Magnetron sputtering for corrosion protection of liquid cyclotron target for Fluorine-18 production

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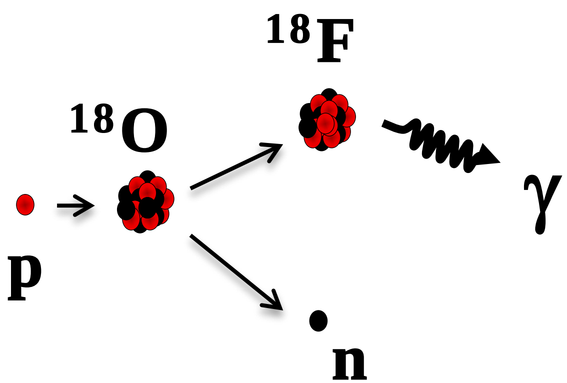
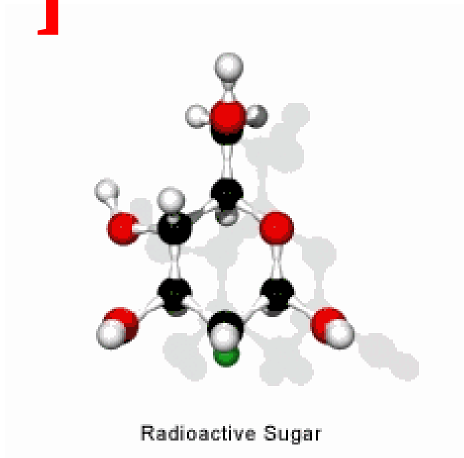
R&D of thin film deposition techniques

in order to provide:

"… a chemical passive surface of targets for radioisotope production …"

Particularly, the [^{18}O]H_2O target for production of [$^{18}F^-$]



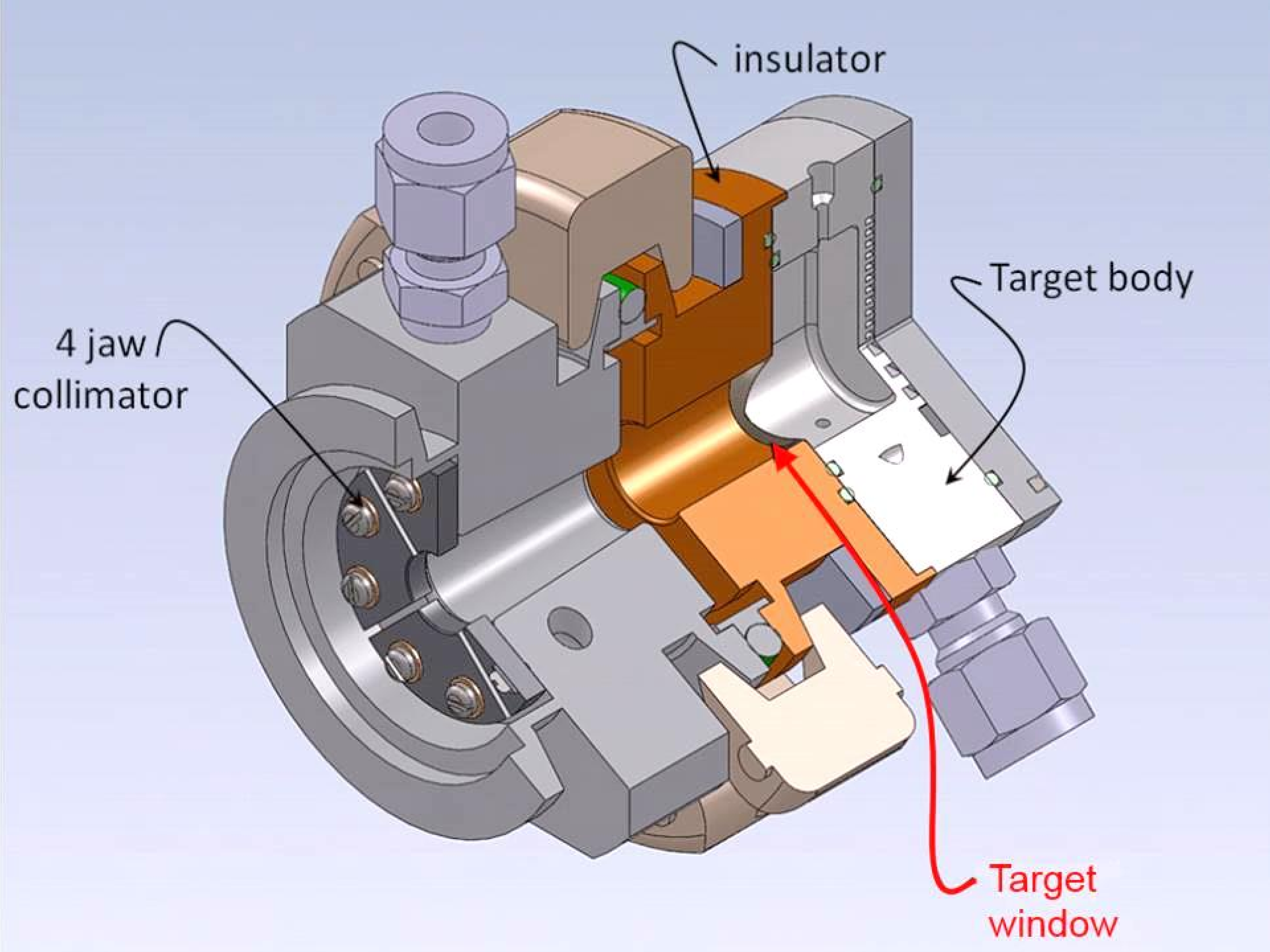


FDG is the most used radioisotope in PET

Proton irradiation cause water radiolysis

$$H_2O \rightarrow H_2,\ O_2,\ H_2O_2,\ OH,\ H,\ e^-_{aq},$$
$$HO_2,\ O_2^-,\ HO_2^-,\ OH^-,\ H^+, \ldots$$

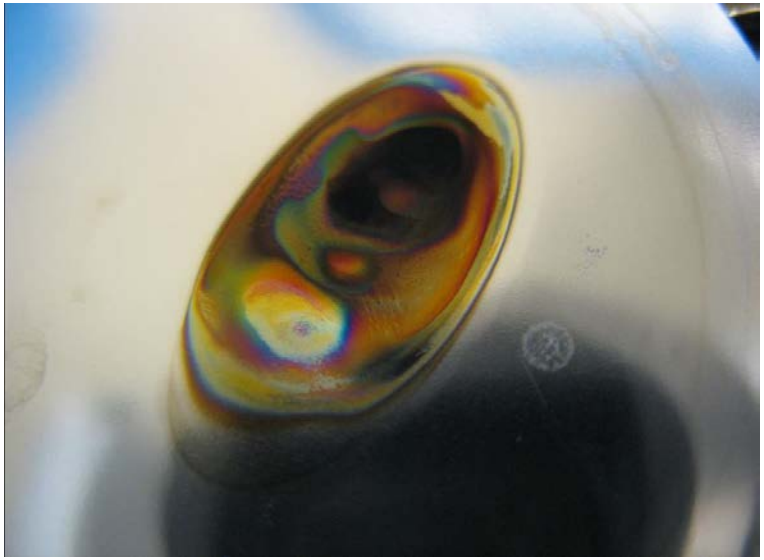
Proton irradiated water is extremely corrosive!


insulator
Target body
4 jaw
collimator
Target
window

Entrance beam window:

- **high tensile strength substrate**

  e.g. **Havar®** **(Co, Cr, Fe, Ni, W, Mo, Mn, C)**



Havar® foil corroded on beam spot

Need of corrosion resistant top-coating

onto the **Havar® beam window**

Candidates:

Nb, Ta, Pt, Zr…

Chemical inertness is mandatory,

but not enough

Microstructure has a great influence on corrosion process

Microstructure requirements:

