Effects of deposition parameters on superconducting NbTiN thin films for use in SRF multilayer structures

11th International Workshop on Thin Films and New Ideas for Pushing the Limits of RF Superconductivity - TFSRF2024, Université Paris-Saclay, France

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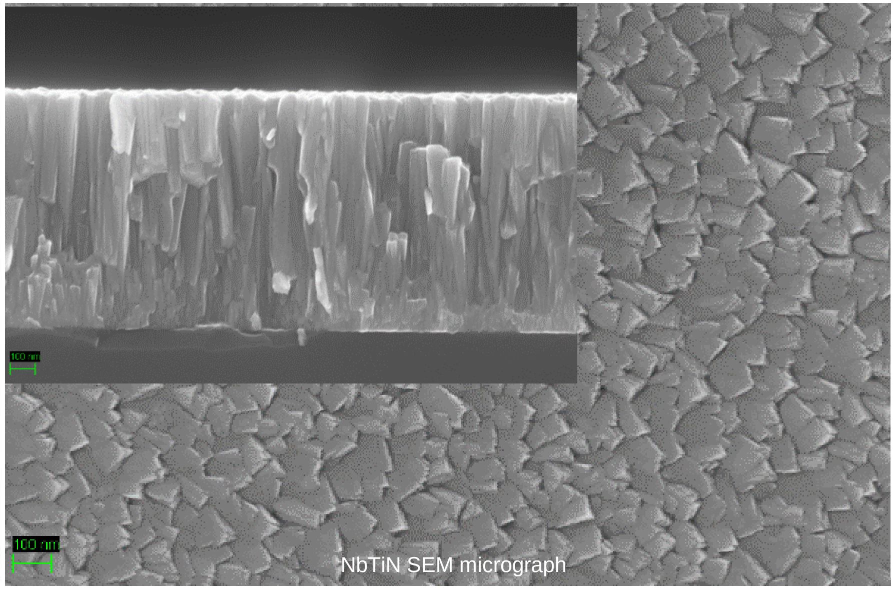
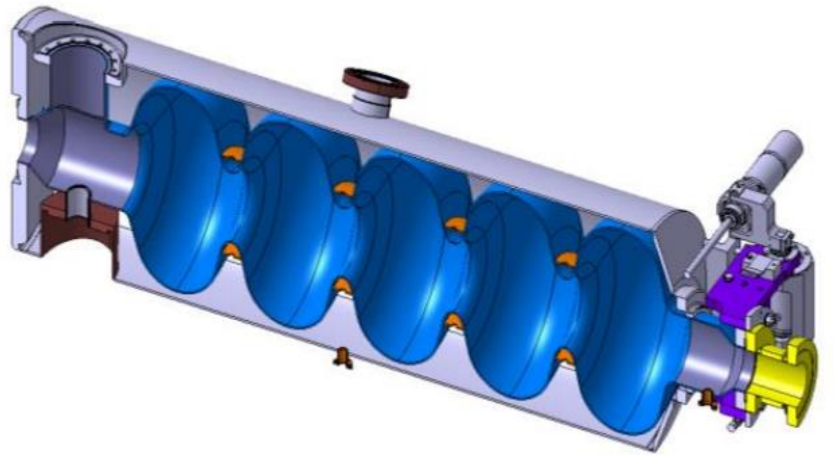
Industrial PVD machine



NbTiN SEM micrograph


Elliptical SRF Cavity

Introduction

Particle Accelerators

Key Component

⇓

Radiofrequency Cavities (Cu)
Dis: Much of the RF power dissipates in cavity walls

⇓

Superconducting Radiofrequency Cavities (Bulk Nb)
Adv: - Most of the RF power transfers to the particle beam
Dis: - Poor thermal conductor, expensive, difficult to process

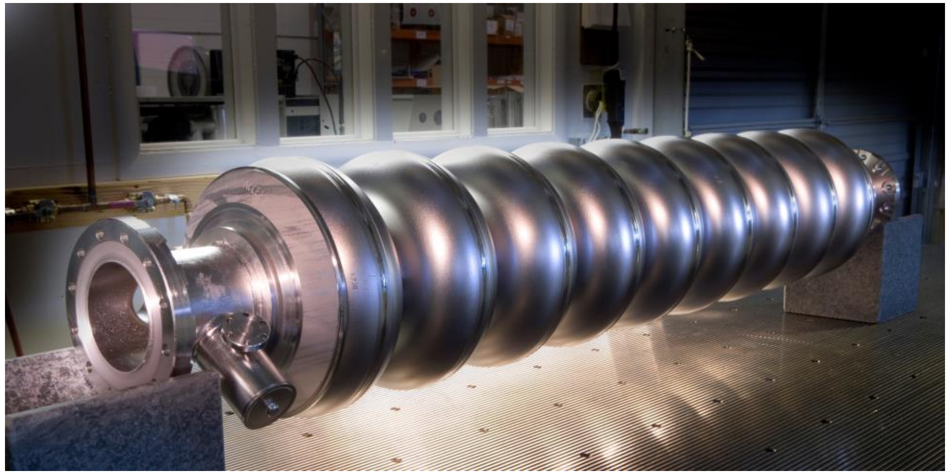
⇓

Thin Film (Nb) based SRF Cavities

⇓

Alternative Superconducting Thin Films
$NbN, NbTiN, Nb_3Sn$, V_3Si, Nb_3Al, Mo_3Re, MgB_2, etc

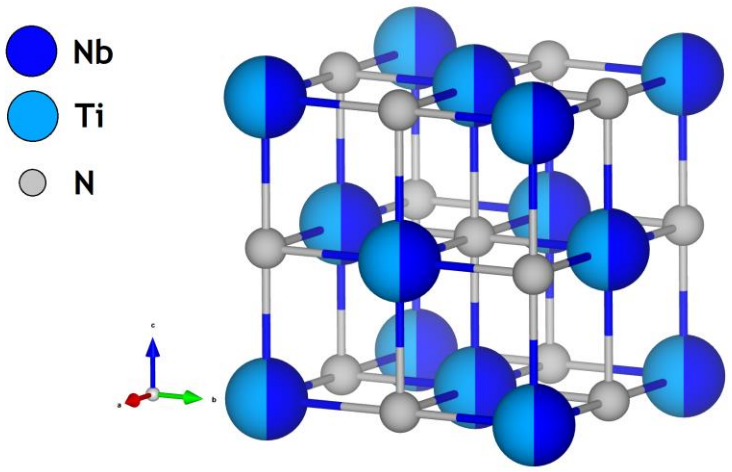
The European XFEL **(**Hard X-ray Free Electron Laser) **[DESY campus in Hamburg]**[1]



