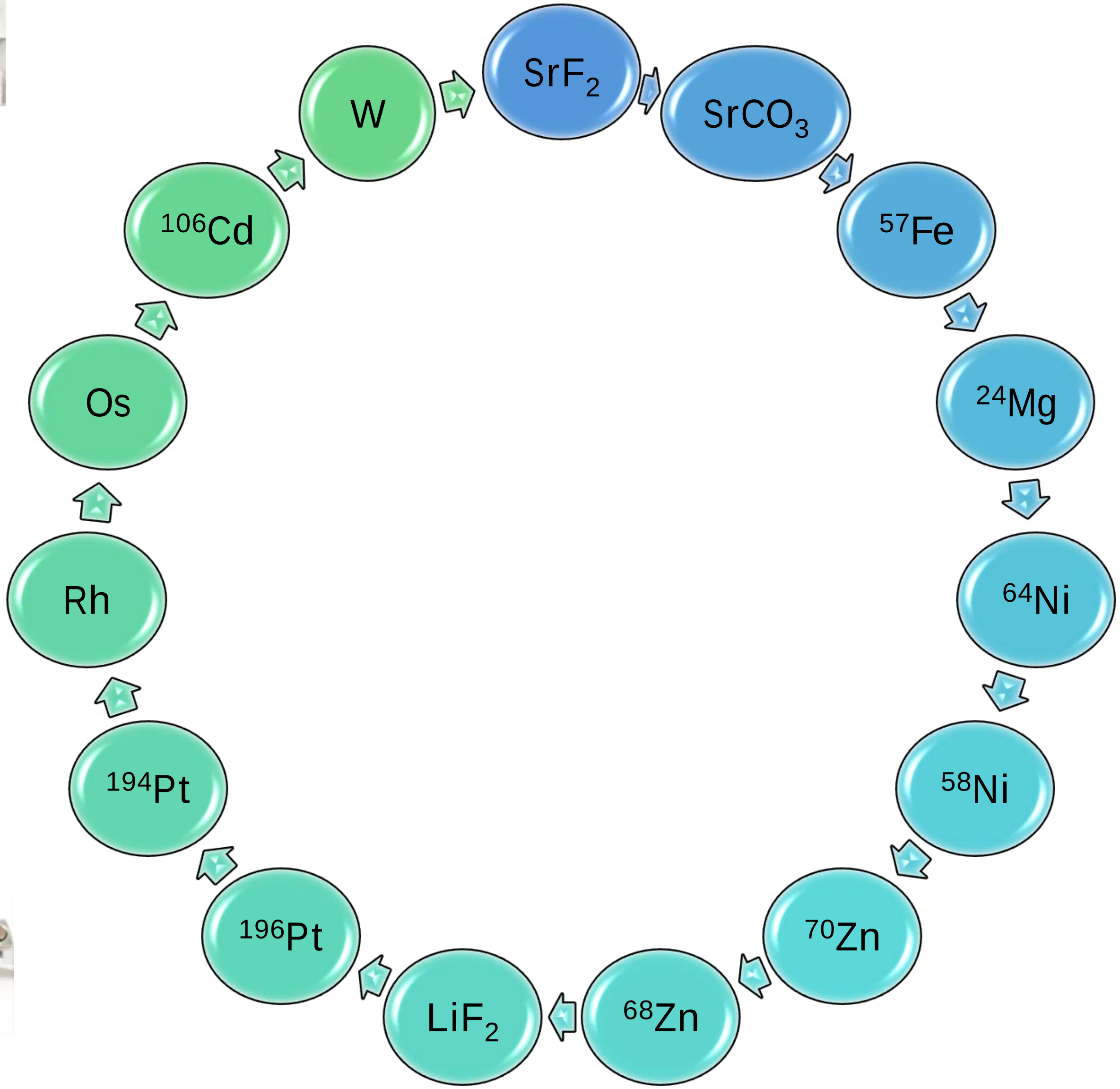
Thickness determination methods- one of the most important target parameters, which can be measured during the deposition process (in situ monitoring) or after it (outside the deposition chamber)

- **Quartz crystal monitor-** quartz crystal is placed in the vacuum chamber, usually close to the substrate holder
- **Rutherford Backscattering Spectrometry (RBS)-** a non-destructive nuclear analytical method. It implies:
  - the use of a rather simple scattering geometry
  - the use of a beam composed of light nuclei
  - a particle detector placed at backwards angles
- **Weighting method-** very important as a primary technique in thickness determination of foils or deposited targets, combined with the precise surface measuring using an optical microscope

Mettler Toledo XP56/ M analytical balance

- **Alpha transmission**

Targets


SrF_2
$SrCO_3$
^{57}Fe
^{24}Mg
^{64}Ni
^{58}Ni
^{70}Zn
^{68}Zn
LiF_2
^{196}Pt
^{194}Pt
Rh
Os
^{106}Cd
W

Specific examples and particularities

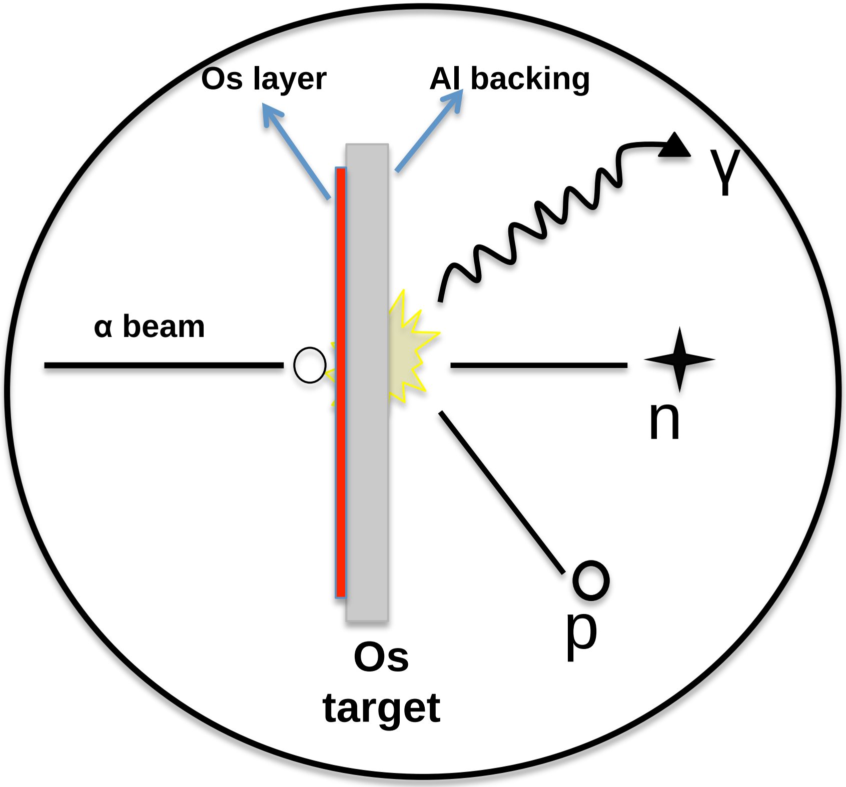
I. Preparation and characterization of thick osmium targets

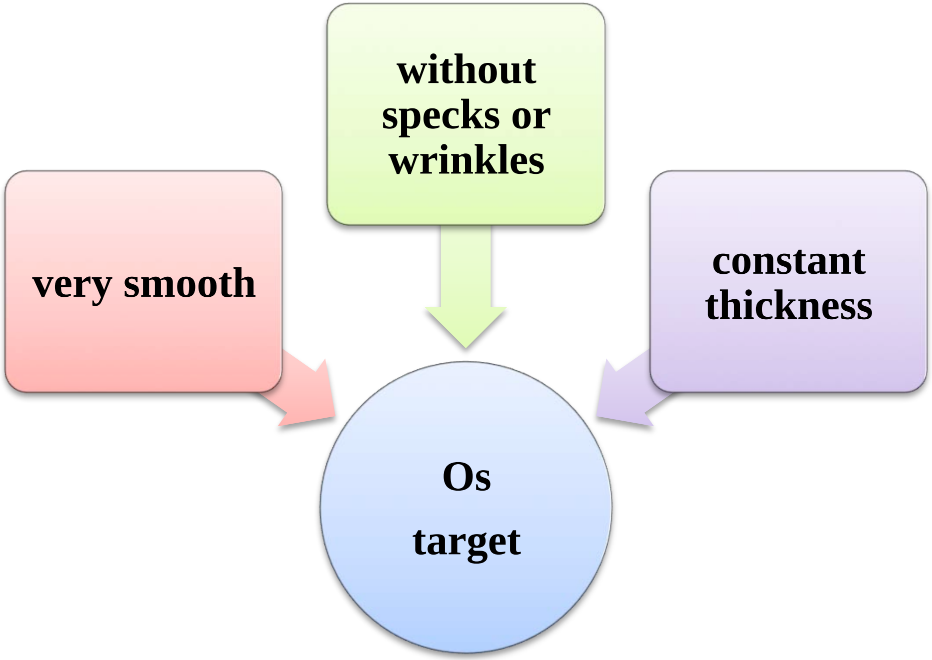
II. Strontium targets for nuclear astrophysics experiments

III. Decontamination of ^{64}Ni targets

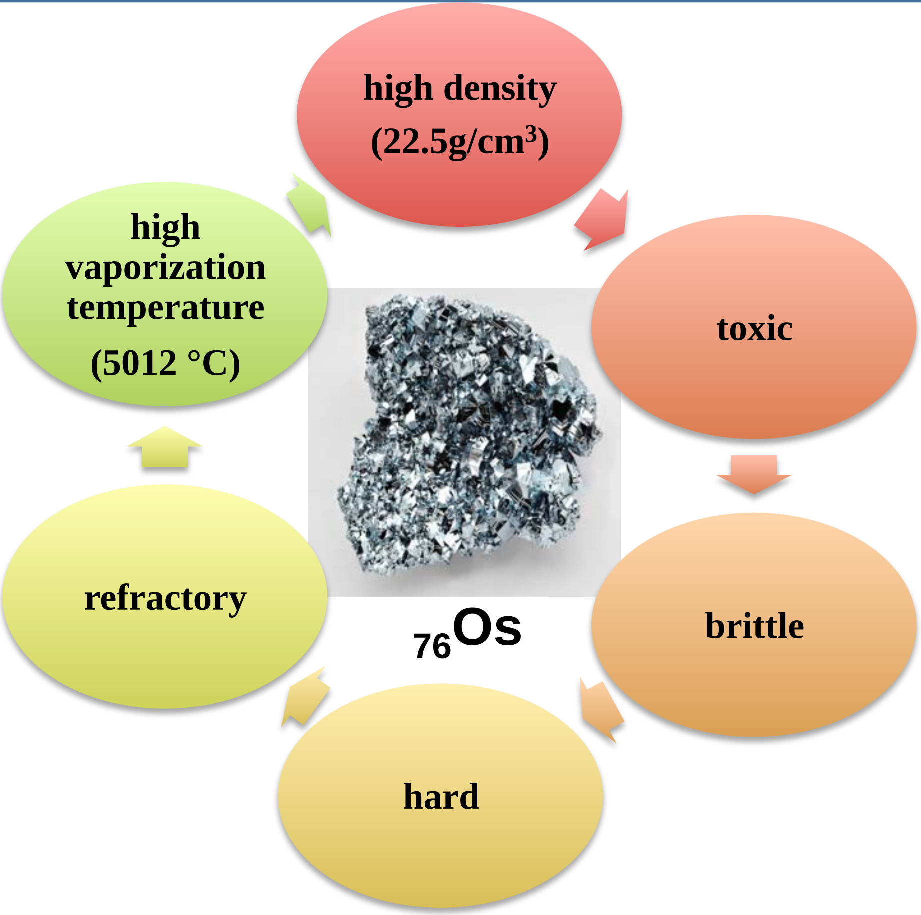
Preparation and characterization of thick osmium targets

- ✓ preparation and characterization of Os targets for nuclear astrophysics experiments with alpha particles





Physical properties

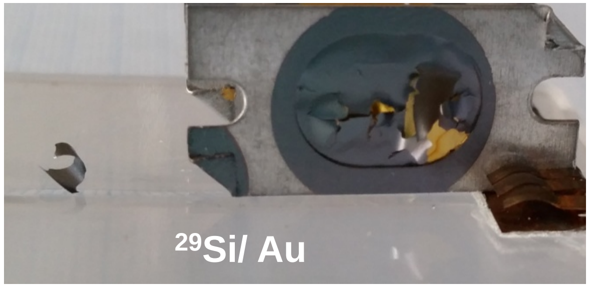


Because of its
[hardness, brittleness, low vapor and very high melting point (3033 °C, the fourth highest of all elements, after only C, W, and Re), solid osmium is difficult to machine, form or work.]