- Coating must be dense with minimal distance between grain boundaries

- **The best possible Diffusion Barriers are Amorphous!**

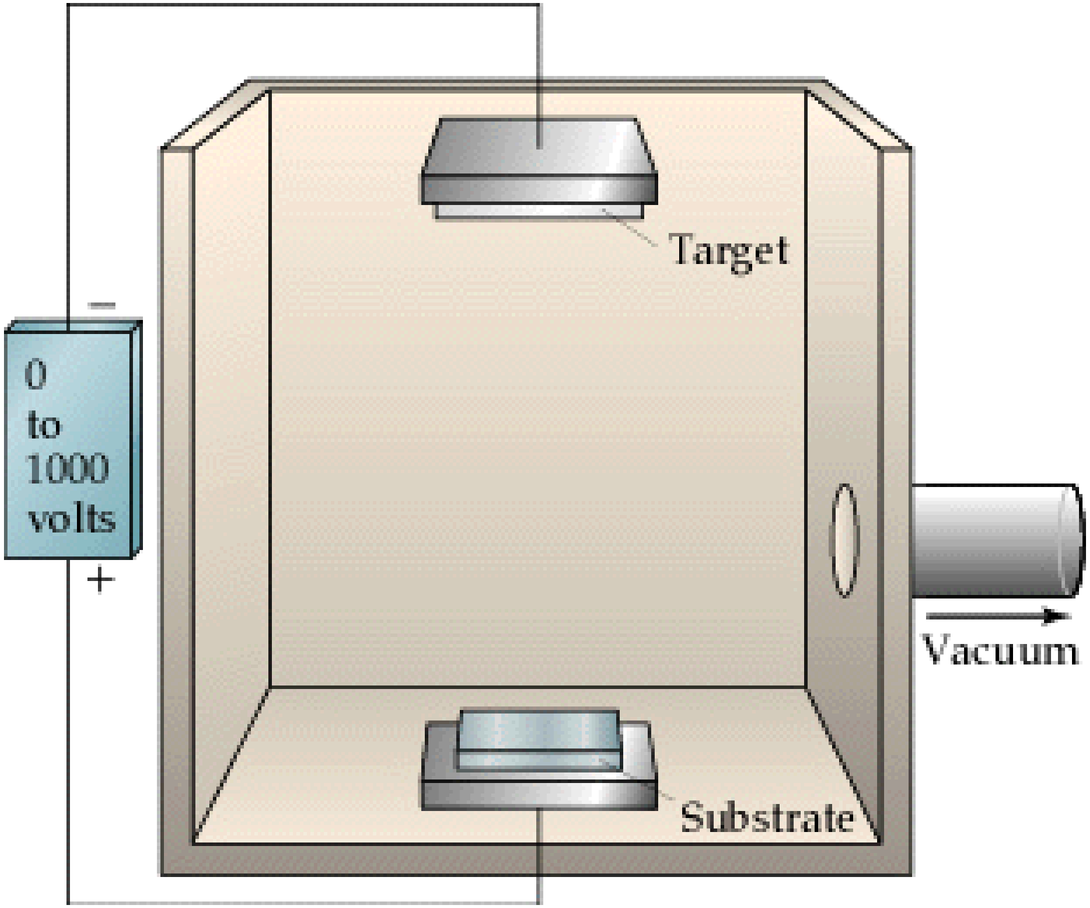
- **Requirement from *BEST Cyclotron Systems Inc.*:**
  - **Uniform thickness**

- **INFN Suggestions:**
  - **Absence of pin-holes**
  - **Low porosity**
  - **Low diffusion across grain boundaries**

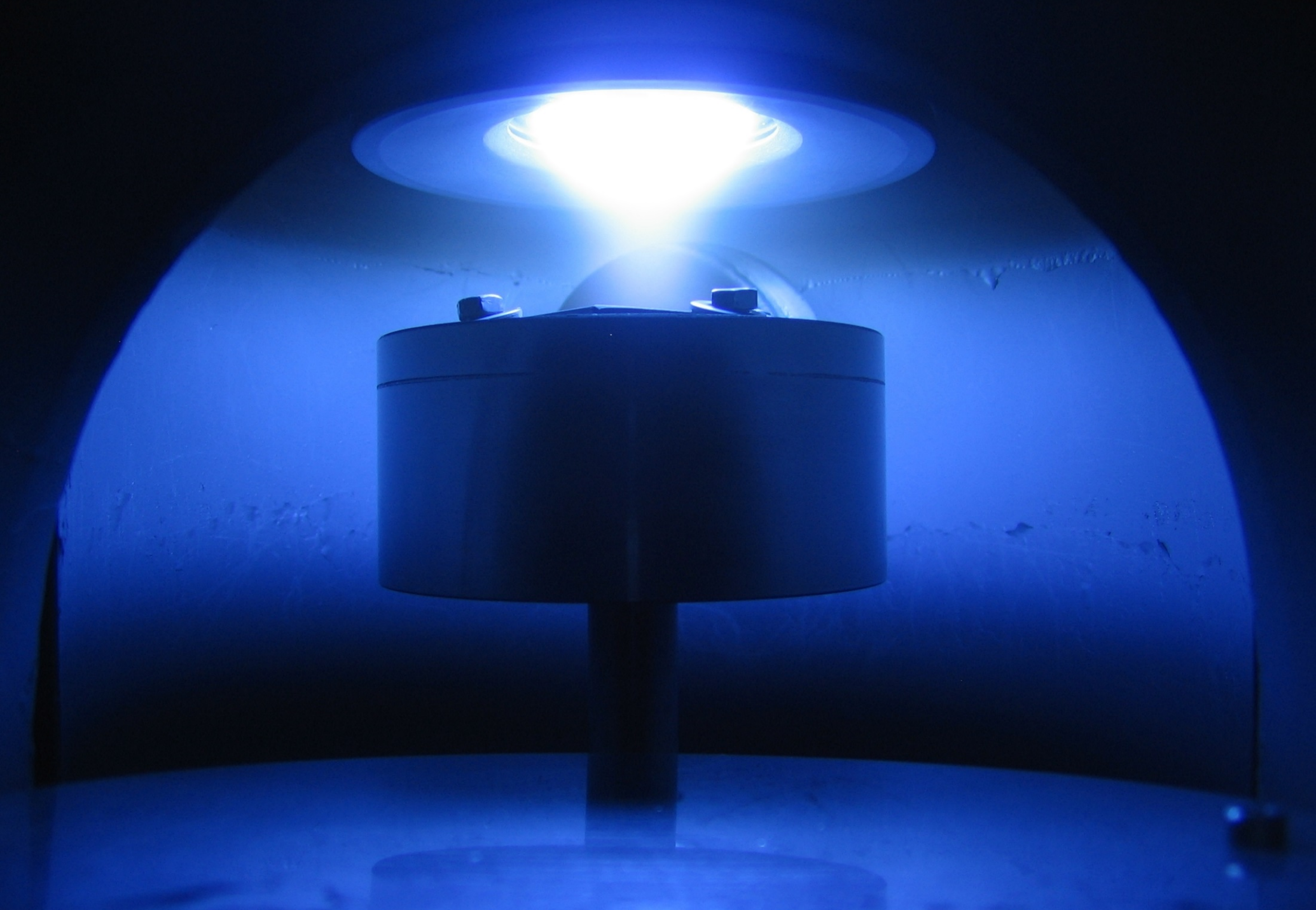
Deposition method:

Sputtering is a method to deposit thin film onto a surface (substrate)