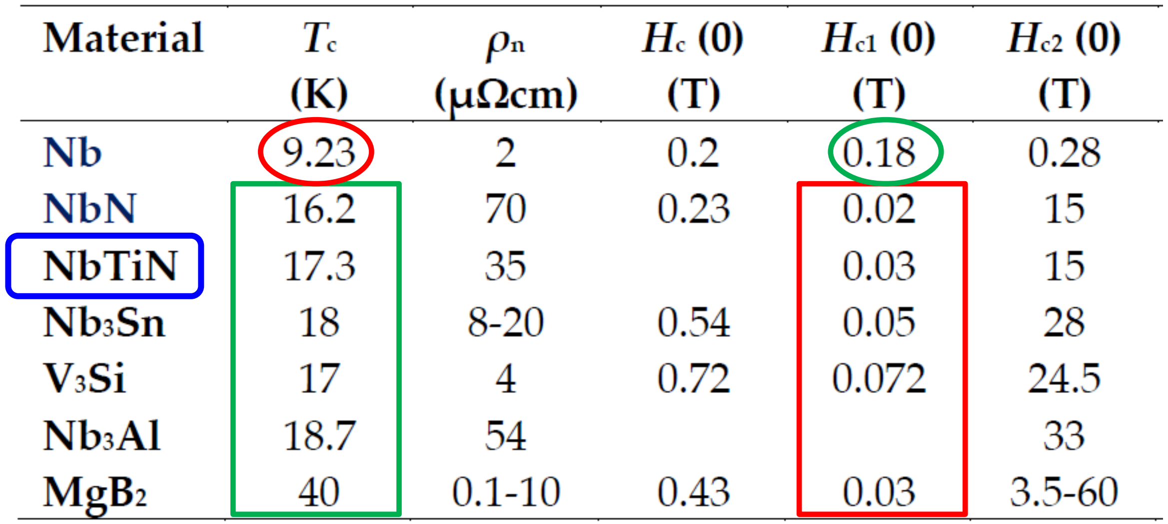
Nine-cell Nb SRF Cavity **[*Fermilab*]**[2]

1. https://xfel.desy.de/
2. https://td.fnal.gov/srf-rd/

Candidate Materials for SRF Cavities



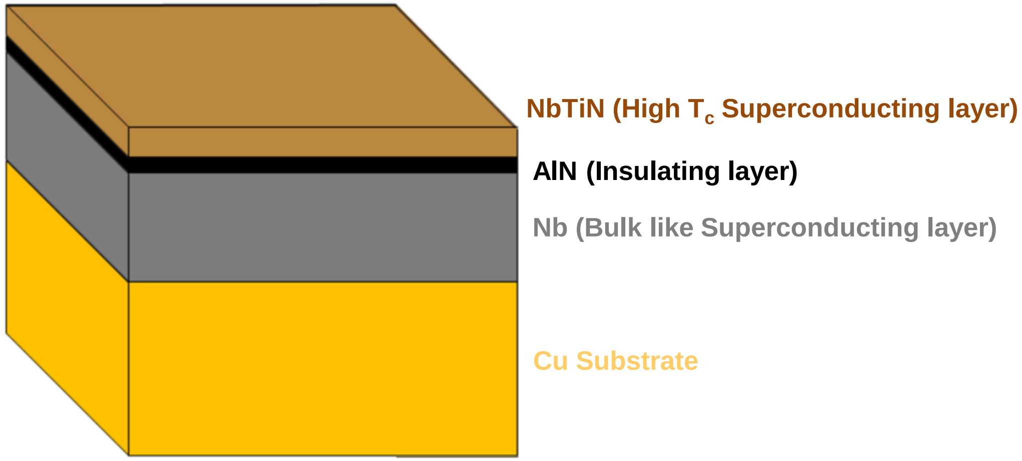
Crystal system: FCC (NaCl – type)
Lattice parameter =4.3600 Å
Space Group: F m -3 m



Material	T_c (K)	ρ_n (μΩcm)	H_c (0) (T)	H_{c1} (0) (T)	H_{c2} (0) (T)
Nb	9.23	2	0.2	0.18	0.28
NbN	16.2	70	0.23	0.02	15
NbTiN	17.3	35		0.03	15
Nb_3Sn	18	8-20	0.54	0.05	28
V_3Si	17	4	0.72	0.072	24.5
Nb_3Al	18.7	54			33
MgB_2	40	0.1-10	0.43	0.03	3.5-60

- **High Transition temperature (T_c)** – Reduced refrigeration costs and high Q factors
- **Low lower critical magnetic field (H_{c1}) –** Lower accelerating gradients
- **Superconducting-Insulator-Superconducting (SIS) multilayer structures -** exploiting the advantages associated with high T_c SC without being limited by their low H_{c1} (*method of providing magnetic shielding to an underlying superconducting layer*)

Ref: Valente-Feliciano, Anne Marie, Superconducting RF materials other than bulk niobium: A review; 2016 Supercond. Sci. Technol. 29 113002
Ref: Stewart Leith; Development of Novel Superconducting Thin Films for use in Superconducting Radio Frequency Cavities; Siegen, Univ., Diss., 2021
Ref: Alex Gurevich; Enhancement of rf breakdown field of superconductors by multilayer coating; February 2006, Applied Physics Letters 88(1):012511 - 012511-3



SIS Scheme

Experimental: NbTiN Thin Film Sputter Deposition

HiPIMS (High Power Impulse Magnetron Sputtering) technique is used to deposit NbTiN films

Parameters	DCMS	HiPIMS
Peak power density	10^3 W/cm^2	$>$ 10^3 W/cm^2
Peak current density	0.01 – 0.1Acm^{-2}	0.01 – $10^3 Acm^{-2}$
Discharge voltage	500 V	500 – 1300 V
Plasma density	$10^{16}/m^3$	10^{25} – $10^{16}/m^3$
Degree of metal ionization	< 5 %	30 – 100 %
Average metal ion energy	2- 10 eV	50 – 100 eV
Deposition rate	1	0.2 – 0.8

CC800 Industrial Coating Machine

NbTi (67/33 .%at) Target size: 100 $\times$88 $\times$ 9 mm^3
Process gas: Ar
Target to substrate distance: 55 mm
Base pressure: 0.05 mPa
Substrate: Silicon

Heating
Substrate etching
Target sputter cleaning
Coating

Ref: H. Zhang, J. S. Cherng, and Q. Chen, “Recent progress on high power impulse magnetron sputtering (HiPIMS): The challenges and applications in fabricating VO2 thin film,” AIP Advances, vol. 9, no. 3, Mar. 2019
Ref: J. T. Gudmundsson, “Physics and technology of magnetron sputtering discharges,” Plasma Sources Science and Technology, vol. 29, no. 11, p. 113001, Nov. 2020.