Limitations of the classical evaporation methods

- given by the material properties -

- **vaporization temperature:** the above-mentioned evaporation techniques allow preparation of well-controlled, good quality thin films from materials with high vaporization temperatures (~ 3000°C), but there is an increased demand for thin films from materials with extremely high vaporization temperatures in the range of 4500-5500°C (e.g. Os, Hf, W);



- **low adhesion of some materials;**

- **the evaporation is not done directed on the substrate**

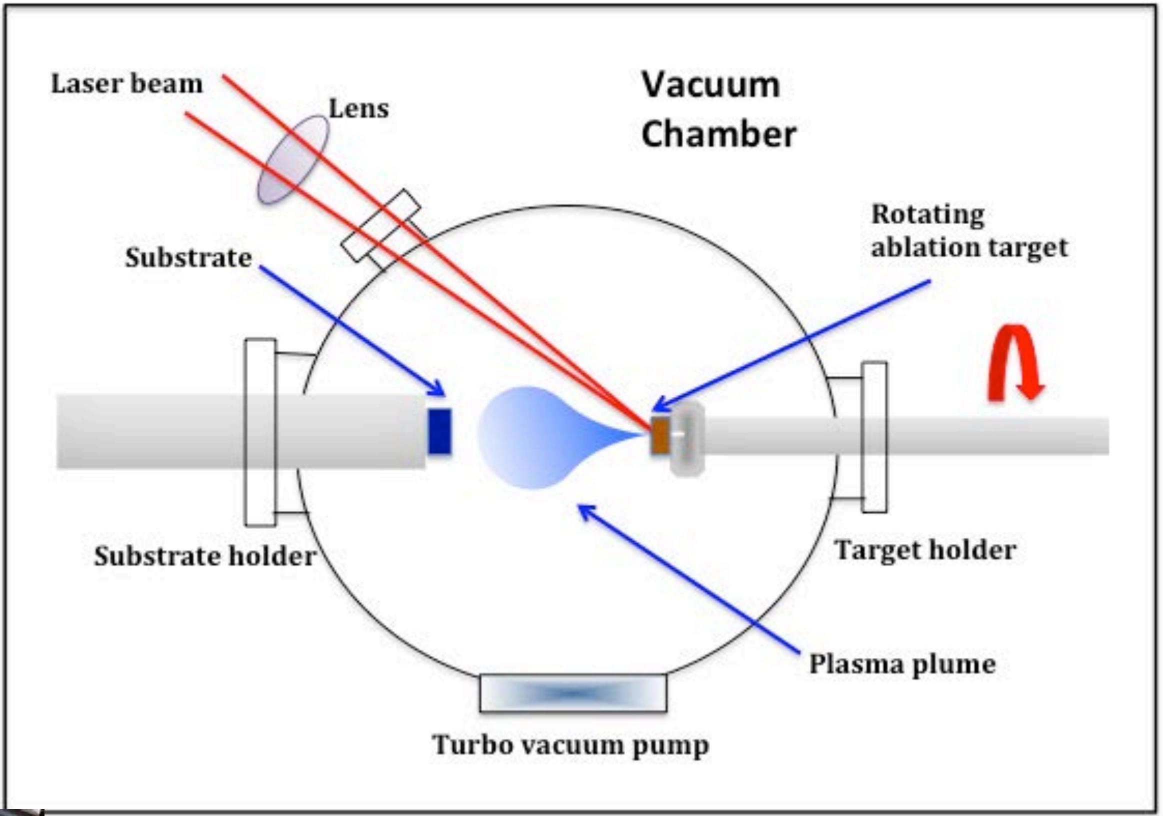
Target preparation method

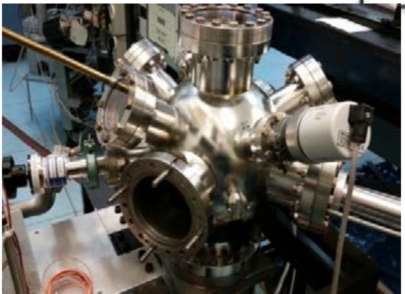
PULSED LASER DEPOSITION (PLD)

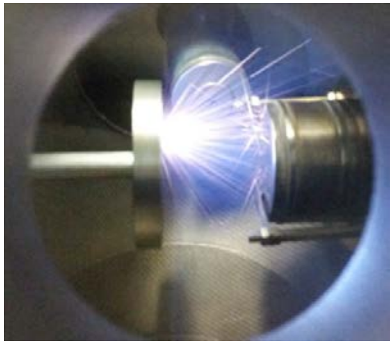
Developed in collaboration with INFLPR

National Institute for Laser, Plasma and Radiation Physics

(Bucharest-Magurele, Romania)







The advantages of PLD method:

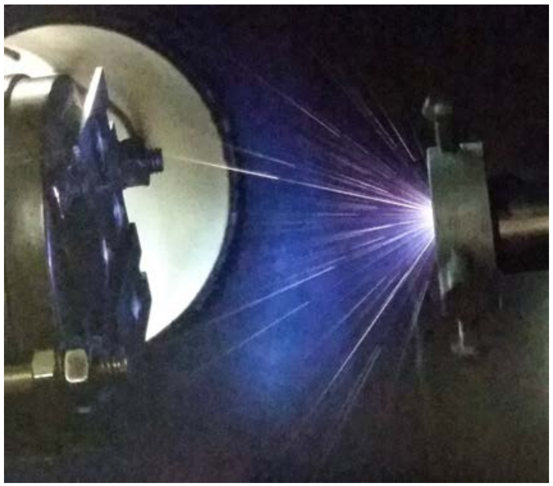
- It allows deposition of chemical elements, compounds and some polymers;

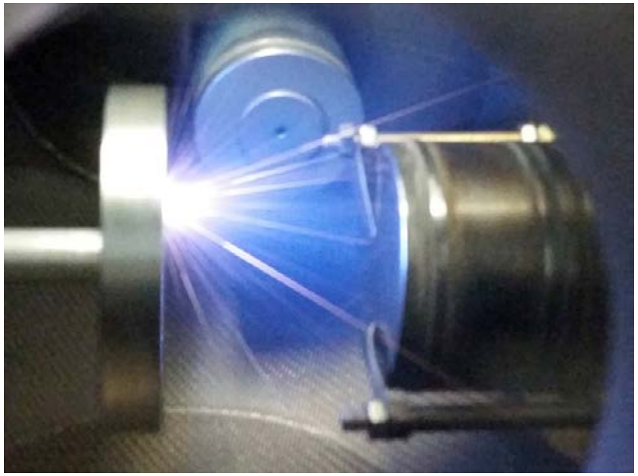
- It allows obtaining thin layers of better quality compared to other methods:

- ✓ **high density**
- ✓ **thickness uniformity on the defined surface**
- ✓ **controllable thickness**
- ✓ **high purity**
- ✓ **durability**

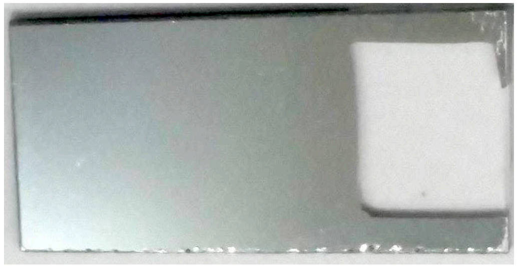
Os targets

Nd:YAG Laser: 1064 nm (IR range)





Os [2000nm] / Al [20um]

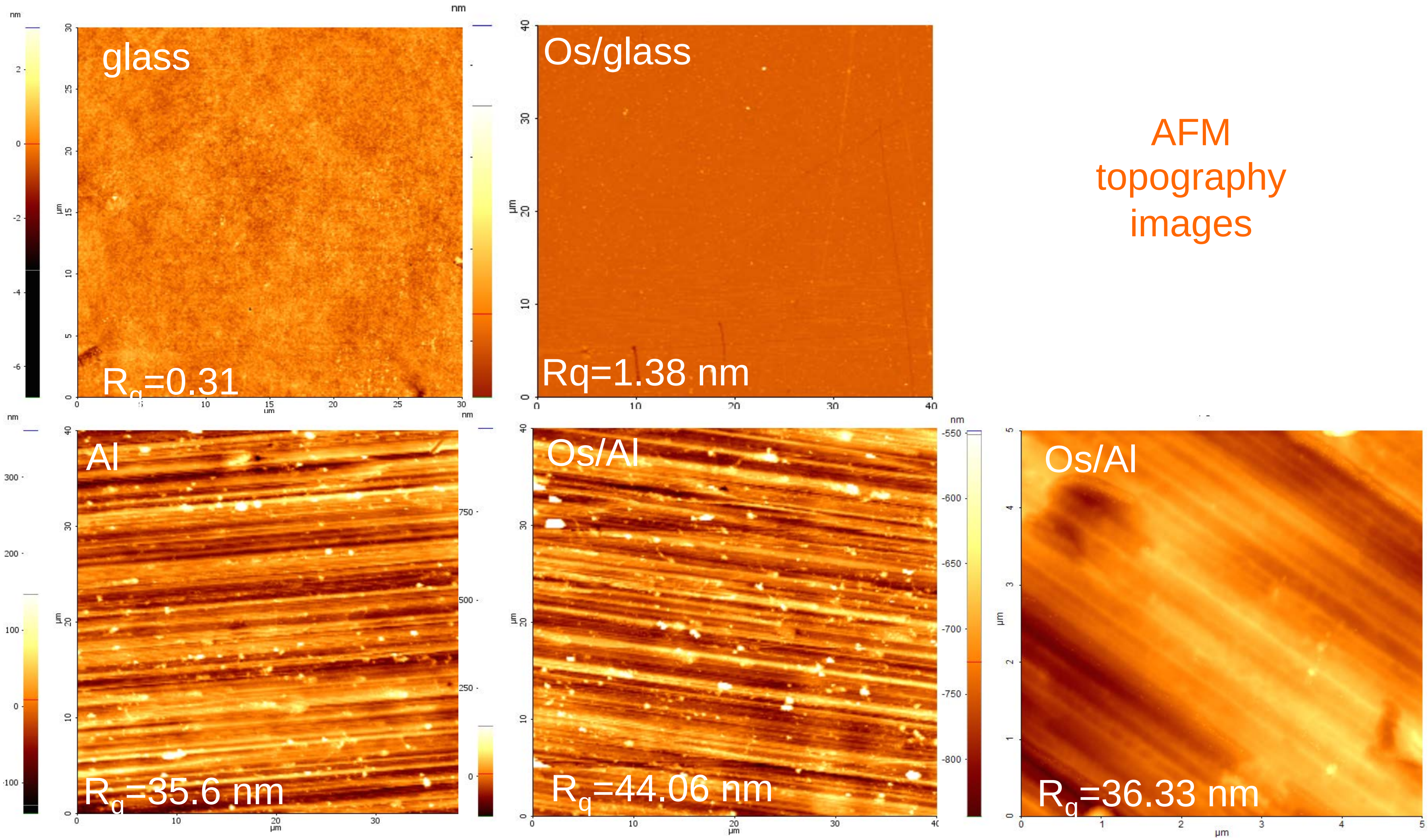


Os [100nm] / glass (test)

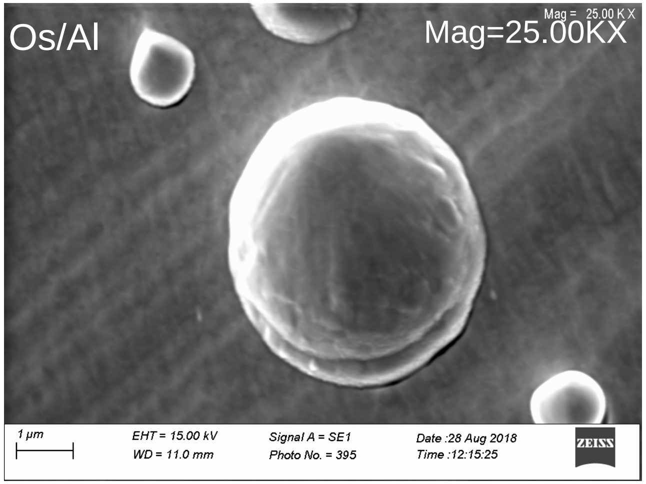
Morphological features

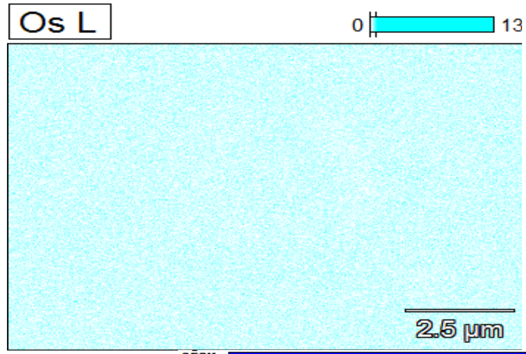
Backing	Short deposition (5 h)	Long deposition (30 h)