Sputtering

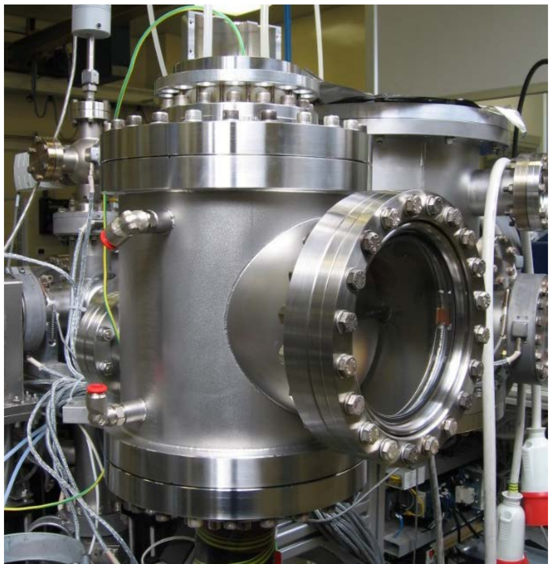


Magnetron sputtering

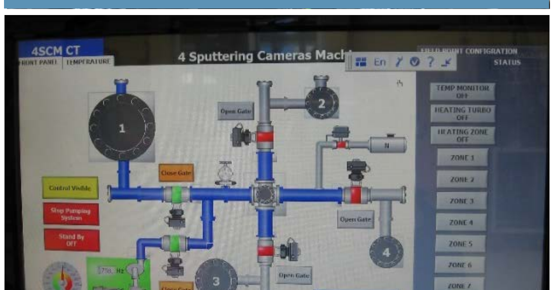


Our sputtering facility:

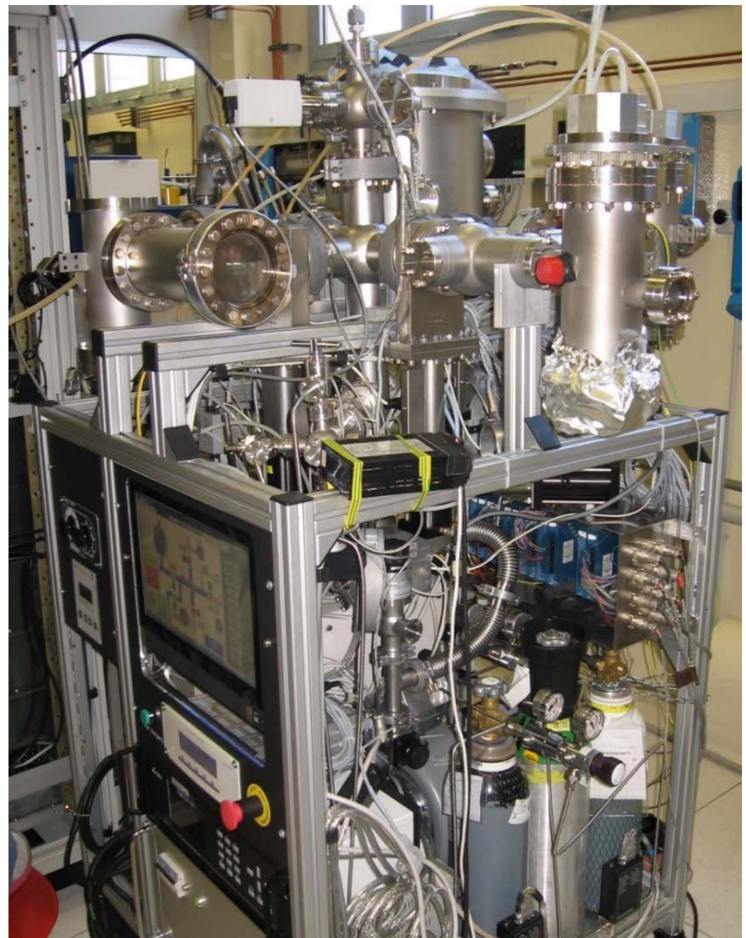
Laboratories for Surface and Material Treatments in Nuclear Physics