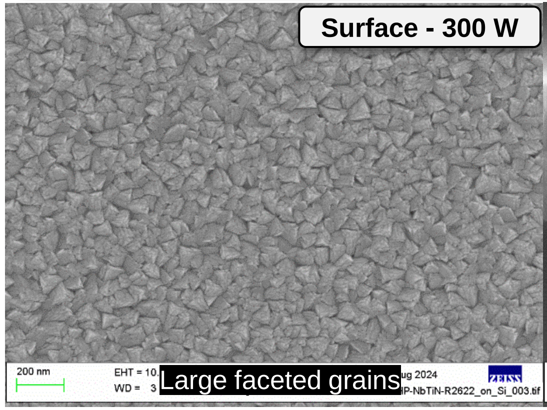
Results and Discussion

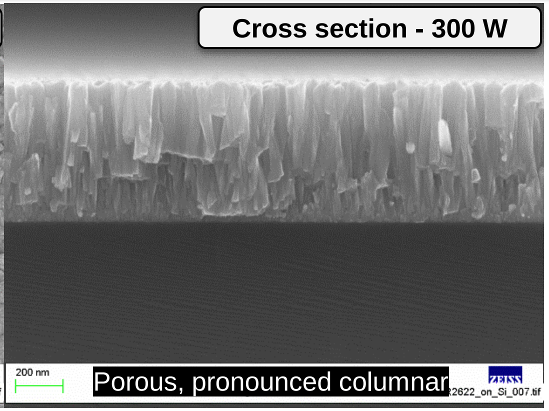
- **Goal:** Optimization (*for High T_c and entry field*) of HiPIMS deposited NbTiN thin films
- **Method:** Studying the influence of process parameters on thin film deposition

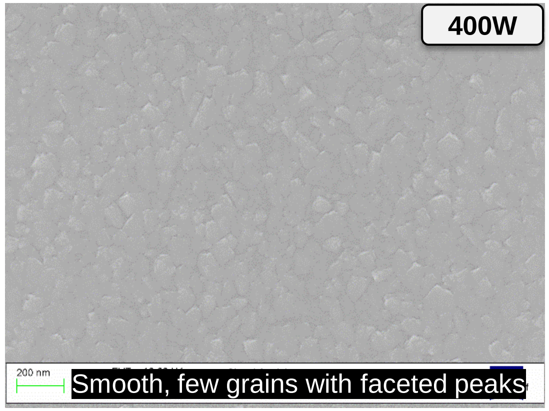
  1. **Cathode power study**
  2. **Deposition pressure study**
  3. **Duty cycle study**
  4. **Substrate bias study**

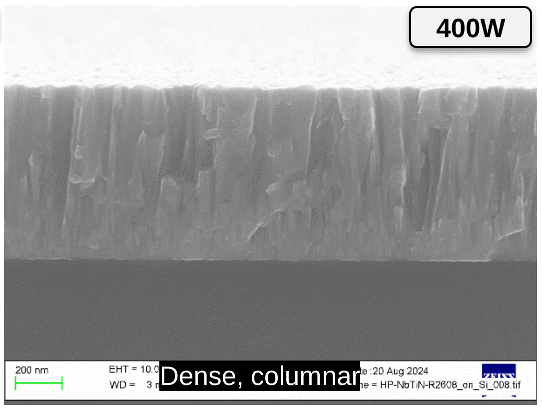
1. Cathode Power

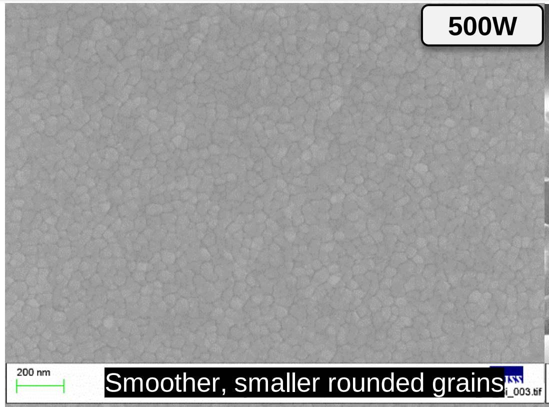
Deposition pressure = 2.0 Pa, N_2%= 10, Substrate bias = 50V, Duty cycle = 10%, Substrate temperature = 150 ℃
Cathode power = 300, 400, 500, and 600 W

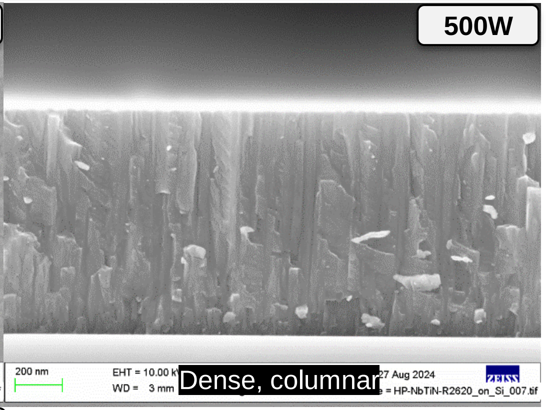

Surface - 300 W
Large faceted grains

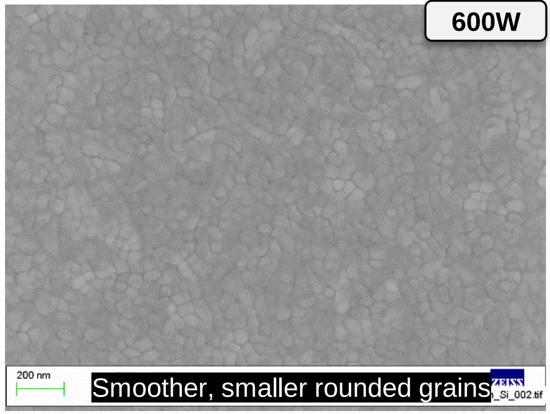

Cross section - 300 W
Porous, pronounced columnar

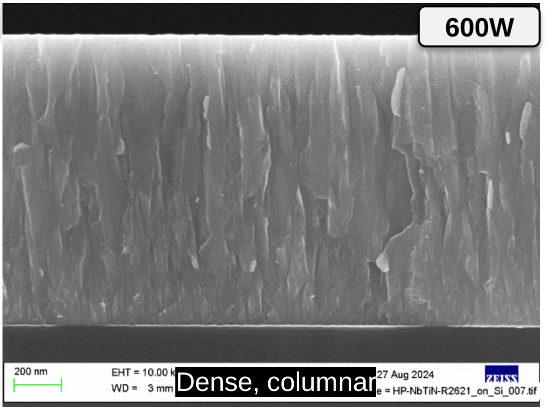

400W
Smooth, few grains with faceted peaks


400W
Dense, columnar


500W
Smoother, smaller rounded grains


500W
Dense, columnar


600W
Smoother, smaller rounded grains


600W
Dense, columnar

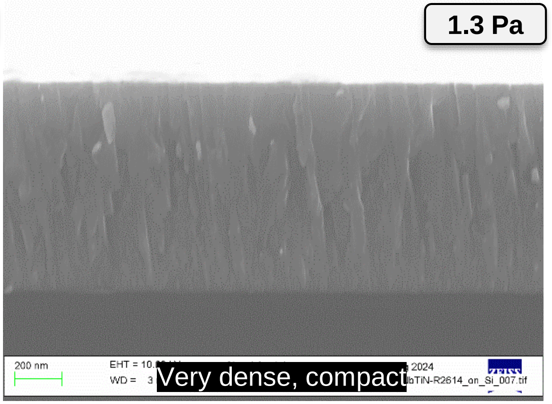
Cathode Power ↑ energy of the gas ions bombarding the target surface ↑ sputtering yield ↑ deposition rate ↑ nucleation growth and surface diffusion ↓ → **grain size ↓ film density ↑**