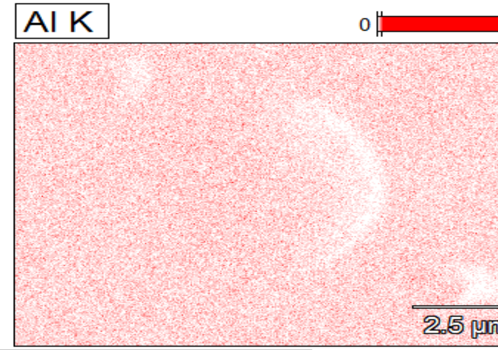
AFM topography images

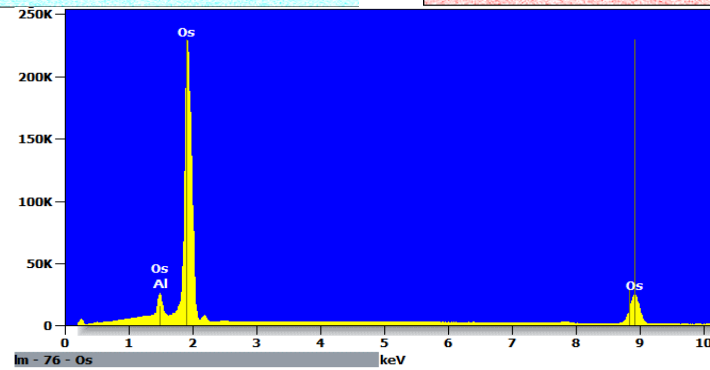


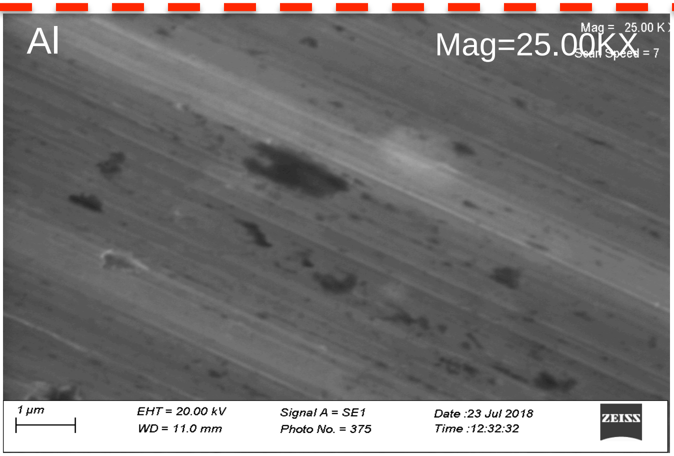
SEM and EDX Os/Al mapping micrographs

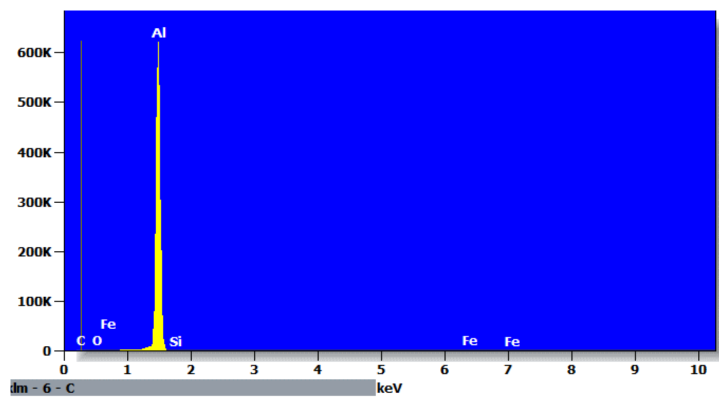




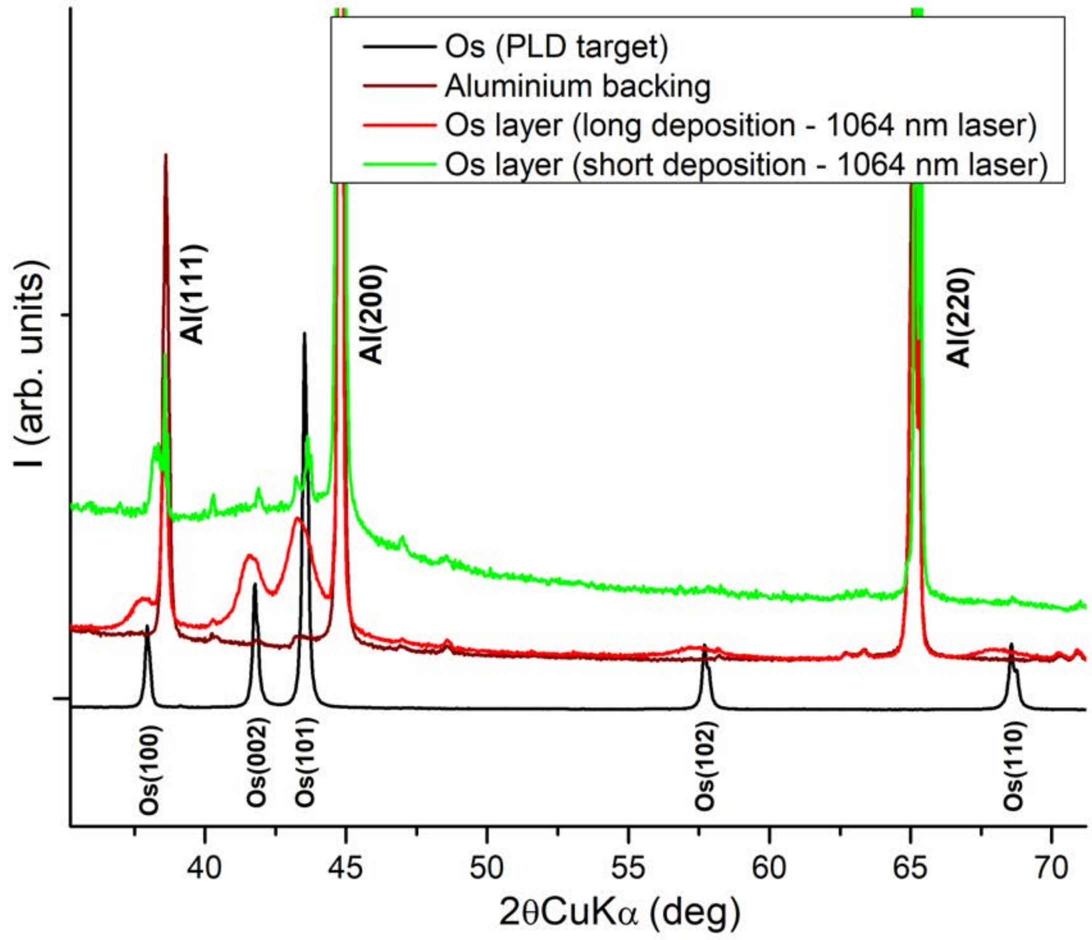








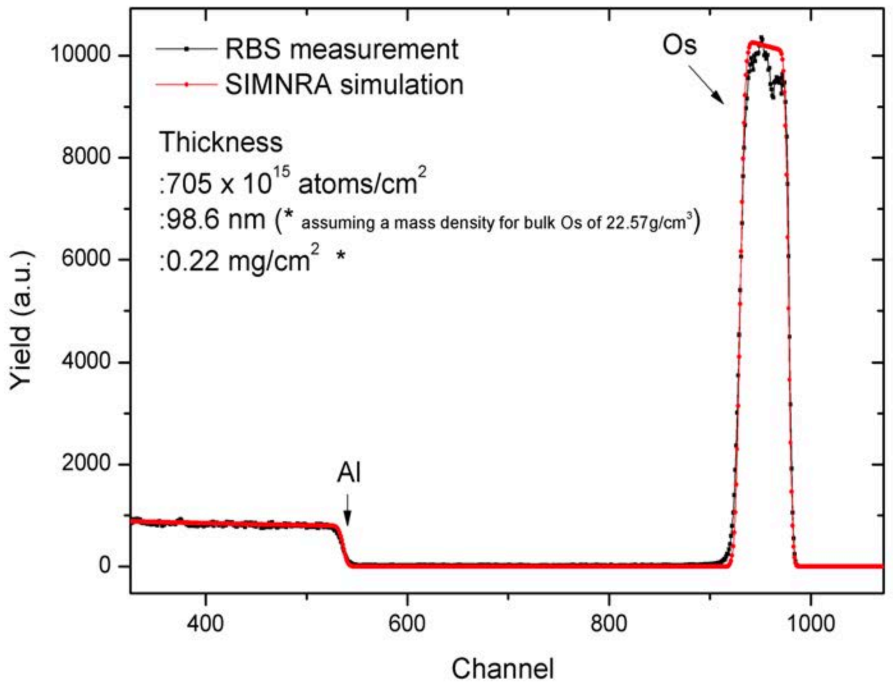
XRD spectra

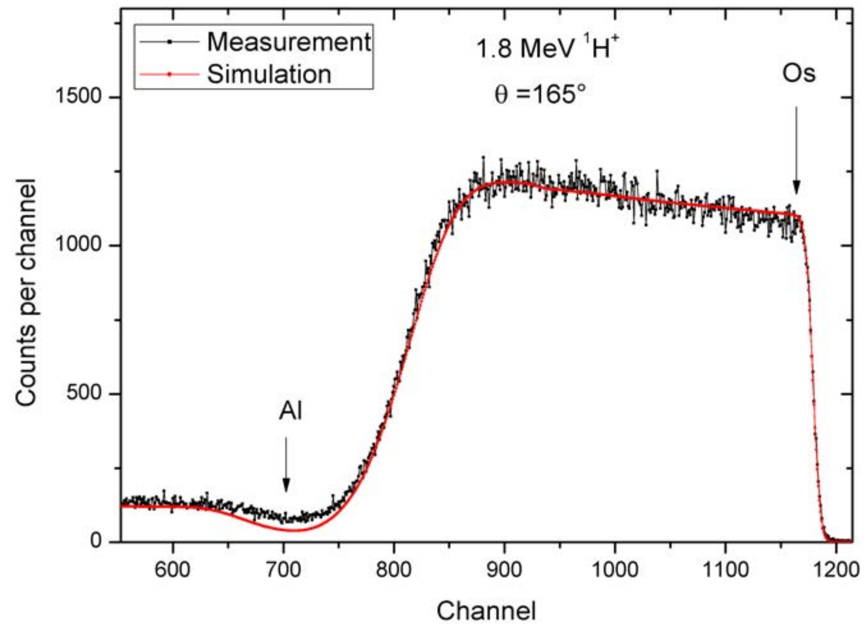


Os (PLD target)
Aluminium backing
Os layer (long deposition - 1064 nm laser)
Os layer (short deposition - 1064 nm laser)
I (arb. units)
Al(111)
Al(200)
Al(220)
Os(100)
Os(002)
Os(101)
Os(102)
Os(110)
40
45
50
55
60
65
70
2θCuKα (deg)

RBS spectra of Os deposited on Al, measured by the RBS fixed detector at 165°

It implies - the use of a rather simple scattering geometry
- the use of a beam composed of light nuclei
- a particle detector placed at backwards angles



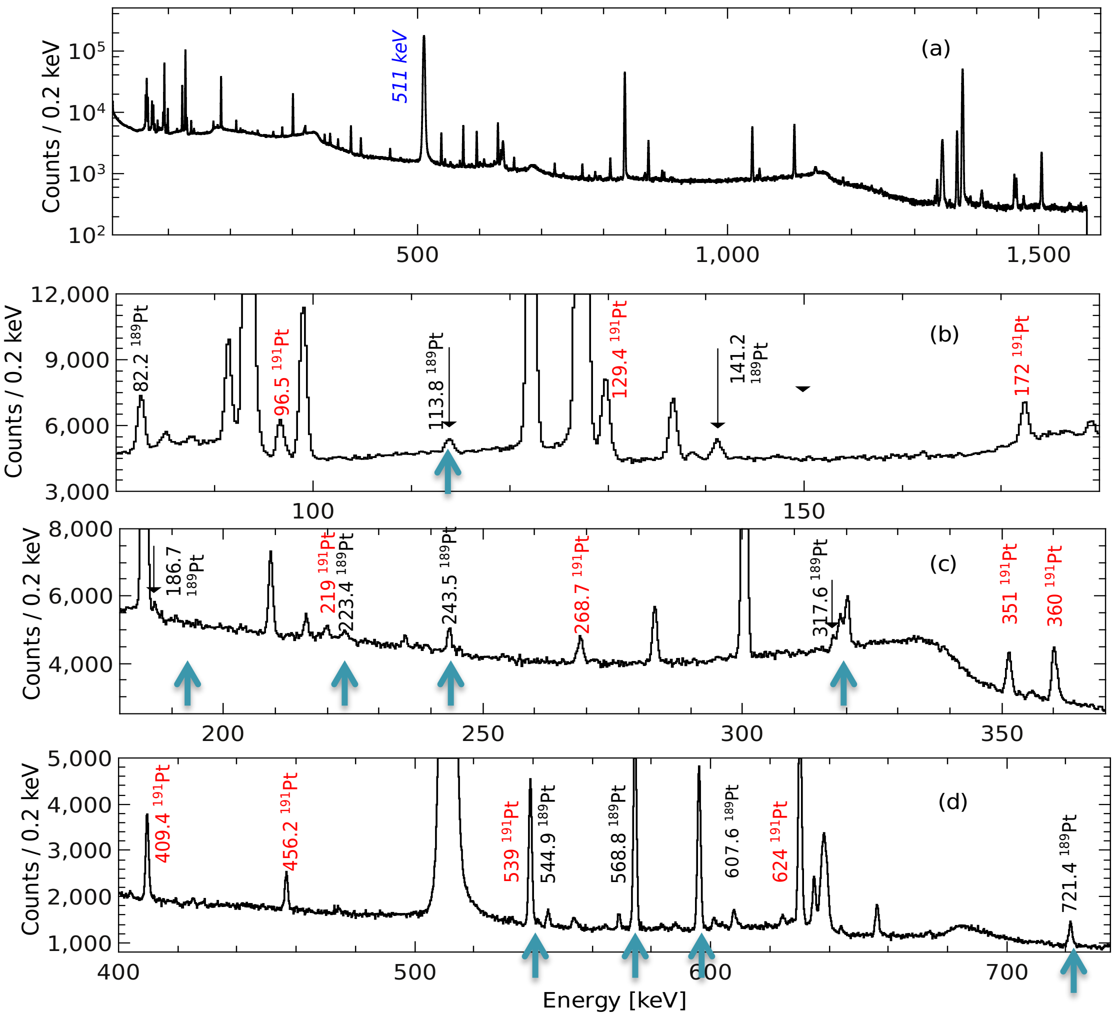


The thickness is: **98.6 nm**, corresponding to 0.222 mg/cm^2

The thickness is: **2240 nm**, corresponding to 5.05 mg/cm^2

GSI, Darmstadt 2019

Experimental results



Example of one of the reaction of interest:

$$^{4}_{2}He + ^{186}_{76}Os \rightarrow ^{189}_{78}Pt + n$$

Os stable isotopes:

^{184}Os	0.02 %
^{186}Os	1.59 %
^{187}Os	1.96 %
^{188}Os	13.24 %
^{189}Os	16.15 %
^{190}Os	26.26 %
^{192}Os	40.78 %

Vacuum
Volume 161, March 2019, Pages 162-167

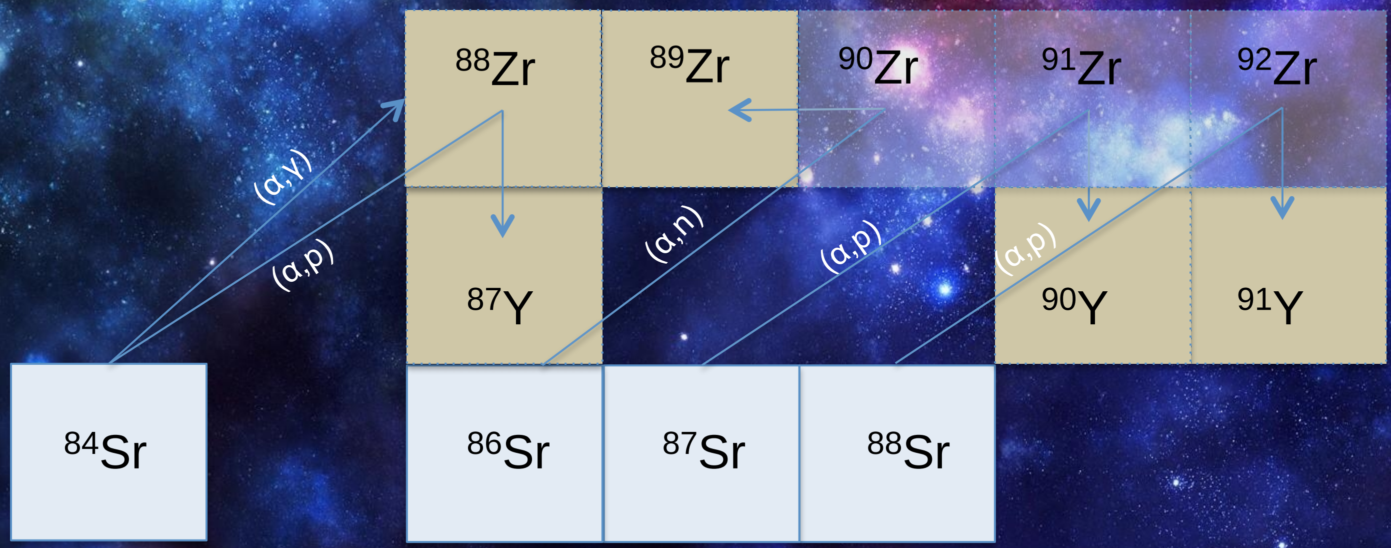
Refractory osmium targets for accelerator based nuclear activation experiments prepared by Pulsed Laser Deposition technique

Andreea Mitu [a, b], Marius Dumitru [c], Rareș Șuvăilă [a], Andreea Oprea [a], Ioana Gheorghe [a], Paul Mereuță [a], Simona Brajnicov [c], Ion Burducea [a], Nicoleta M. Florea [a], Nicolae Mărginean [a], Tudor Glodariu [a, 1], Maria Dinescu [c], Gheorghe Căta – Danil [b]

Strontium targets for nuclear astrophysics experiments

Proposed experiment

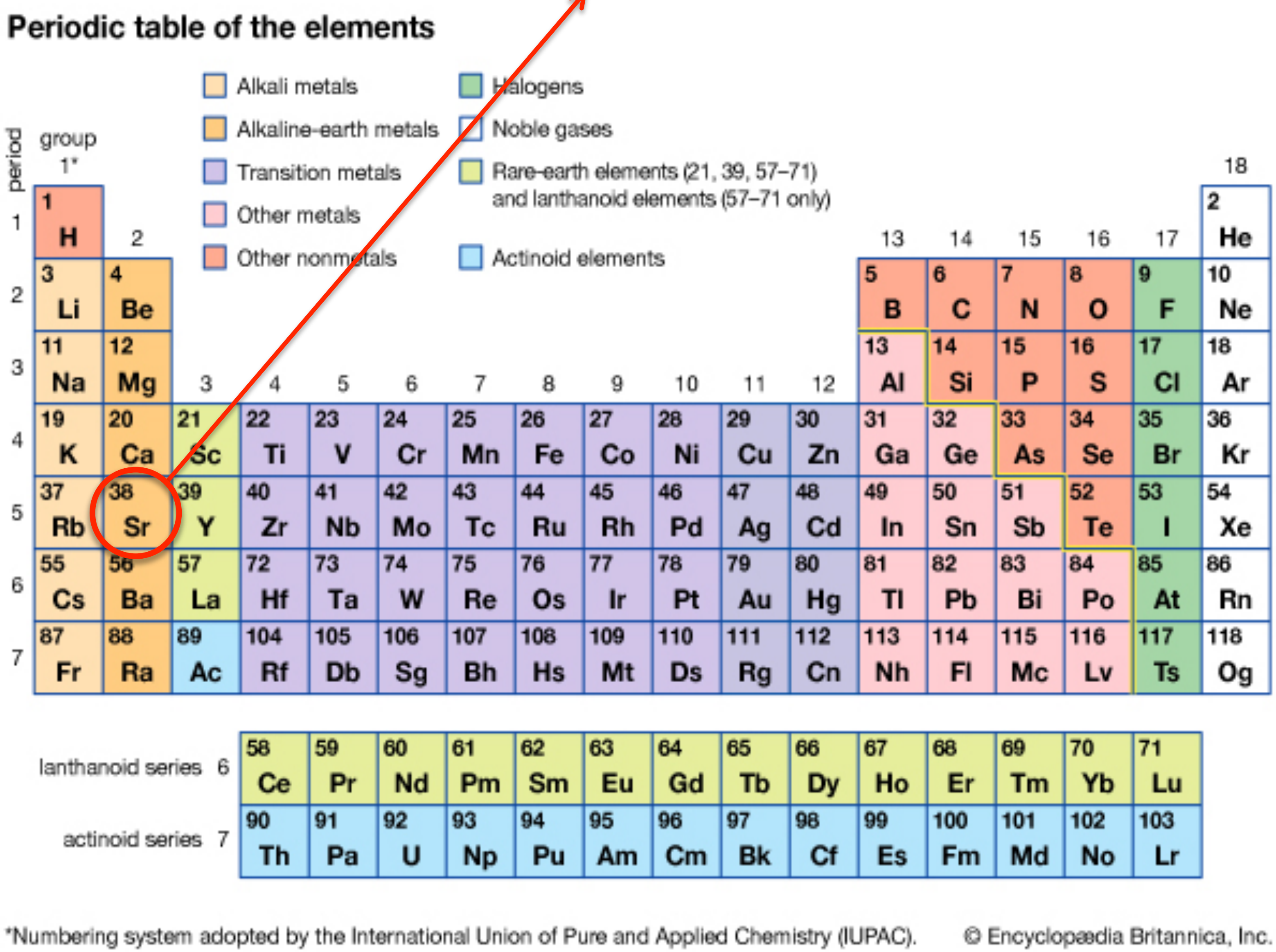
0.56 %	^{84}Sr
9.86 %	^{86}Sr
7.00 %	^{87}Sr
82.58 %	^{88}Sr



The isotopes produced on natural strontium targets by α irradiation

Ideal target

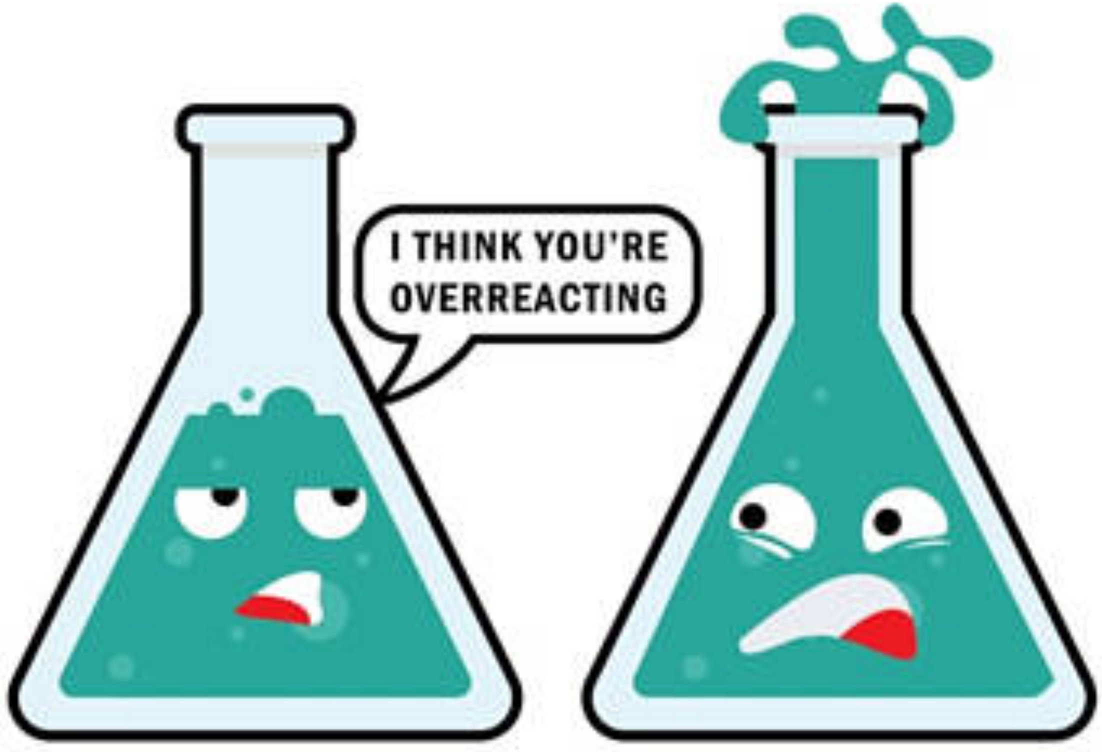
IFIN-HH



The question is…

NO!

Trouble…



Metallic Sr targets exhibit a significant chemical activity and oxidize very fast in regular environment (air)!

Solutions?

- This is a major constraint for measuring absolute cross-sections for the alpha particle activation of Sr isotopes.

Strontium compounds

Goal of the present study: overcome this constraint by using targets consisting of strontium compounds which are not reactive in air!