Working chamber



Control panel

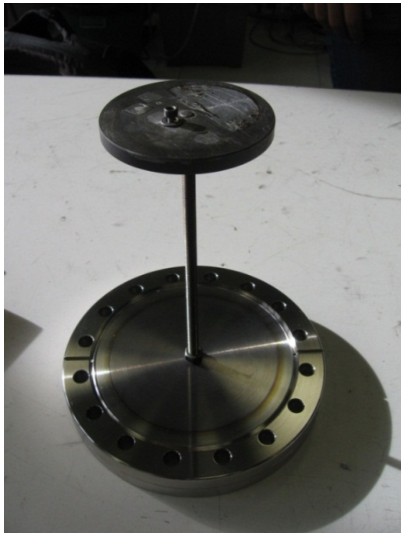


Full sputtering system

INFN
LNL
Istituto Nazionale di Fisica Nucleare

Substrate holders

- grounded



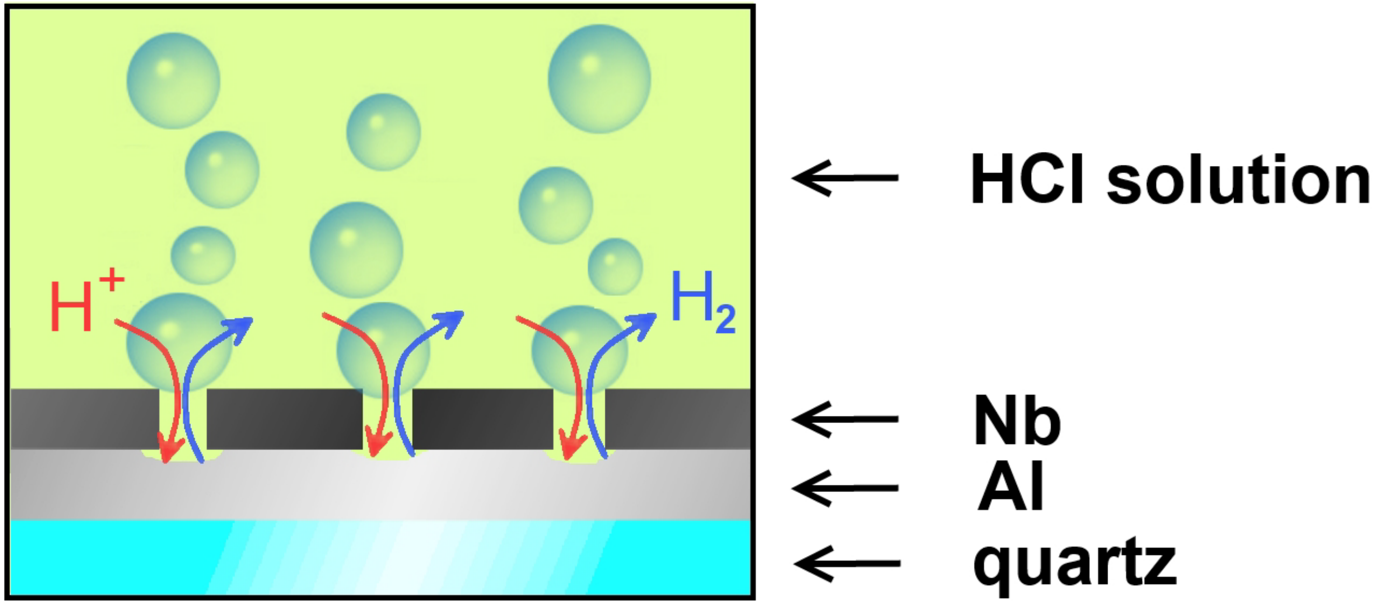
- liquid nitrogen-cooled

- heated

- water-cooled

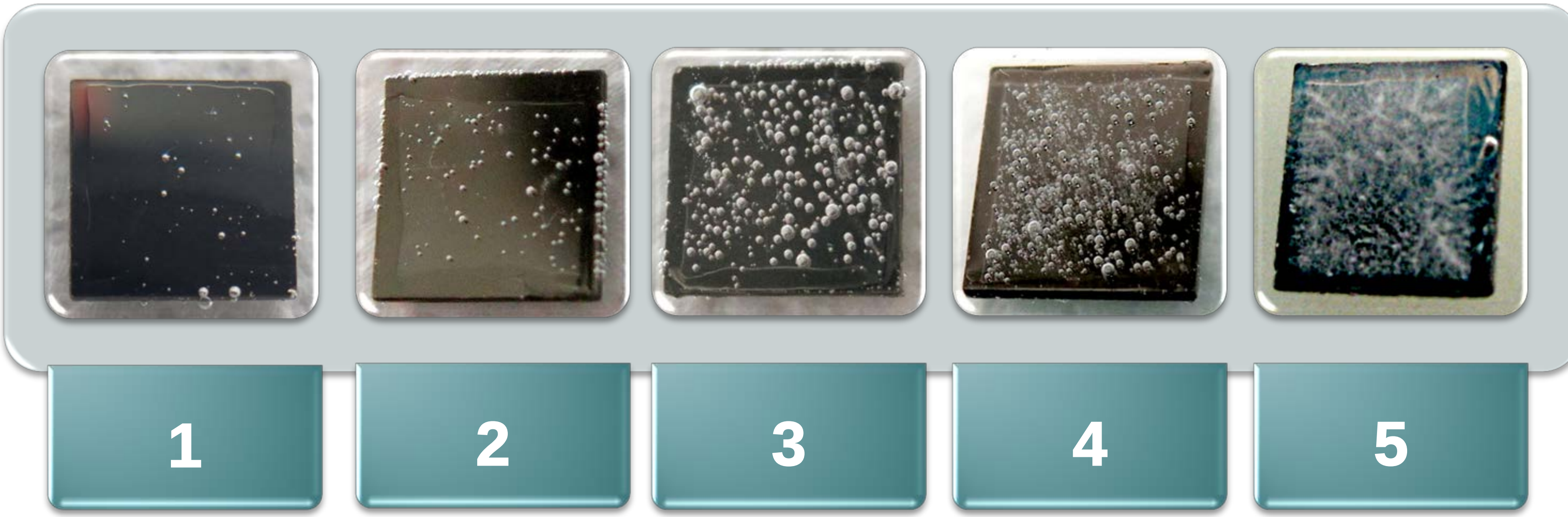
Analyzing technique

Acid porosity test:

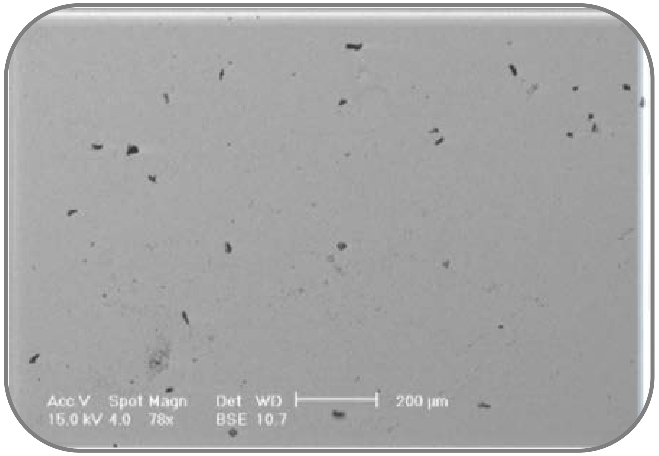


Analyzing technique

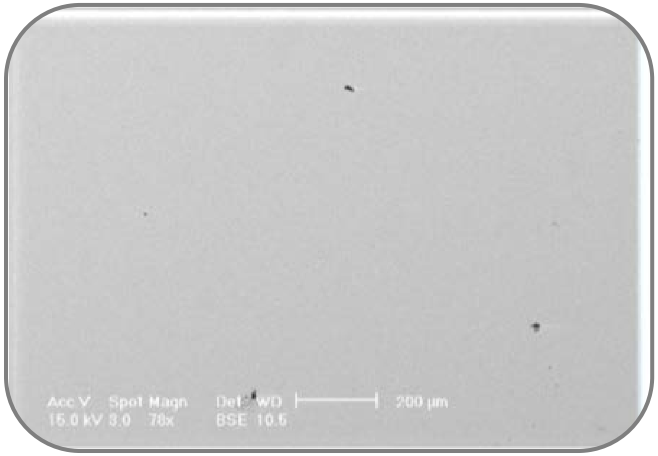
Acid porosity test: **10% HCl , 30°C, 10 min**