2. Deposition Pressure Study

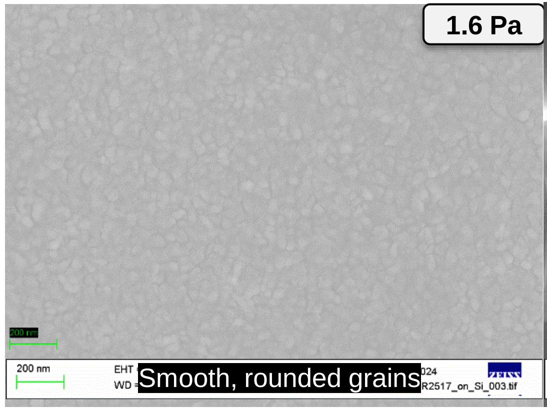
Cathode power = 400W, N_2%= 10, Substrate bias = 50V, Duty cycle = 10%, Substrate temperature = 150 ℃
Deposition pressure =1.3, 1.6, 2.0, 2.8 Pa


1.3 Pa

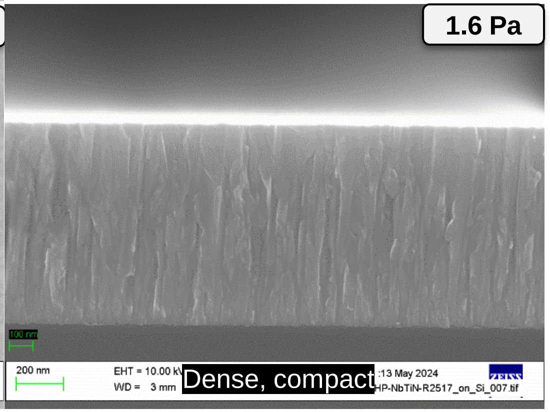
Very smooth, rounded grains


1.3 Pa

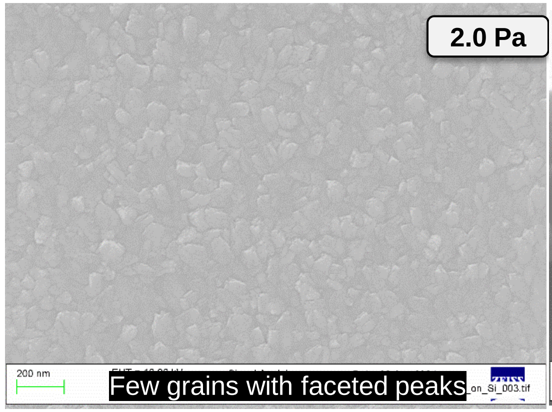
Very dense, compact


1.6 Pa

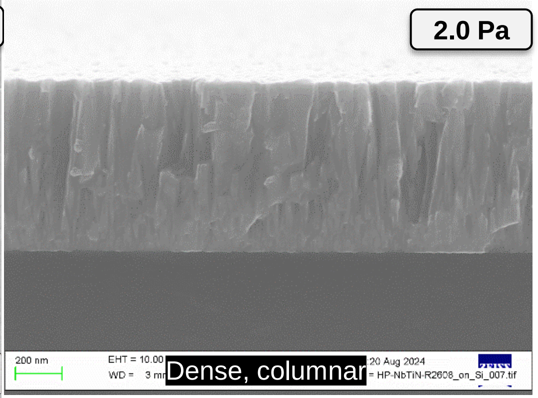
Smooth, rounded grains


1.6 Pa

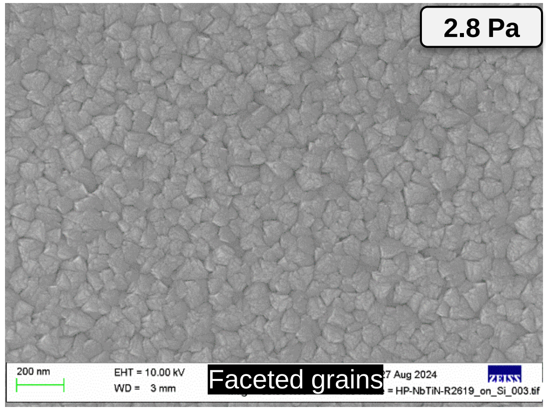
Dense, compact


2.0 Pa

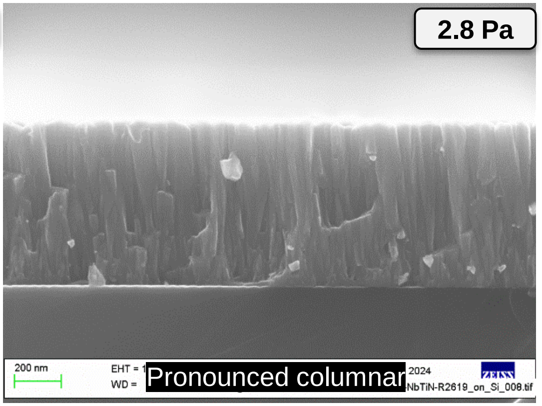
Few grains with faceted peaks


2.0 Pa

Dense, columnar


2.8 Pa

Faceted grains


2.8 Pa

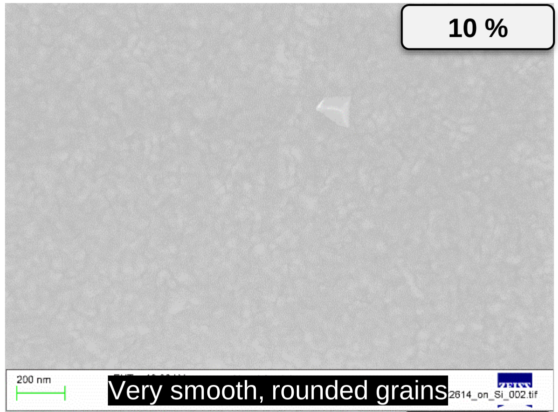
Pronounced columnar

Deposition pressure ↓ mean free path ↑ collision probability ↓ → particles arrive substrate surface with high energy → surface diffusion ↑ **film density ↑**