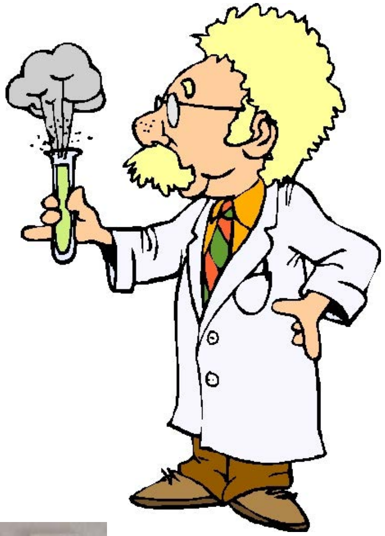
Targets

^{84}Sr

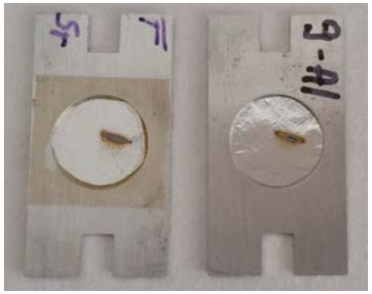
^{86}Sr

^{87}Sr

^{88}Sr



SrF_2



$SrCO_3$

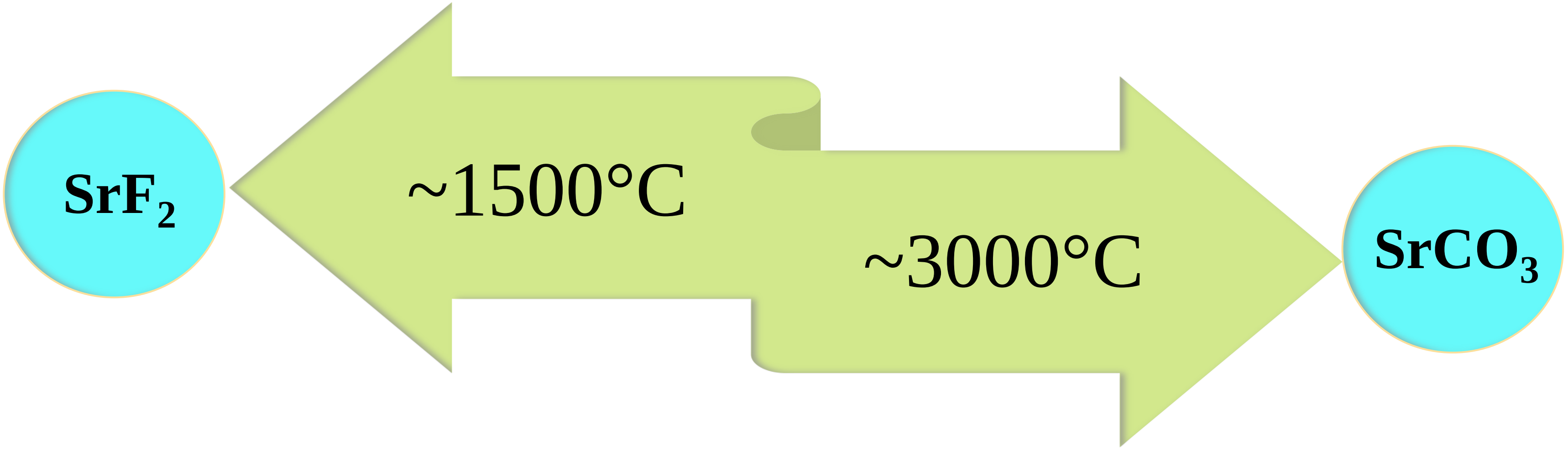
Preparation of Sr targets

1. Pressing powder

2. Heating pellet

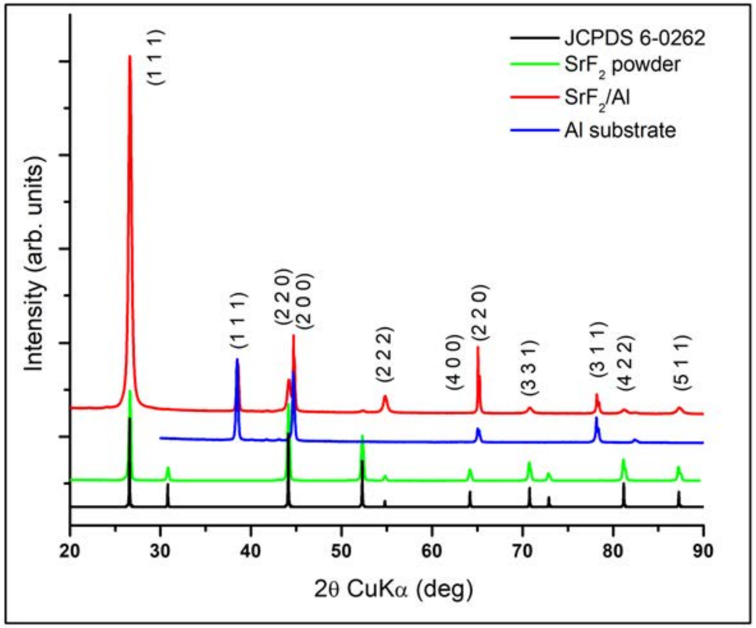
3. Evaporation of the Sr compound on Al backing

Vaporization temperature



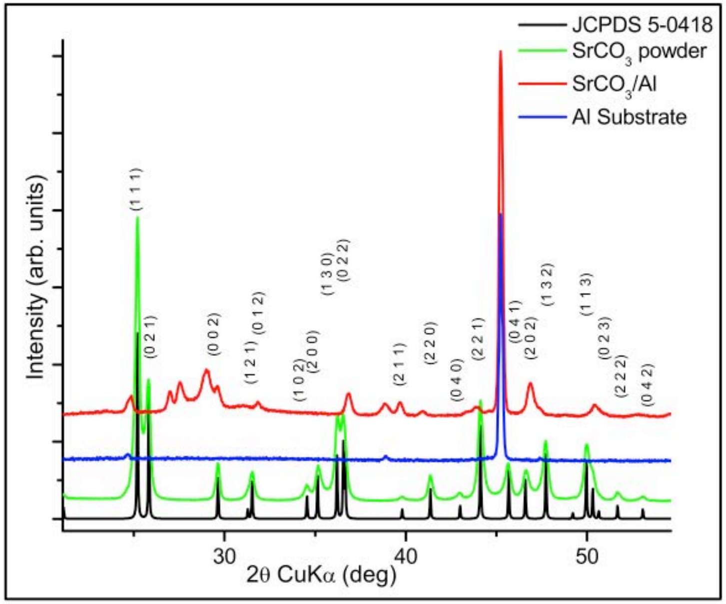
X-ray diffraction

SrF_2



XRD patterns of Al substrate, SrF_2 powder and SrF_2/ Al target

$SrCO_3$



XRD patterns of Al substrate, $SrCO_3$ powder and $SrCO_3$/ Al target

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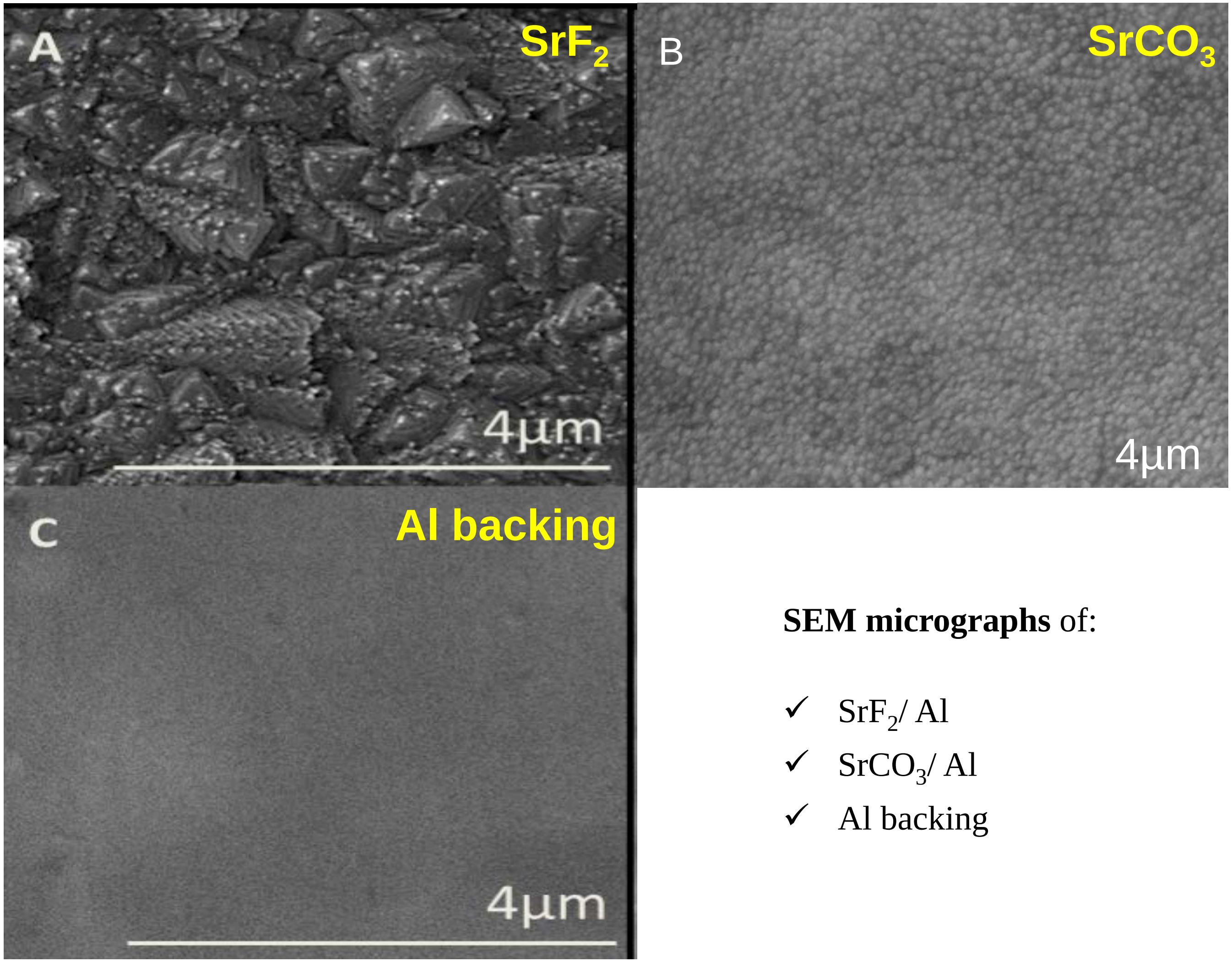
SEM micrographs



SEM micrographs of:

- ✓ SrF_2/ Al
- ✓ $SrCO_3$/ Al
- ✓ Al backing

EDX analyses

Table 1: SrF_2 target composition by EDX analysis

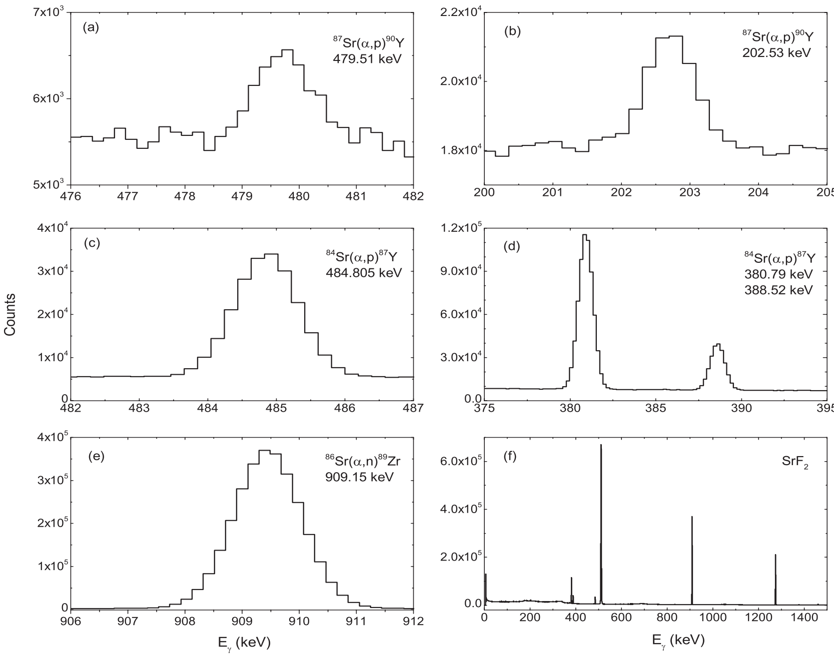
Element	Wt %	At %	K-Ratio
F K	26.06	61.91	0.0689
Sr K	73.94	38.09	0.6806
Total	100.00	100.00	
kV: 20.01	Det Type: SUTW, Sapphire		

Table 2: $SrCO_3$ target composition by EDX analysis

Element	Wt %	At %	K-Ratio
C K	12.455	31.807	0.0749
O K	20.950	40.642	0.1884
Cu L	31.192	15.140	0.2607
Sr L	35.403	12.411	0.2799
Total	100.00	100.00	
kV: 5.01	Det Type: SUTW, Sapphire		

Copper contamination!