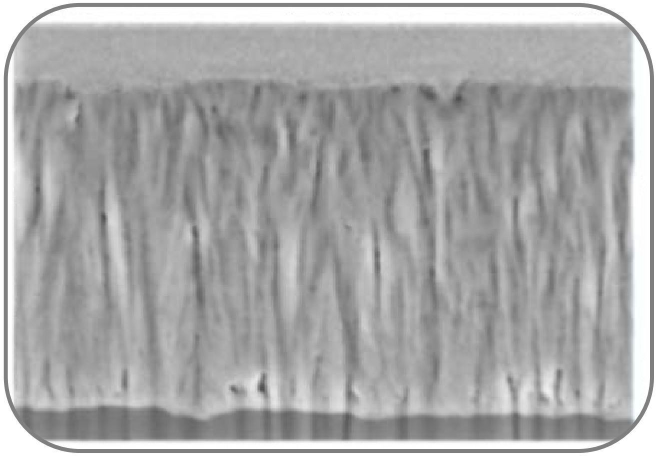
Analyzing technique: SEM, FIB SEM



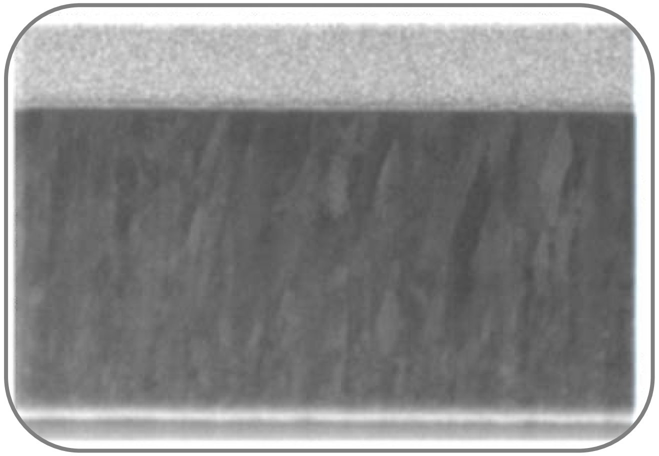
SEM



SEM



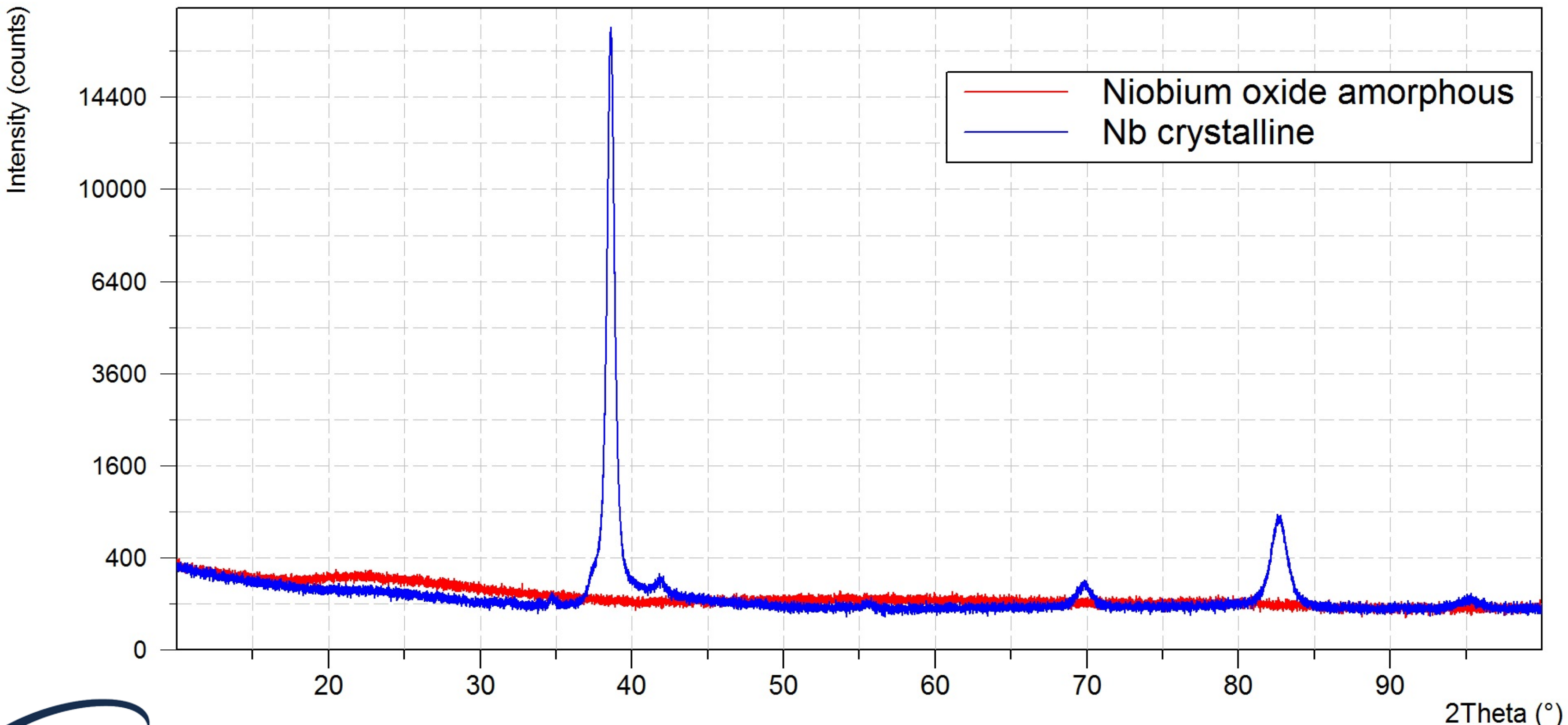
FIB SEM



FIB SEM

Analyzing technique

X-ray diffractometry



Coating systems investigated:

- **Nb**
- **Nb_2O_5**
- **Nb/Nb_2O_5 multilayers**
- **Nb-Ta, Nb-Zr, Ta-Zr**

Parameters investigated for Nb coatings:

substrate temperature

- -100°C ÷ 500°C

applied bias

- -150 V ÷ +80 V

sputtering gas pressure

- $3 \cdot 10^{-3}$ mbar ÷ $3 \cdot 10^{-2}$ mbar

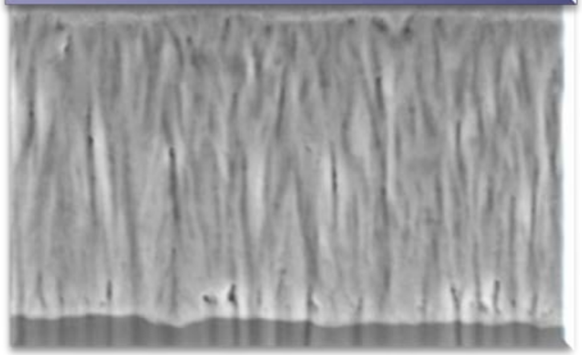
deposition rate

- 0.5 nm/sec ÷ 5 nm/sec

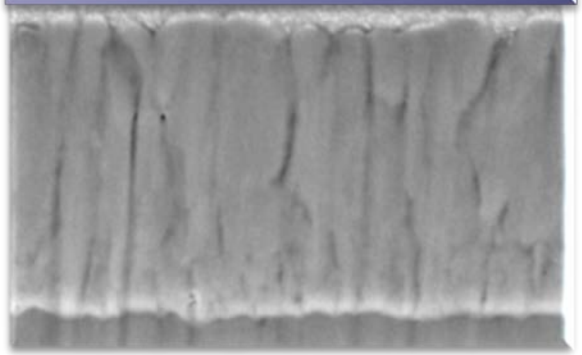
Niobium-based sputtered thin films for Corrosion Protection of proton-irradiated liquid water targets for [^{18}F] production , H. Skliarova, O. Azzolini, O. Dousset, R. R. Johnson, V. Palmieri, *Journal of Physics D Applied Physics* 08/2013; 47(4).

Temperature influence:

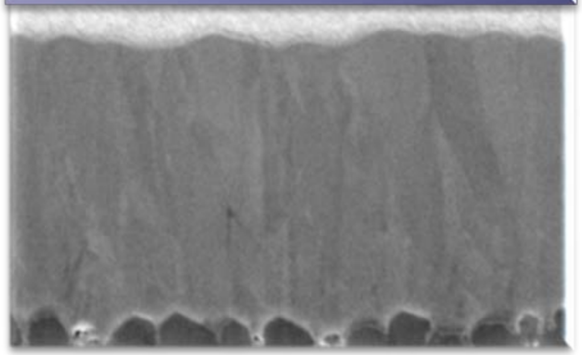
-100 °C



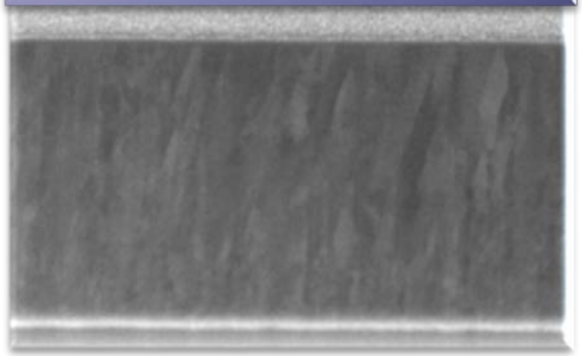
0 °C



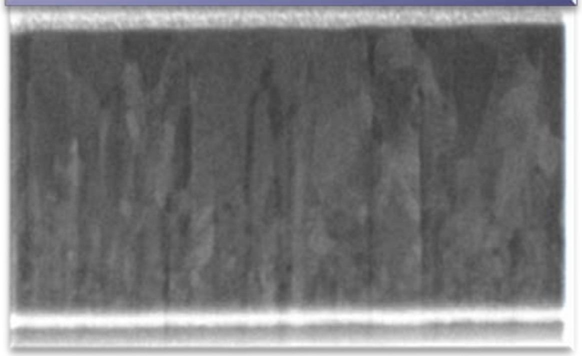
Floating



500 °C



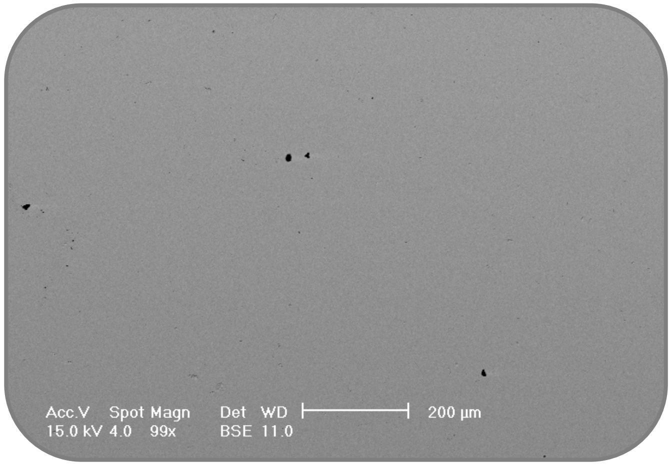
800 °C



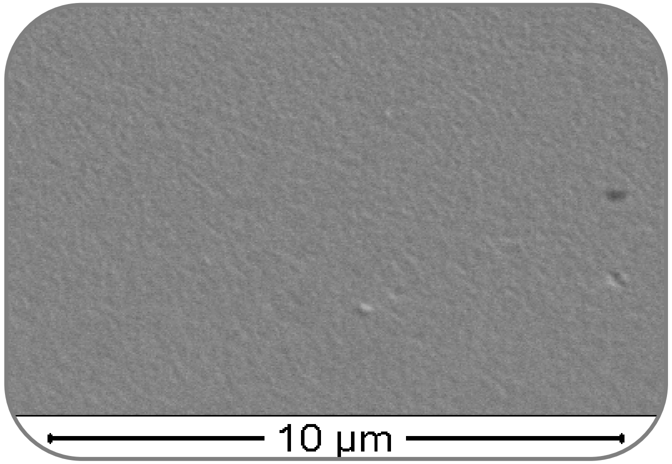
Best Nb deposition recipe:

P_{Ar}:	I_{DC}:	Substrate bias:	Substrate temperature
$3 \cdot 10^{-3}$ mbar	0.5 A	-80 V	no heating/ no cooling (~250°C)

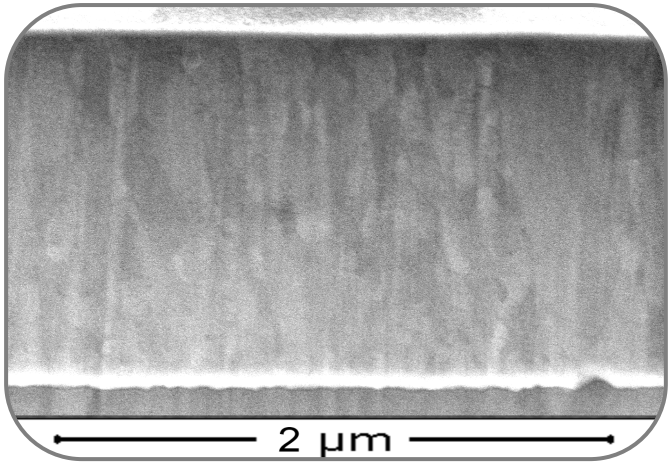
DC-biased MS of Nb:



SEM



SEM



FIB SEM

Reactive sputtering of Nb_2O_5 :

Sputtering gas pressure

- $8 \cdot 10^{-3}$ mbar ÷ $7 \cdot 10^{-2}$ mbar

Stoichiometry: Ar/O_2

- Ar/O_2

Applied bias

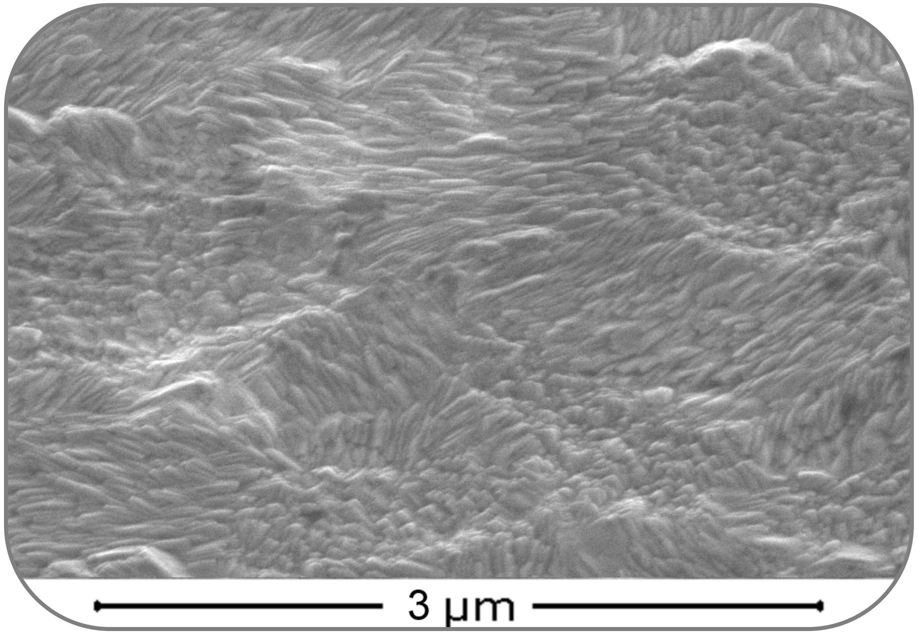
- -80 V ÷ 0 V

Niobium-based sputtered thin films for Corrosion Protection of proton-irradiated liquid water targets for [^{18}F] production , H. Skliarova, O. Azzolini, O. Dousset, R. R. Johnson, V. Palmieri, *Journal of Physics D Applied Physics* 08/2013; 47(4).

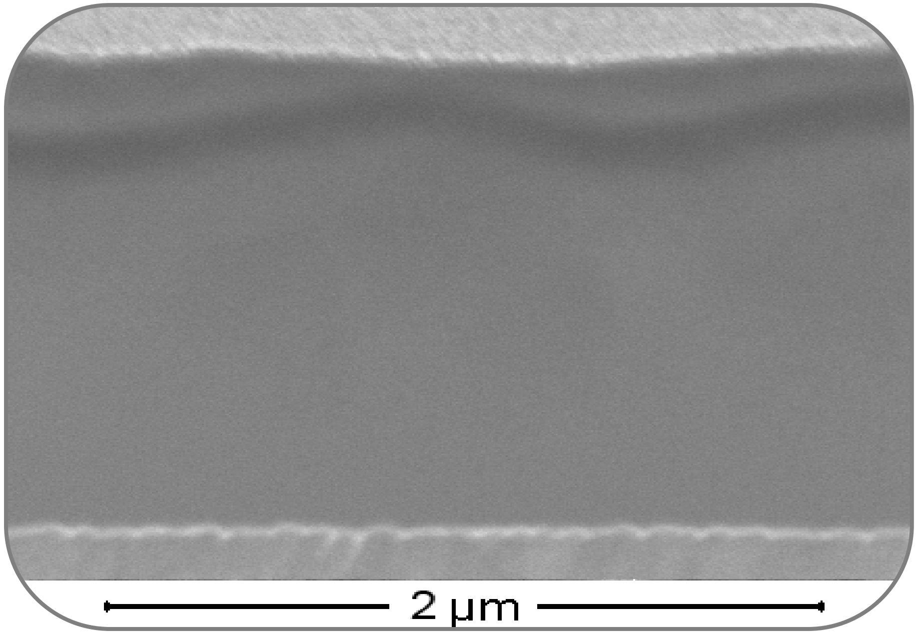
Amorphous Nb_2O_5 deposition recipe:

Ar flux:	O_2 flux:	Sputtering pressure:	I_{DC}:
3 sccm	7 sccm	$1 \cdot 10^{-2}$ mbar	0.5 A

Amorphous Nb_2O_5:



SEM



FIB SEM

Multilayer Nb/Nb_2O_5 coatings

combine:

high ductility of Nb with

excellent barrier properties of Nb_2O_5

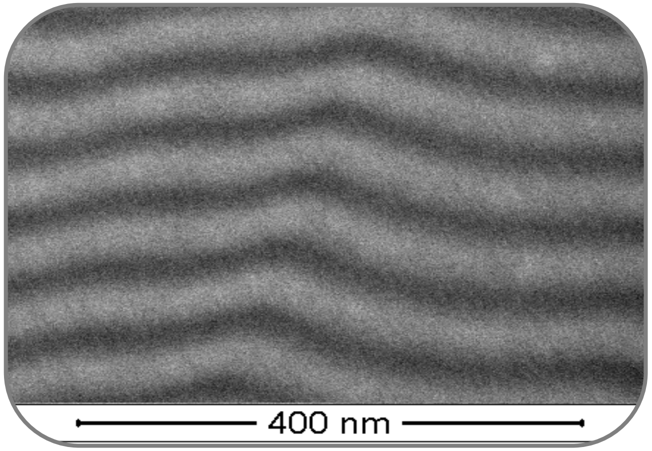
Best Nb/Nb_2O_5 multilayer recipe:

Ar flux:	O_2 flux:	Sputtering pressure:	Layer thickness:
3 sccm	0/7 sccm	$3 \cdot 10^{-3}$/ $1 \cdot 10^{-2}$ mbar	40/20 nm

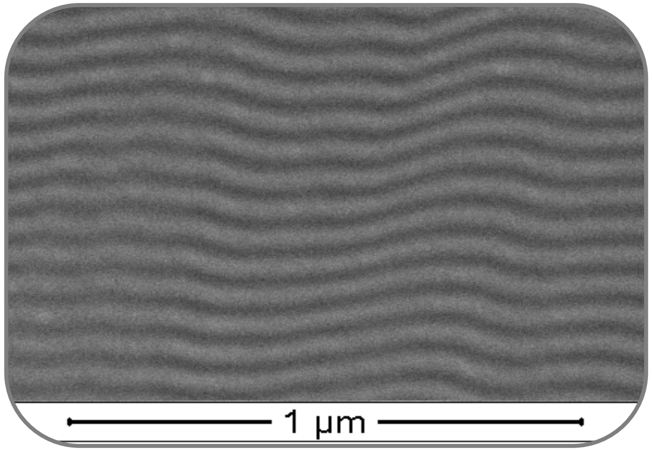
60 nm double-layer coatings showed **high corrosion resistance**

Niobium-niobium oxide multilayered coatings for corrosion protection of proton-irradiated liquid water targets for [^{18}F] production , H. Skliarova, M. Renzelli, O. Azzolini, D. de Felicis, E. Bemporad, Ri R. Johnson, V. Palmieri, *Thin Solid Films* 03/2015; 42.

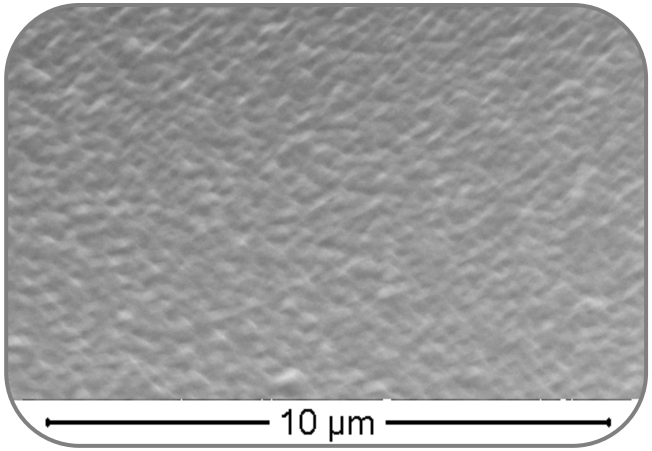
Thin Nb/Nb_2O_5 multilayer:



FIB SEM



FIB SEM



SEM

Nb-Ta, Nb-Zr and Ta-Zr

were co-deposited in different ratios in

order to find amorphous metallic coating

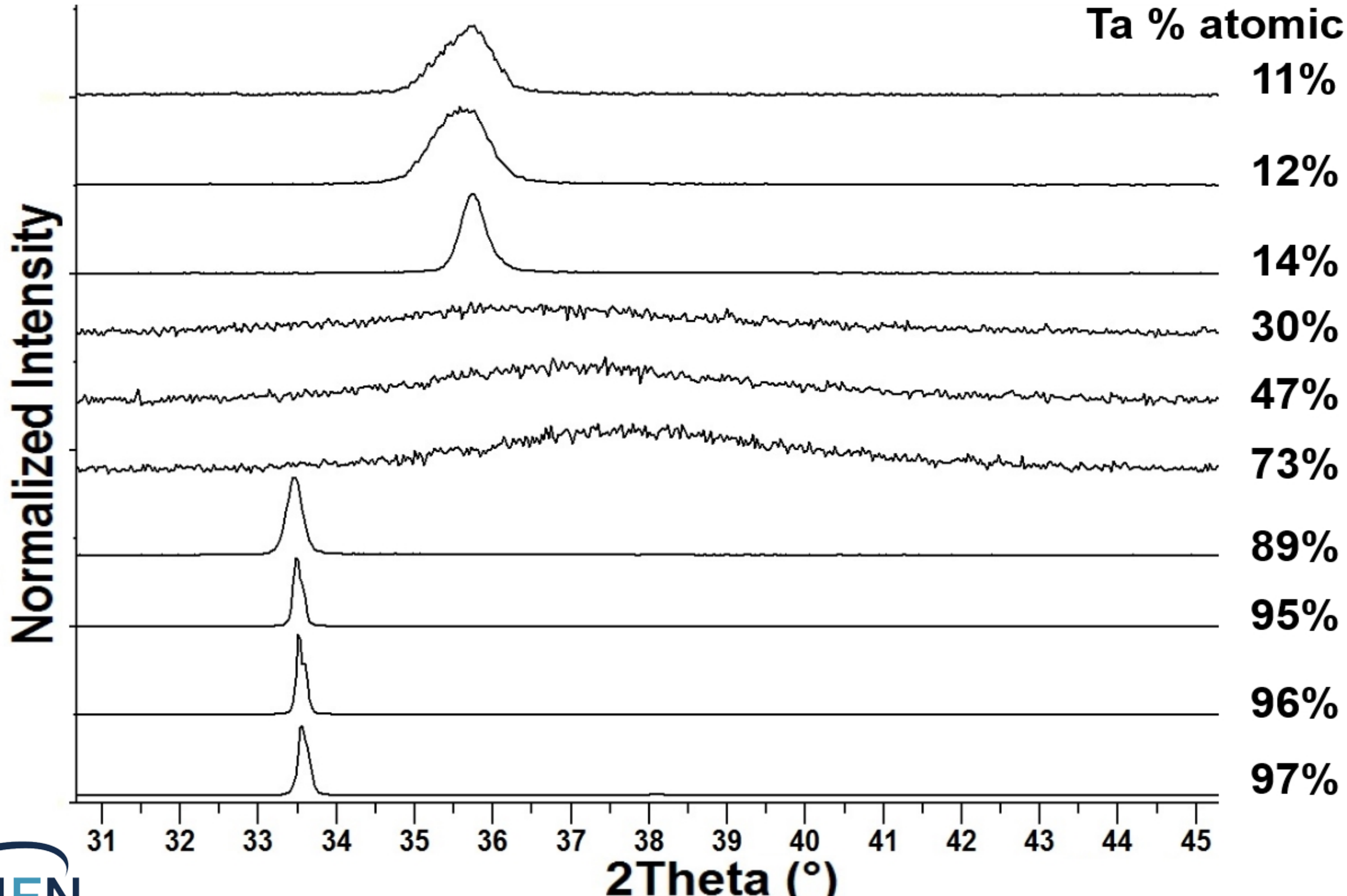
Sample-holder for co-deposition

Nb-Ta, Nb-Zr systems

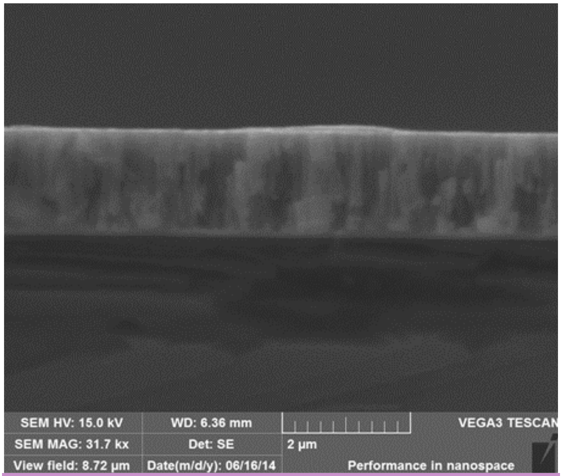
were resulting only in crystalline columnar structures

Co-sputtered amorphous Nb–Ta, Nb–Zr and Ta–Zr coatings for corrosion protection of cyclotron targets for [^{18}F] production , H. Skliarova, O. Azzolini, R. R. Johnson, V. Palmieri, *Journal of Alloys and Compounds* 08/2015; 639:488-495.