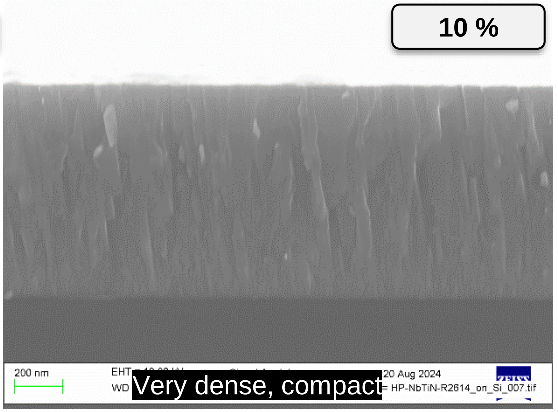
3. Duty Cycle Study

Cathode power = 400W, N_2%= 10, Deposition pressure = 1.3 Pa, Substrate bias = 50 V, D. P = 1.3 Pa, Substrate temperature = 150 ℃

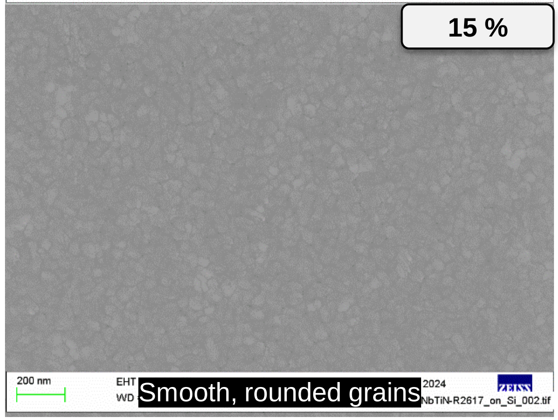
Duty cycle = 10, 15, and 20%



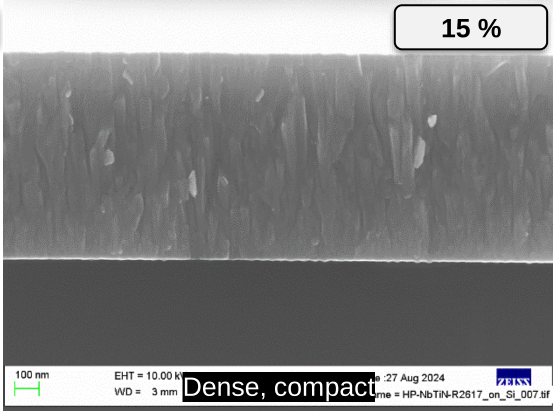
Very smooth, rounded grains



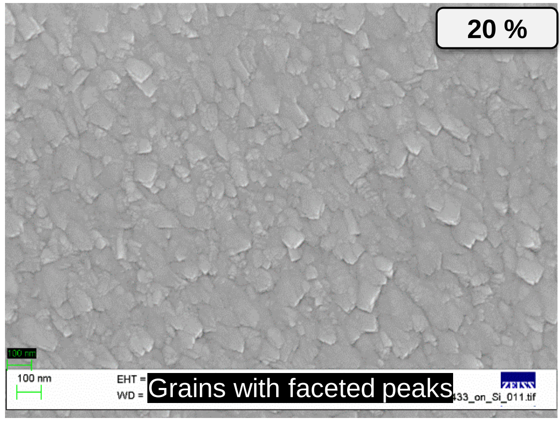
Very dense, compact



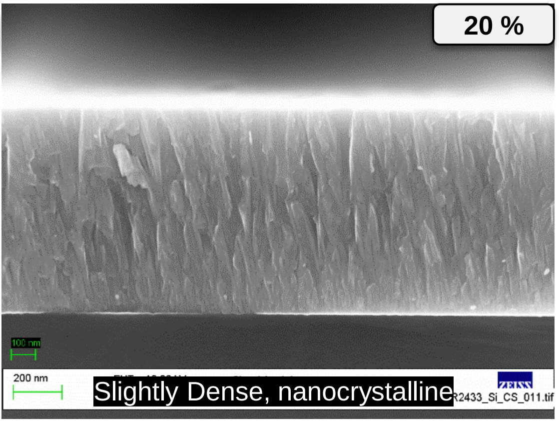
Smooth, rounded grains



Dense, compact



Grains with faceted peaks

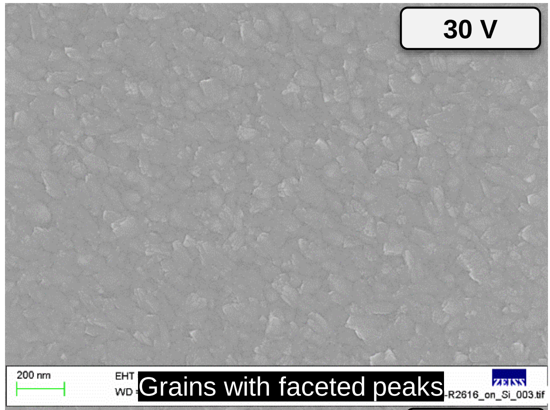


Slightly Dense, nanocrystalline

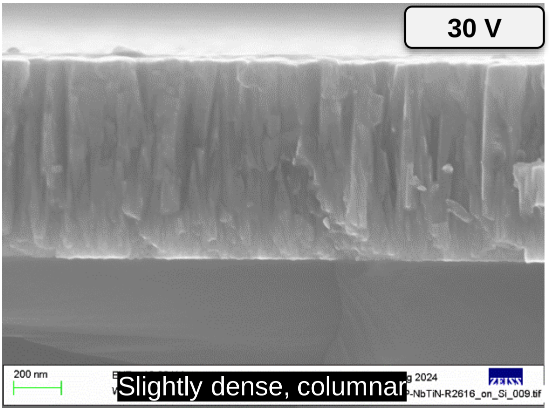
Duty Cycle ↓ peak power density ↑ plasma density ↑ ionization of sputtered species ↑ ionized species arrive substrate surface with high energy → surface diffusion ↑ **film density** ↑

4. Substrate Bias Study

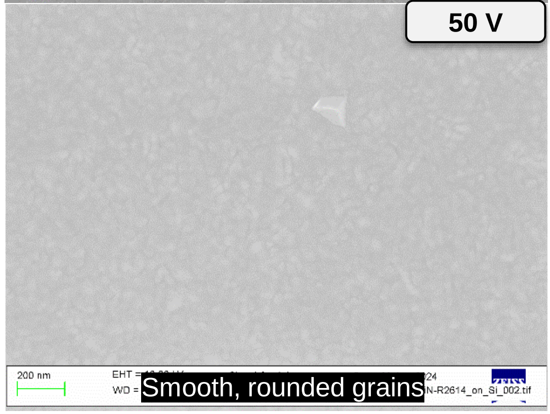
Cathode power = 400W, N_2% = 10, Deposition pressure = 1.3 Pa, Duty cycle = 10%, Substrate temperature = 150 ℃
Substrate bias = 30, 50, 75, and 100 V