Nuclear activation spectra



SrF_2

$^{87}Sr(\alpha,p)^{90}Y$

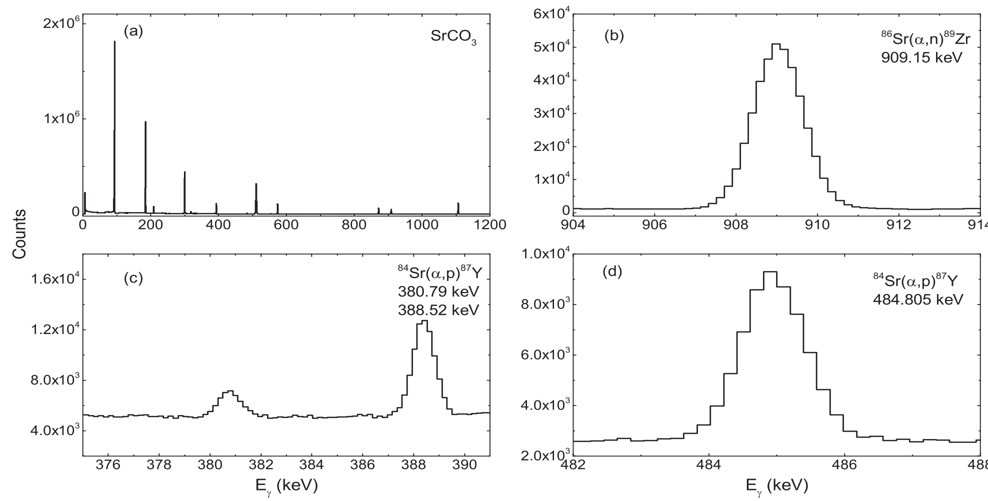
$^{84}Sr(\alpha,p)^{87}Y$

$^{86}Sr(\alpha,n)^{89}Zr$

$SrCO_3$

$^{86}Sr(\alpha,n)^{89}Zr$

$^{84}Sr(\alpha,p)^{87}Y$



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Preparation and characterization of strontium targets for nuclear astrophysics experiments

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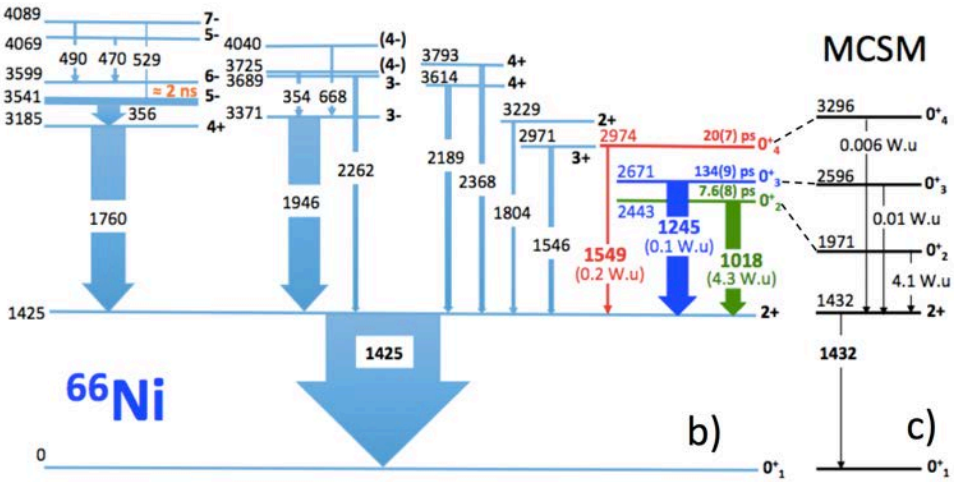
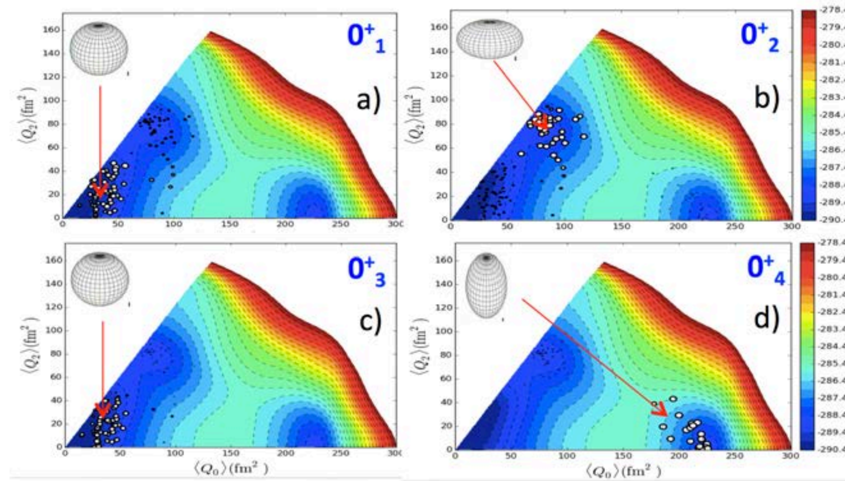
Received: 8 February 2018

Decontamination of ^{64}Ni targets

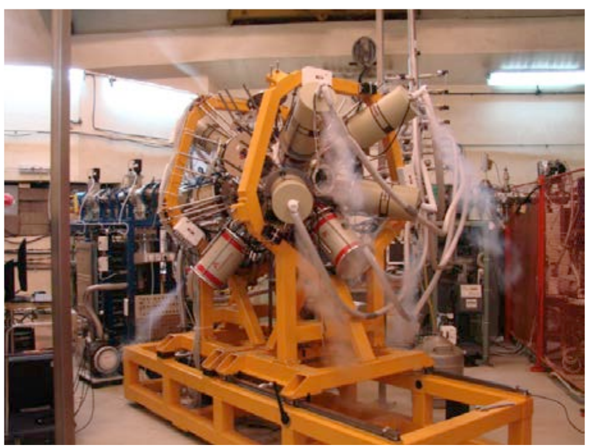
AIM and IMPORTANCE of the experiment

- shape coexistence in ^{66}Ni

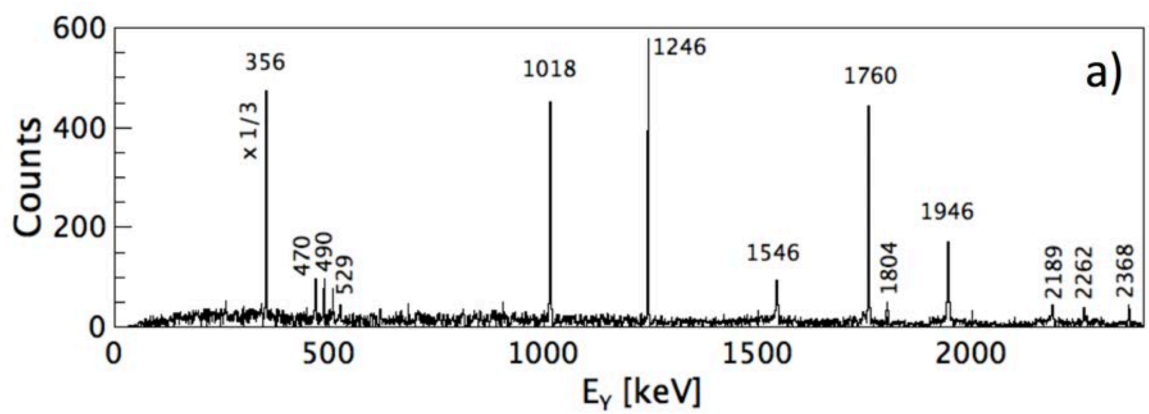




T. Otsuka , private communication



^{64}Ni(^{18}O,^{16}O)^{66}Ni 39 MeV ROSPHERE array



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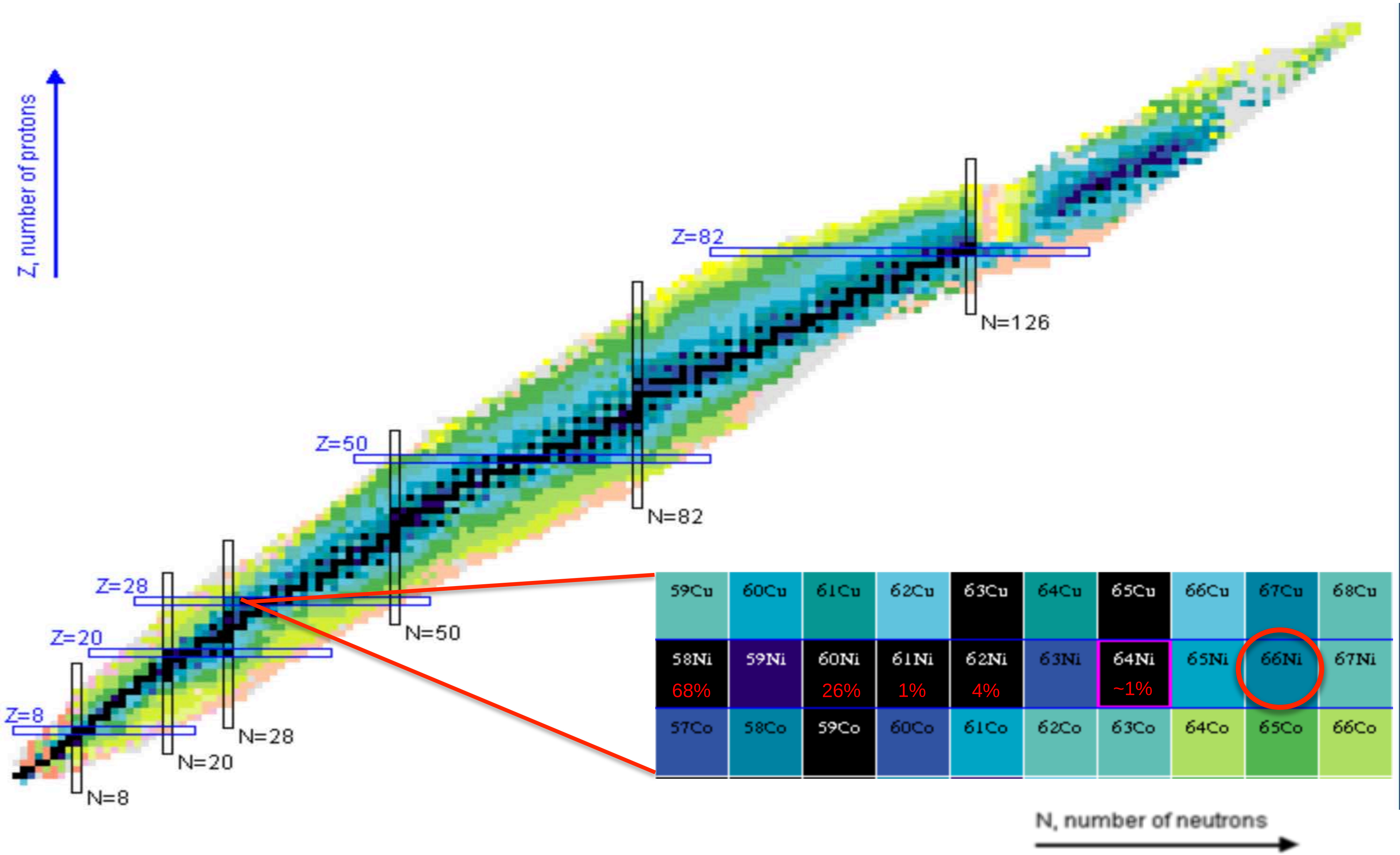
Target used in the experiment



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Targets required for the proposed experiment