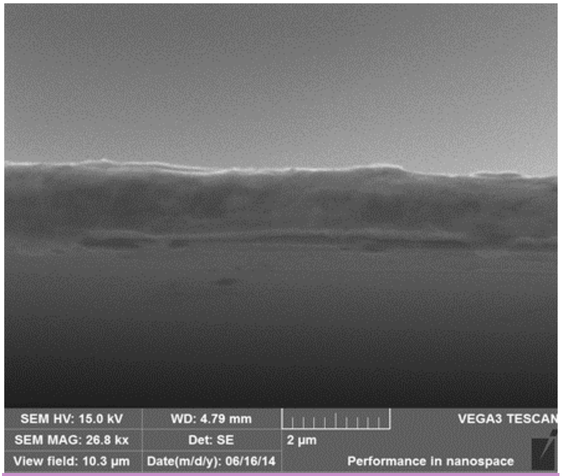
XRD Ta-Zr:


Ta % atomic
11%
12%
14%
30%
47%
73%
89%
95%
96%
97%
Normalized Intensity
31 32 33 34 35 36 37 38 39 40 41 42 43 44 45
2Theta (°)

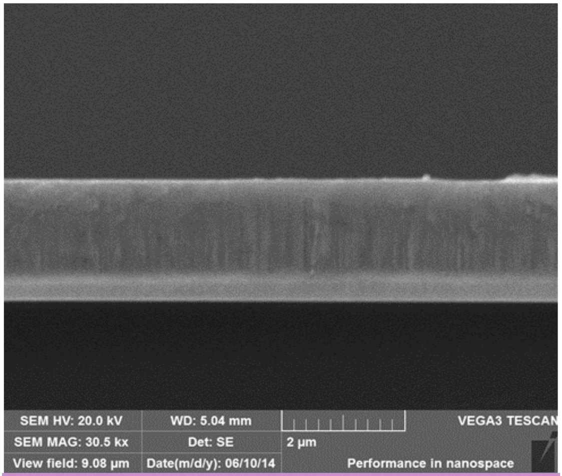
Sample-holder for co-deposition



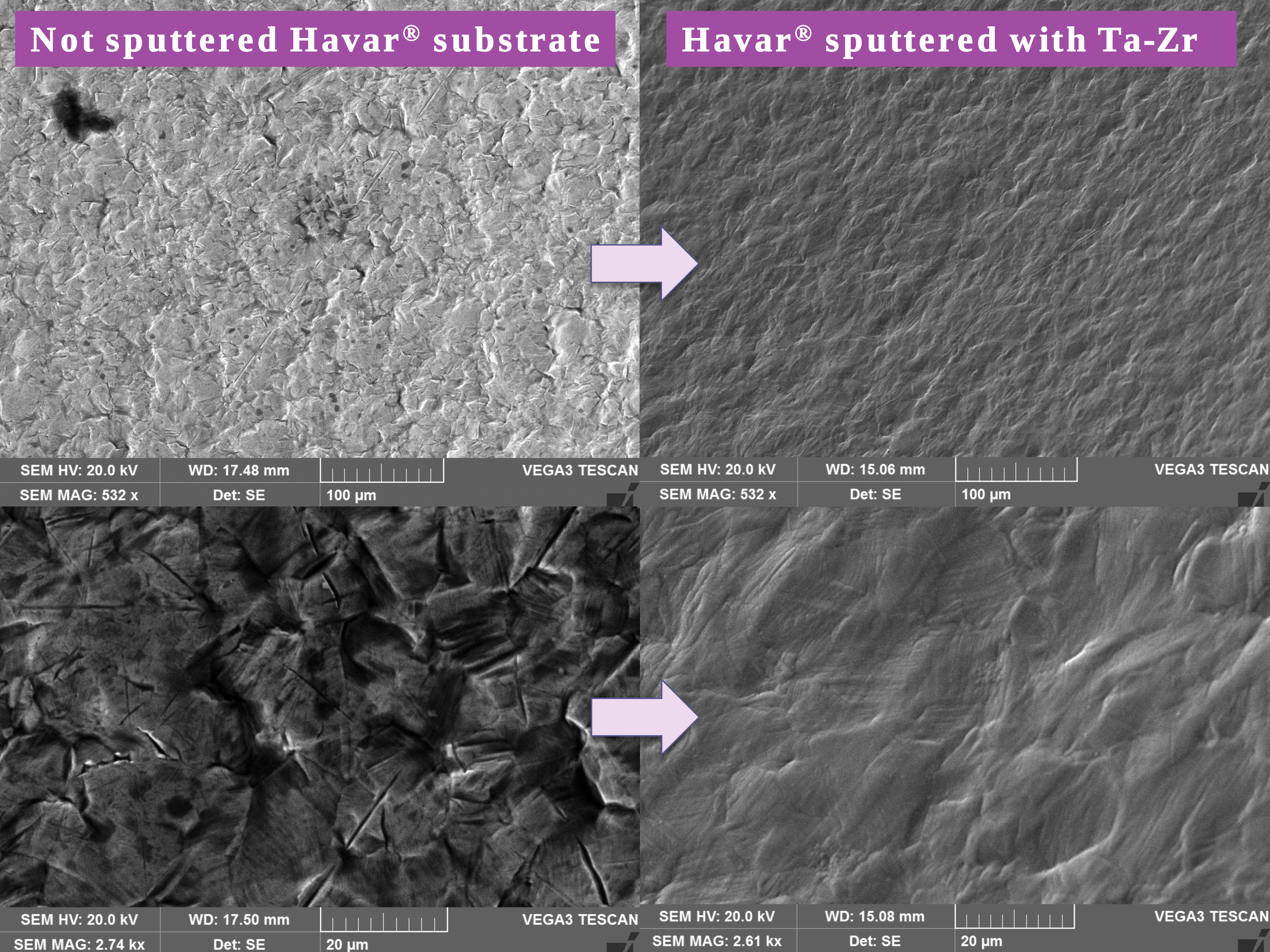
$Ta_{95}Zr_5$



$Ta_{50}Zr_{50}$



$Ta_{15}Zr_{85}$


Not sputtered Havar® substrate
Havar® sputtered with Ta-Zr
SEM HV: 20.0 kV
WD: 17.48 mm
VEGA3 TESCAN
SEM MAG: 532 x
Det: SE
100 μm
SEM HV: 20.0 kV
WD: 15.06 mm
VEGA3 TESCAN
SEM MAG: 532 x
Det: SE
100 μm
SEM HV: 20.0 kV
WD: 17.50 mm
VEGA3 TESCAN
SEM MAG: 2.74 kx
Det: SE
20 μm
SEM HV: 20.0 kV
WD: 15.08 mm
VEGA3 TESCAN
SEM MAG: 2.61 kx
Det: SE
20 μm

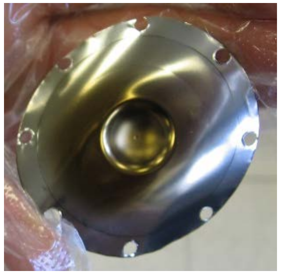
Recipes used for beam window coating:

DC-biased sputtering of Nb

Reactive sputtering of amorphous Nb_2O_5

Nb/Nb_2O_5 thin multilayers

Amorphous Ta-Zr



Thank you
for your attention!