30 V

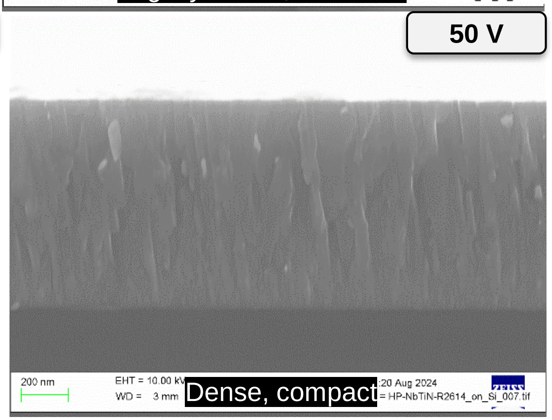
Grains with faceted peaks


30 V

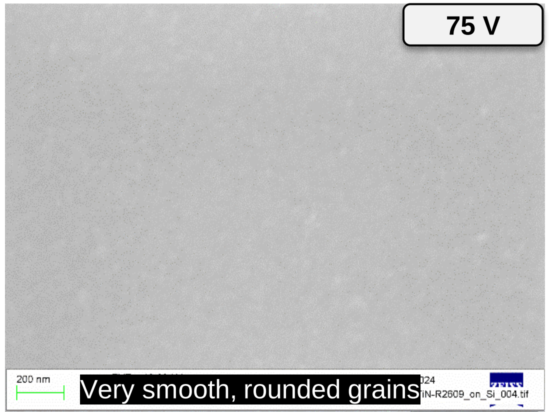
Slightly dense, columnar


50 V

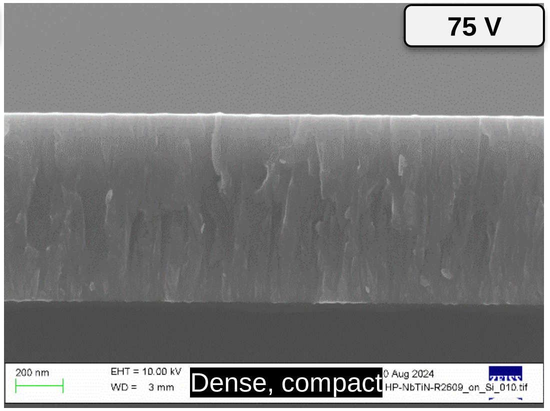
Smooth, rounded grains


50 V

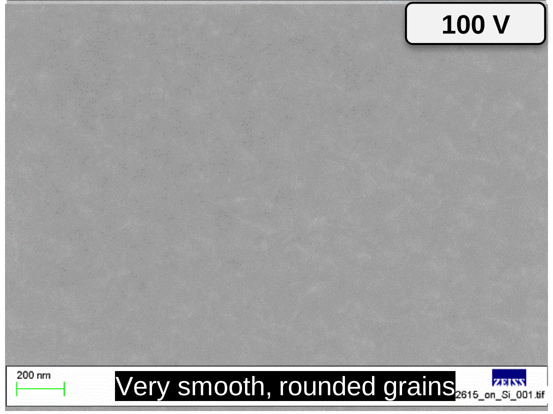
Dense, compact


75 V

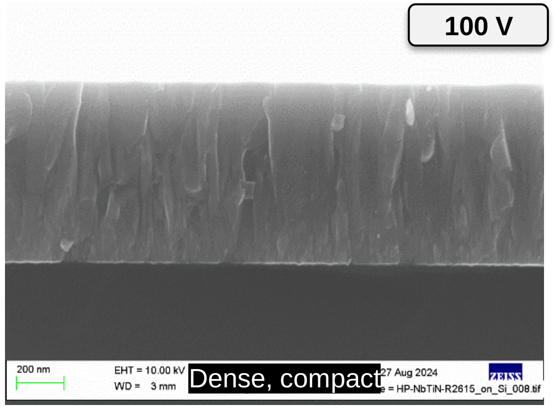
Very smooth, rounded grains


75 V

Dense, compact


100 V

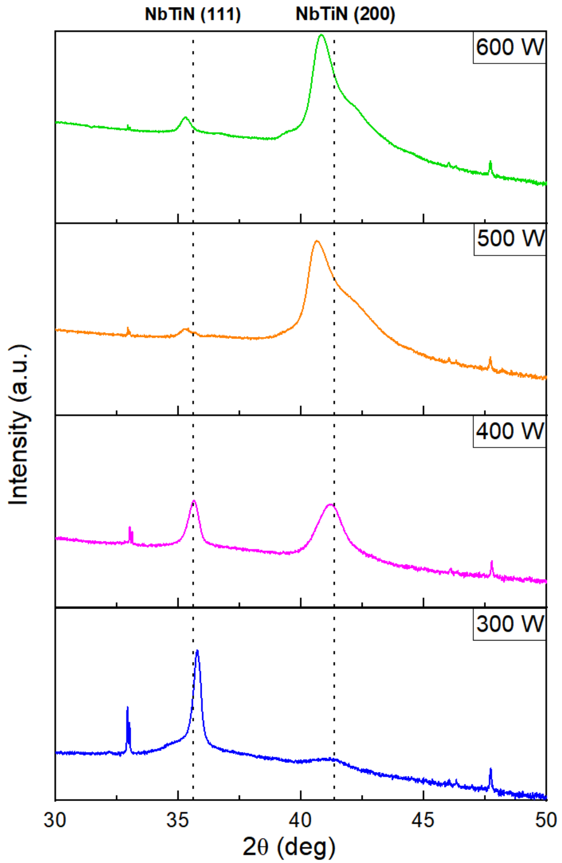
Very smooth, rounded grains


100 V

Dense, compact

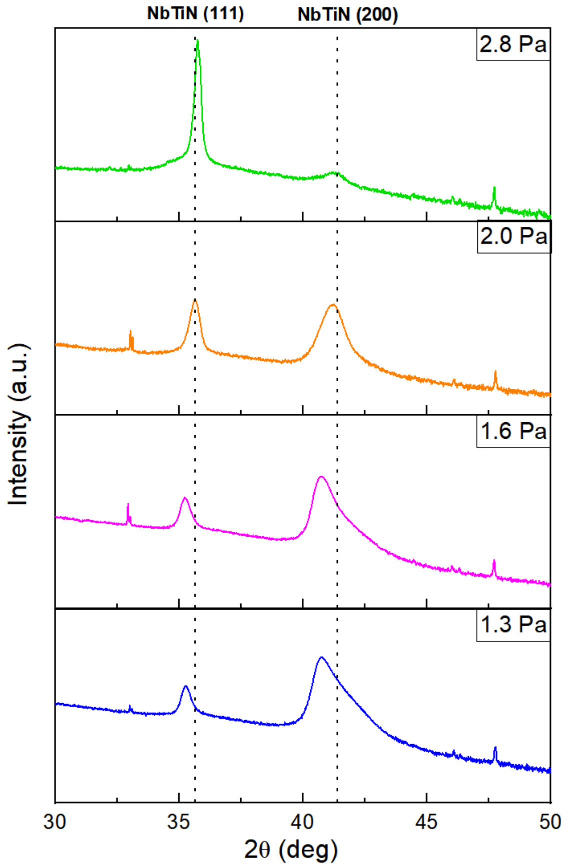
Substrate Bias ↑ acceleration of ions approaching the film surface ↑ → higher-energy ion bombardment → surface mobility and diffusion ↑ **film density** ↑

XRD Spectra



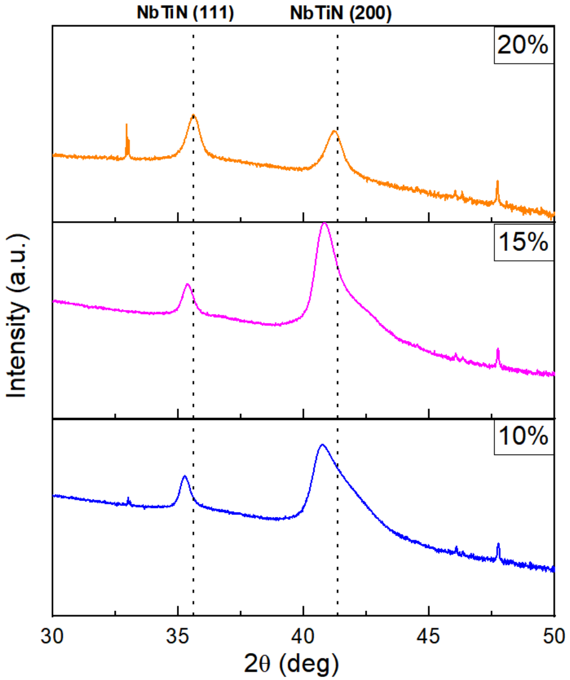
Cathode power study

Films at **High C.P** tend to grow with NbTiN (200)