2 thick targets

5 mg/cm²

3-4 thin targets

1 mg/cm²

To achieve all these targets I started from 100 mg of ^{64}Ni powder

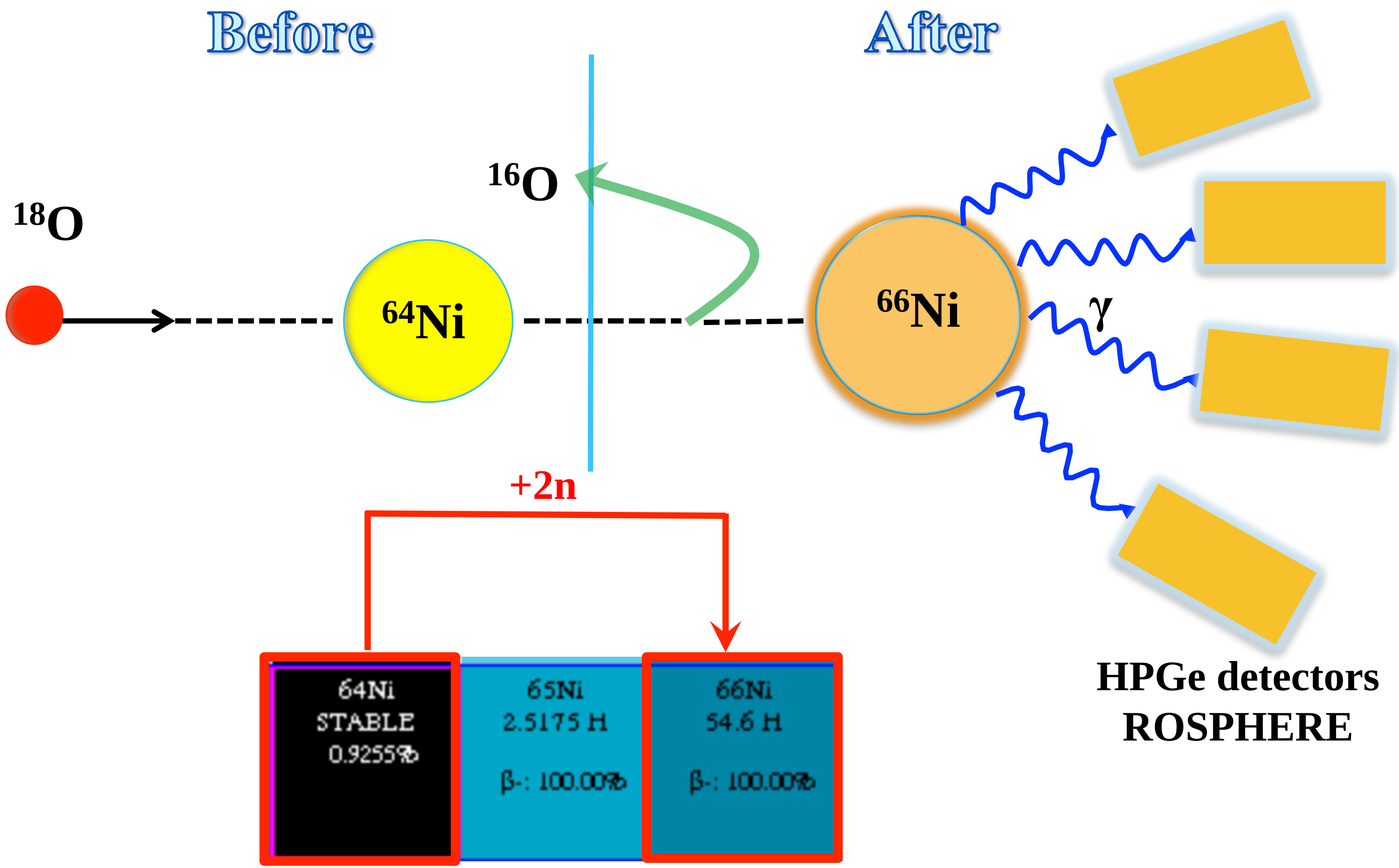
Preparation of ^{64}Ni foils

1. Pressing powder

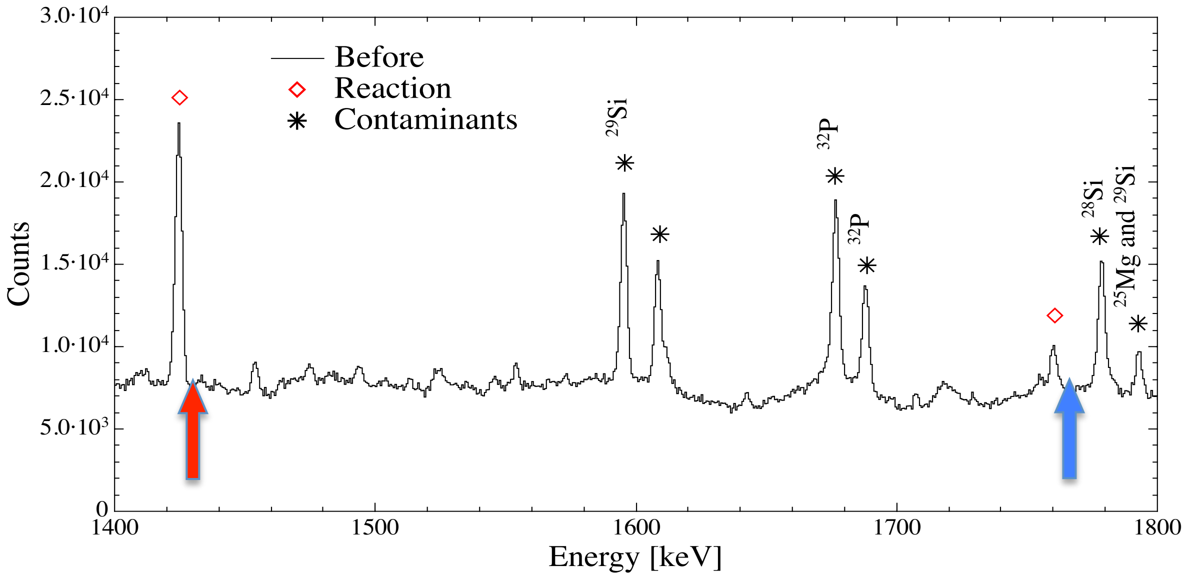
2. Heating pellet

3. Rolling process

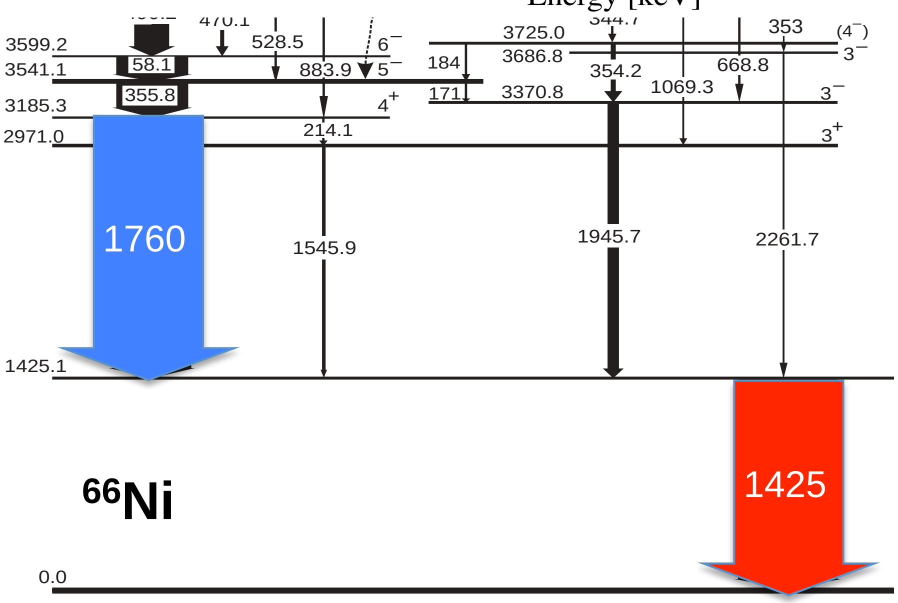
Reaction Kinematics below Coulomb barrier



$^{18}O + ^{64}Ni$ @ 39 MeV



Fusion-evaporation cross-section calculation
Contaminants from ^{18}O on ^{16}O



1.Yields of residual nuclei

Z	N	A		events	percent	x-section(mb)
16	17	33	S	42	0.42%	3.73
15	18	33	P	42	0.42%	3.73
16	16	32	S	1111	11.1%	98.7
15	17	32	P	2021	20.2%	180
14	18	32	Si	68	0.68%	6.04
16	15	31	S	23	0.23%	2.04
15	16	31	P	1906	19.1%	169
14	17	31	Si	151	1.51%	13.4
14	16	30	Si	73	0.73%	6.48
14	15	29	Si	4063	40.6%	361
13	16	29	Al	304	3.04%	27
14	14	28	Si	31	0.31%	2.75
13	15	28	Al	1	0.01%	0.0888
12	14	26	Mg	103	1.03%	9.15
12	13	25	Mg	61	0.61%	5.42
TOTAL				10000	100%	888

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- How to improve the experimental results?

↓

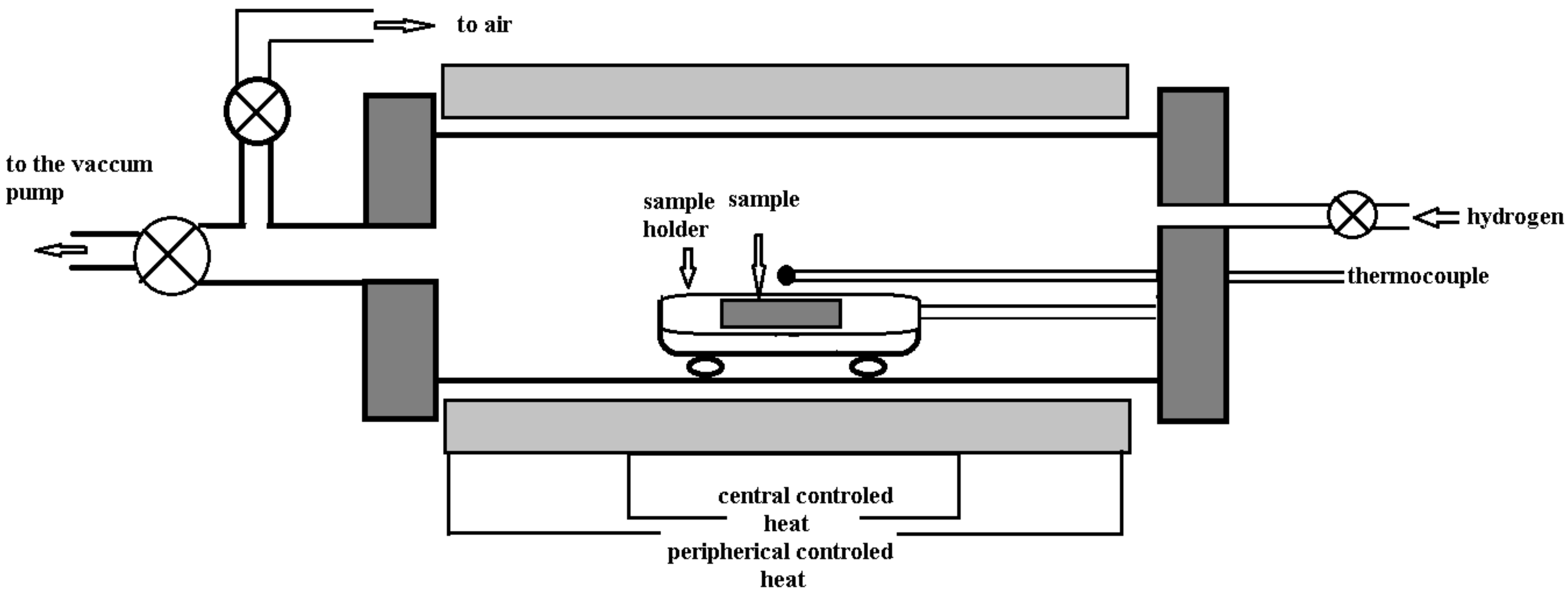
Removing oxygen

How to remove the oxygen?

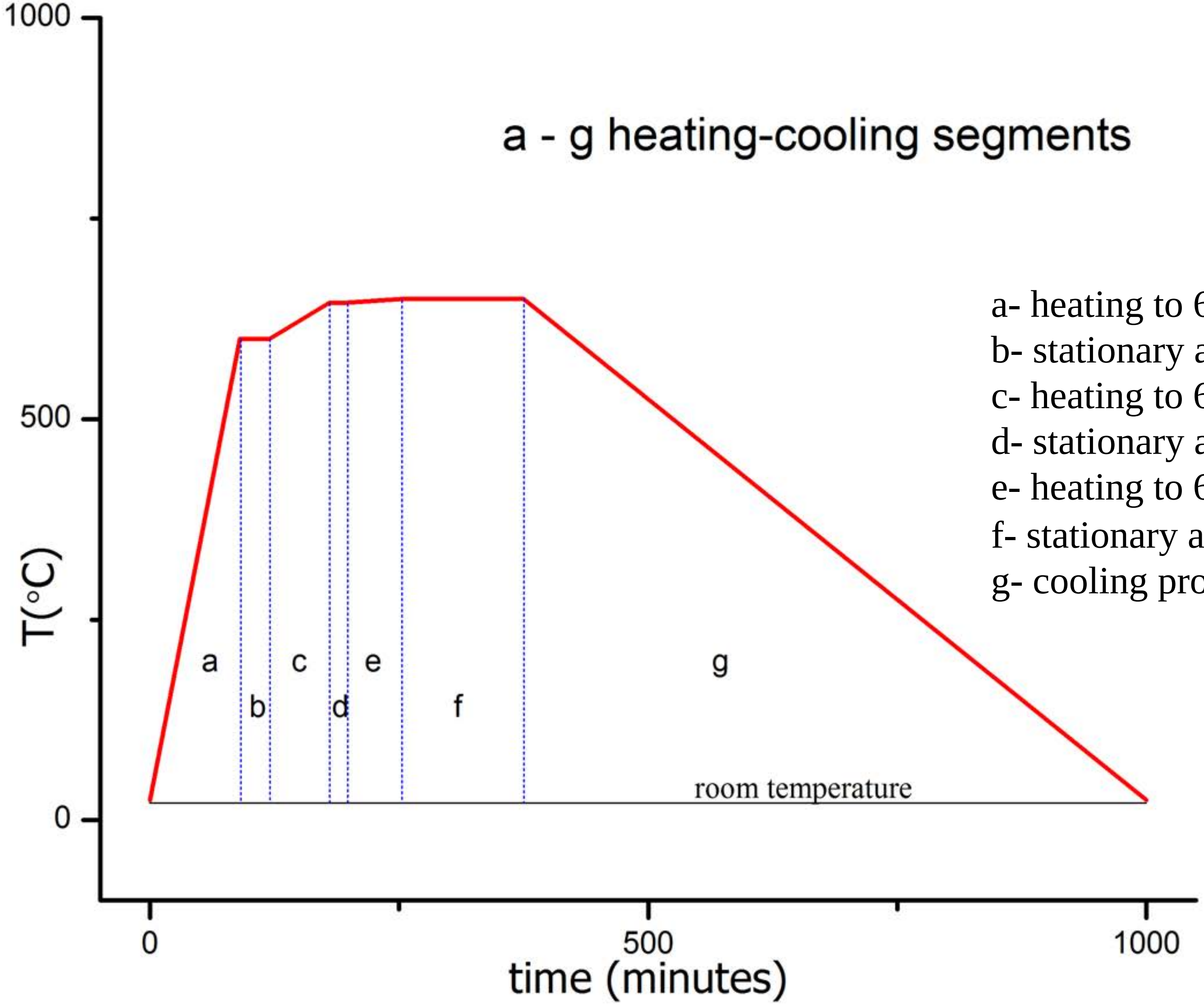
Which is the first formula that we learn in chemistry?

