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 $^{18}\text{O}\text{H}_2\text{O}$ target for production of $^{18}\text{F}^-$.

FDG is the most used radioisotope in PET.
Proton irradiation cause **water radiolysis**

\[ \text{H}_2\text{O} \rightarrow \text{H}_2, \text{O}_2, \text{H}_2\text{O}_2, \text{OH}, \text{H}, \text{e}^-_{\text{aq}}, \text{HO}_2, \text{O}_2^-, \text{HO}_2^-, \text{OH}^-, \text{H}^+, \ldots \]

Proton irradiated water is **extremely corrosive!**
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
Substrate holders

- grounded
- liquid nitrogen-cooled
- heated
- water-cooled
Analyzing technique

Acid porosity test:

- HCl solution
- Nb
- Al
- quartz
Analyzing technique

**Acid porosity test:** 10% HCl, 30°C, 10 min
Analyzing technique: SEM, FIB SEM
Analyzing technique

X-ray diffractometry

![Graph showing intensity counts vs. 2Theta degrees with peaks indicating crystalline and amorphous niobium oxide.]

- Red line: Niobium oxide amorphous
- Blue line: Nb crystalline
Coating systems investigated:

- Nb
- Nb$_2$O$_5$
- Nb/Nb$_2$O$_5$ 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

Temperature influence:
Best Nb deposition recipe:

- $P_{Ar}: 3 \cdot 10^{-3} \text{ mbar}$
- $I_{DC}: 0.5 \text{ A}$
- Substrate bias: $-80 \text{ V}$
- Substrate temperature: no heating/ no cooling ($\sim 250^{\circ}\text{C}$)
DC-biased MS of Nb:

SEM

SEM

FIB SEM
Reactive sputtering of $\text{Nb}_2\text{O}_5$ :

**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

Amorphous \( \text{Nb}_2\text{O}_5 \) deposition recipe:

- **Ar flux:** 3 sccm
- **\( \text{O}_2 \) flux:** 7 sccm
- **Sputtering pressure:** \( 1 \cdot 10^{-2} \) mbar
- **\( I_{\text{DC}} \):** 0.5 A
Amorphous $\text{Nb}_2\text{O}_5$:
Multilayer Nb/Nb$_2$O$_5$ coatings combine:

high ductility of Nb with excellent barrier properties of Nb$_2$O$_5$
Best Nb/Nb$_2$O$_5$ multilayer recipe:

**Ar flux:** 3 sccm

**O$_2$ flux:** 0/7 sccm

**Sputtering pressure:** $3 \cdot 10^{-3}/1 \cdot 10^{-2}$ mbar

**Layer thickness:** 40/20 nm

60 nm double-layer coatings showed high corrosion resistance


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Thin Nb/Nb$_2$O$_5$ 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.
XRD Ta-Zr:

Ta % atomic
11%
12%
14%
30%
47%
73%
89%
95%
96%
97%
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
Recipes used for beam window coating:

- DC-biased sputtering of Nb
- Reactive sputtering of amorphous Nb$_2$O$_5$
- Nb/Nb$_2$O$_5$ thin multilayers
- Amorphous Ta-Zr
Thank you for your attention!