C.P ↑ → a transition from (111) to (200)
At higher C.P → peak broadening
At higher C.P → a negative shift (compressive stress)



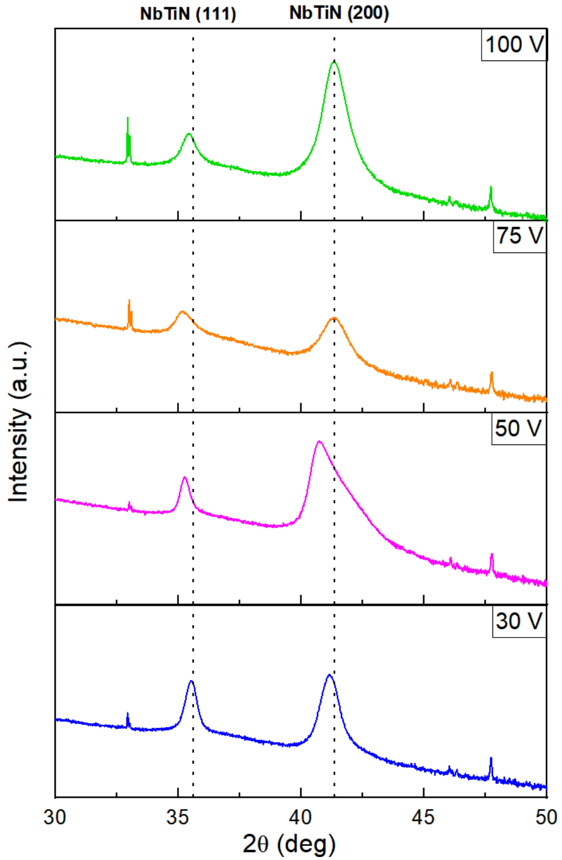
Deposition pressure study

D.P ↑ → transition from (200) to (111)
At lower D.P → a negative shift (compressive stress)
At higher D.P → lower film stress due to disjointed columnar structure – stress relaxation



Duty cycle study

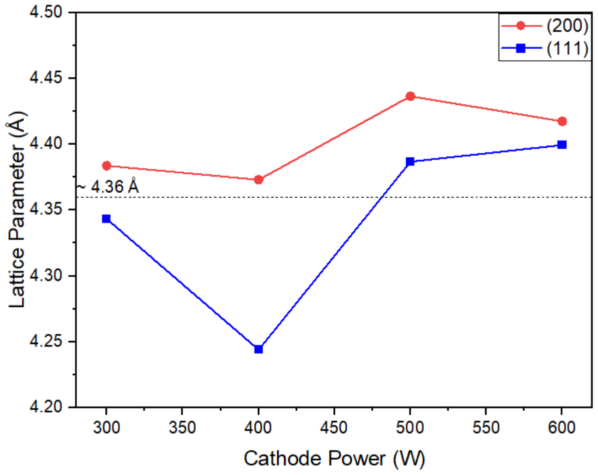
Films at **lower DCyc** tend to grow with NbTiN (200)
At lower DCyc → a negative shift (compressive stress)
At lower DCyc → higher film stress due to higher film density



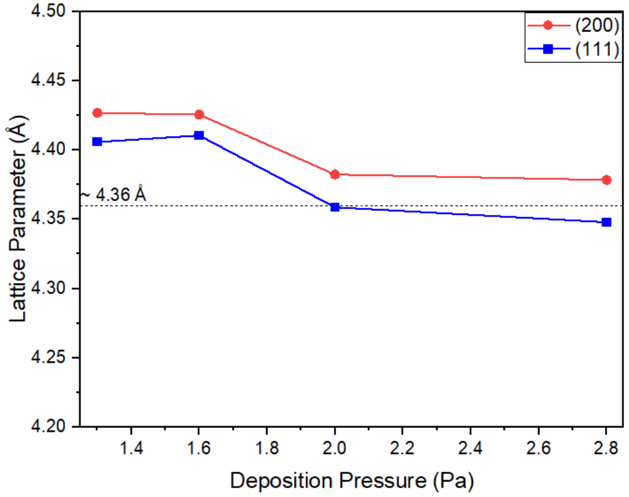
Substrate bias study

All films have a high presence of NbTiN (200)
Only At 50 V → a negative shift (compressive stress)