$$O + 2H = H_2O$$

Hydrogen oven



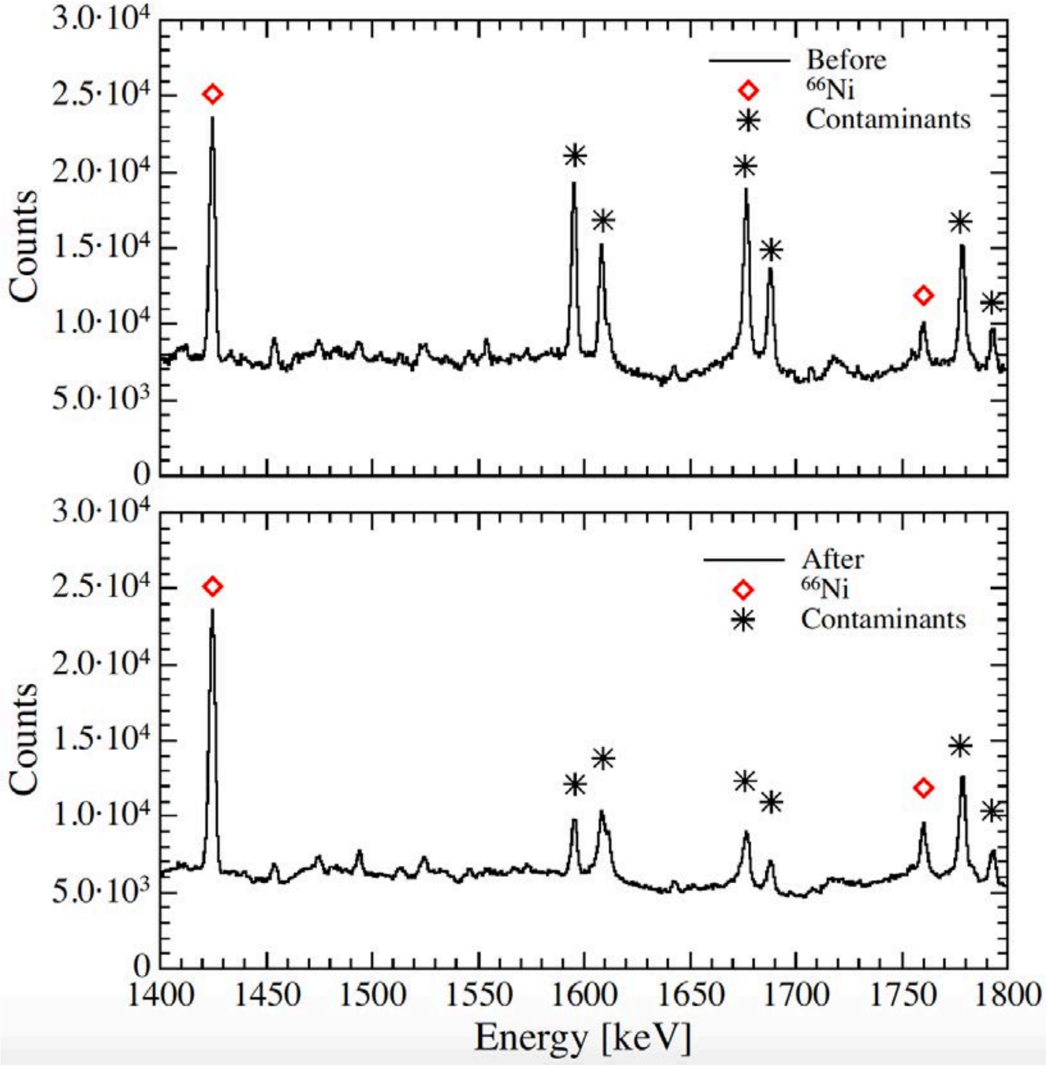
The thermal treatment for the ^{64}Ni target

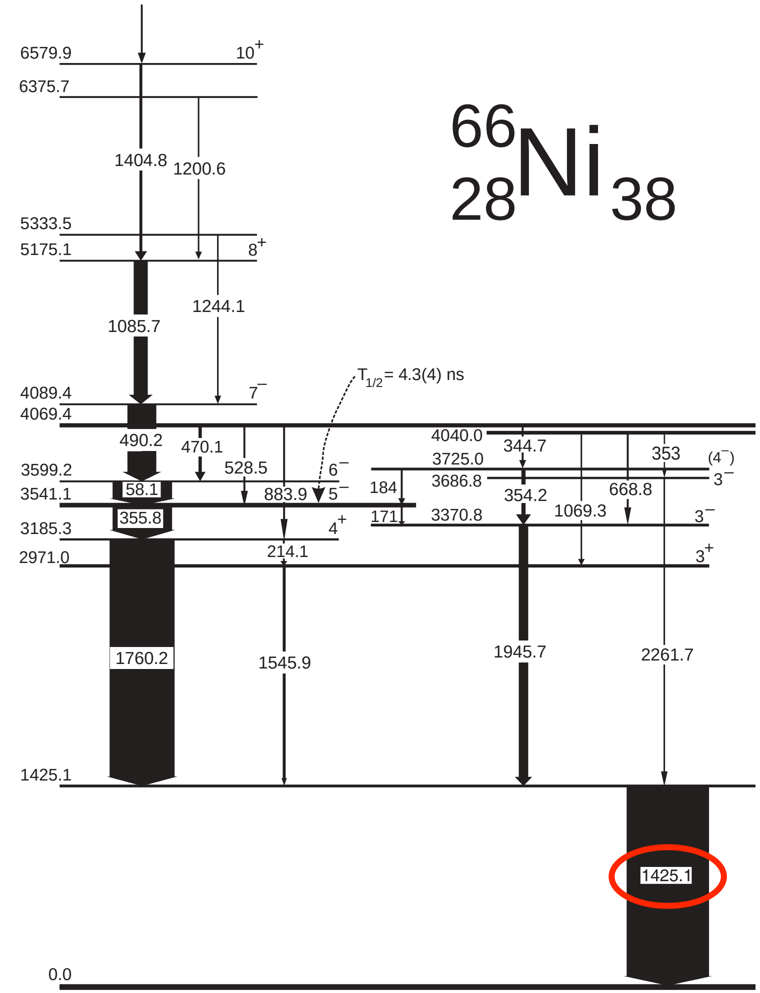


a- heating to 600^0C (60min)
b- stationary at 600^0C (30min)
c- heating to 645^0C (60min)
d- stationary at 645^0C (15min)
e- heating to 650^0C (60min)
f- stationary at 650^0C(120min)
g- cooling process to 25^0C (625min)

Comparison between before (top) and after (bottom) the "target treatment"

The peak at 1425 keV has been used as normalization the two spectra





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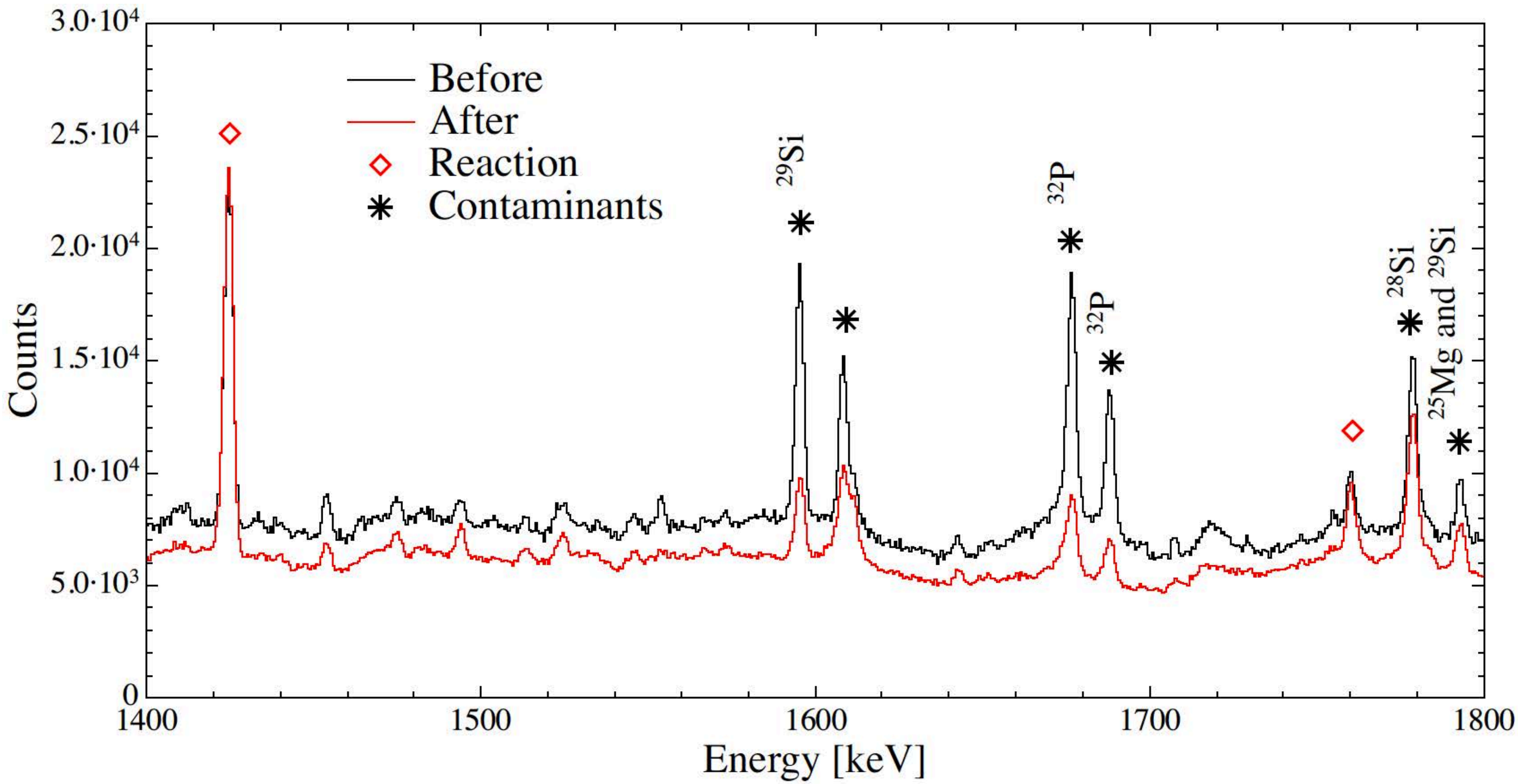
$^{18}O + ^{64}Ni$ @ 39 MeV
($\gamma\gamma$ – total projection)



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HIGH GRADE DECONTAMINATION OF Ni TARGETS FOR SUB-BARRIER TRANSFER REACTIONS

Andreea MITU[1,2], Marius DUMITRU[3], Florian DUMITRACHE[3], Nicolae MARGINEAN[1], Rareș ȘUVĂILĂ[1], Cristina NIȚĂ[1], Maria DINESCU[3], Gheorghe CĂTA –DANIL[2]

PRL **118,** 162502 (2017) PHYSICAL REVIEW LETTERS week ending 21 APRIL 2017

Multifaceted Quadruplet of Low-Lying Spin-Zero States in ^{66}Ni: Emergence of Shape Isomerism in Light Nuclei

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Conclusions and perspectives

ACHIEVEMENTS:

- ✓ ***Thin films preparation for various types of nuclear physics experiments with accelerators***
- ✓ ***Production of targets by pulsed laser deposition***
- ✓ ***Target quality - characterization in terms of thickness, uniformity, purity in collaboration with several departments of our Institute***

LOOKING FORWARD:

- ✓ ***Developing a database of target laboratories, available methods***
- ✓ ***The first nuclear forensics laboratory of Romania***

THANK YOU FOR YOUR ATTENTION!

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