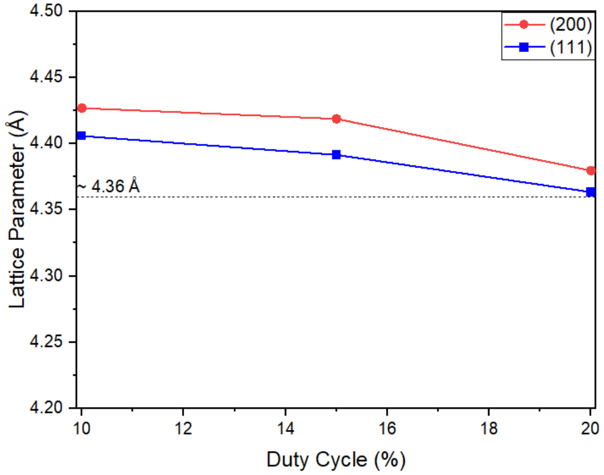
Lattice Parameters



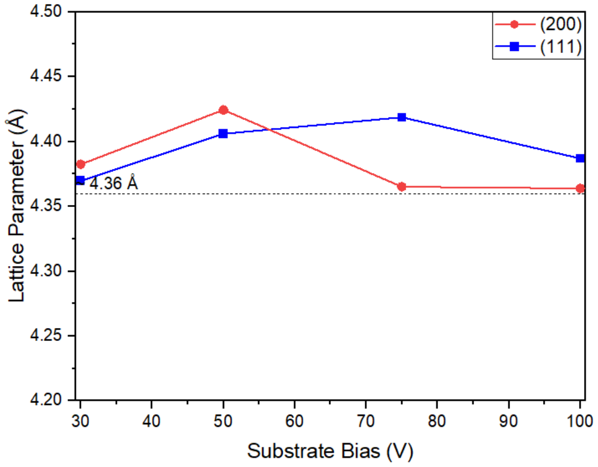
Cathode power study



Deposition pressure Study



Duty cycle Study

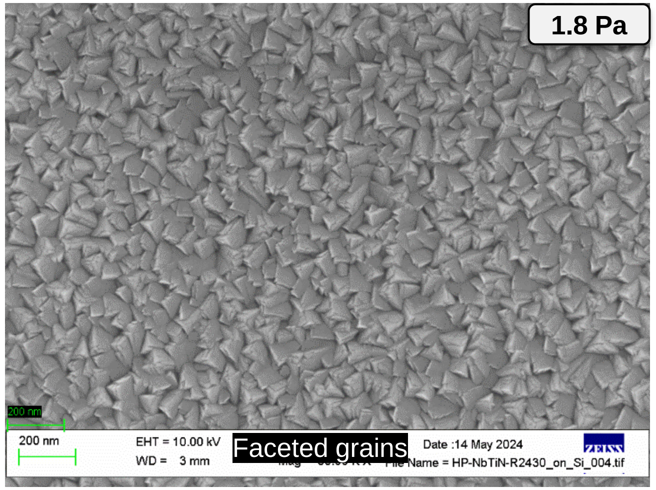


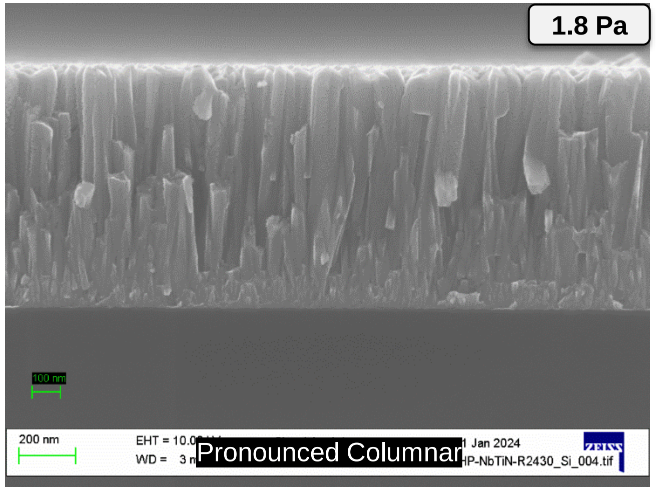
Substrate Bias study

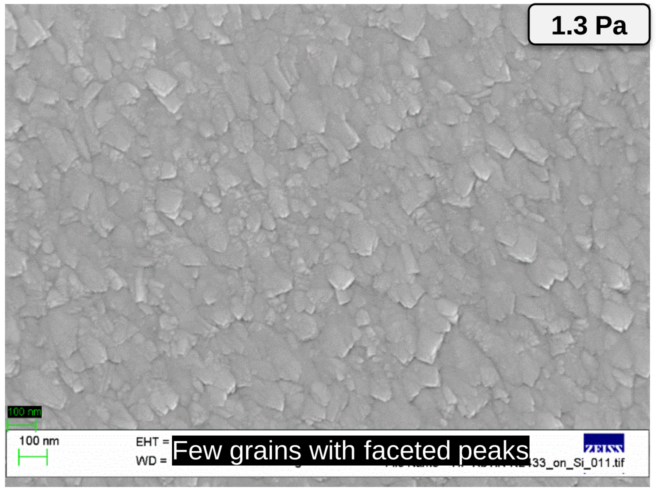
- The lattice parameter decreases with increasing deposition pressure and duty cycle
- The films with less stress state exhibit the lattice parameter value near the reference value for NbTiN ~ 4.360 Å
- Thermal stresses (thermal expansion coefficients) and intrinsic stresses (crystallographic flaws and lattice mismatch) →

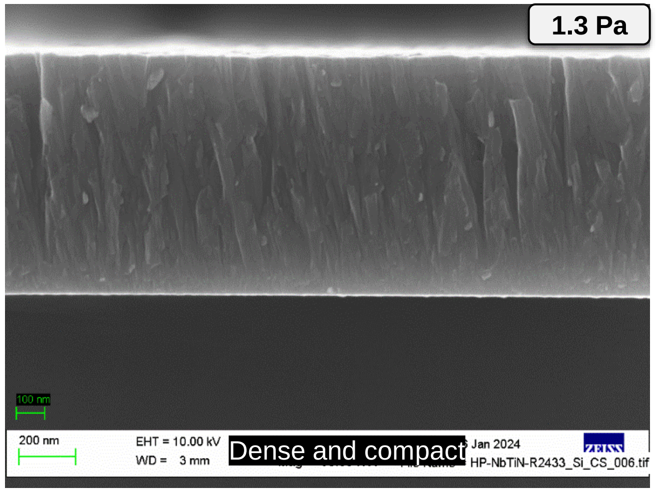
  Cracking or poor adhesion

T_c Variation with Deposition Pressure









T_c= 12.96K

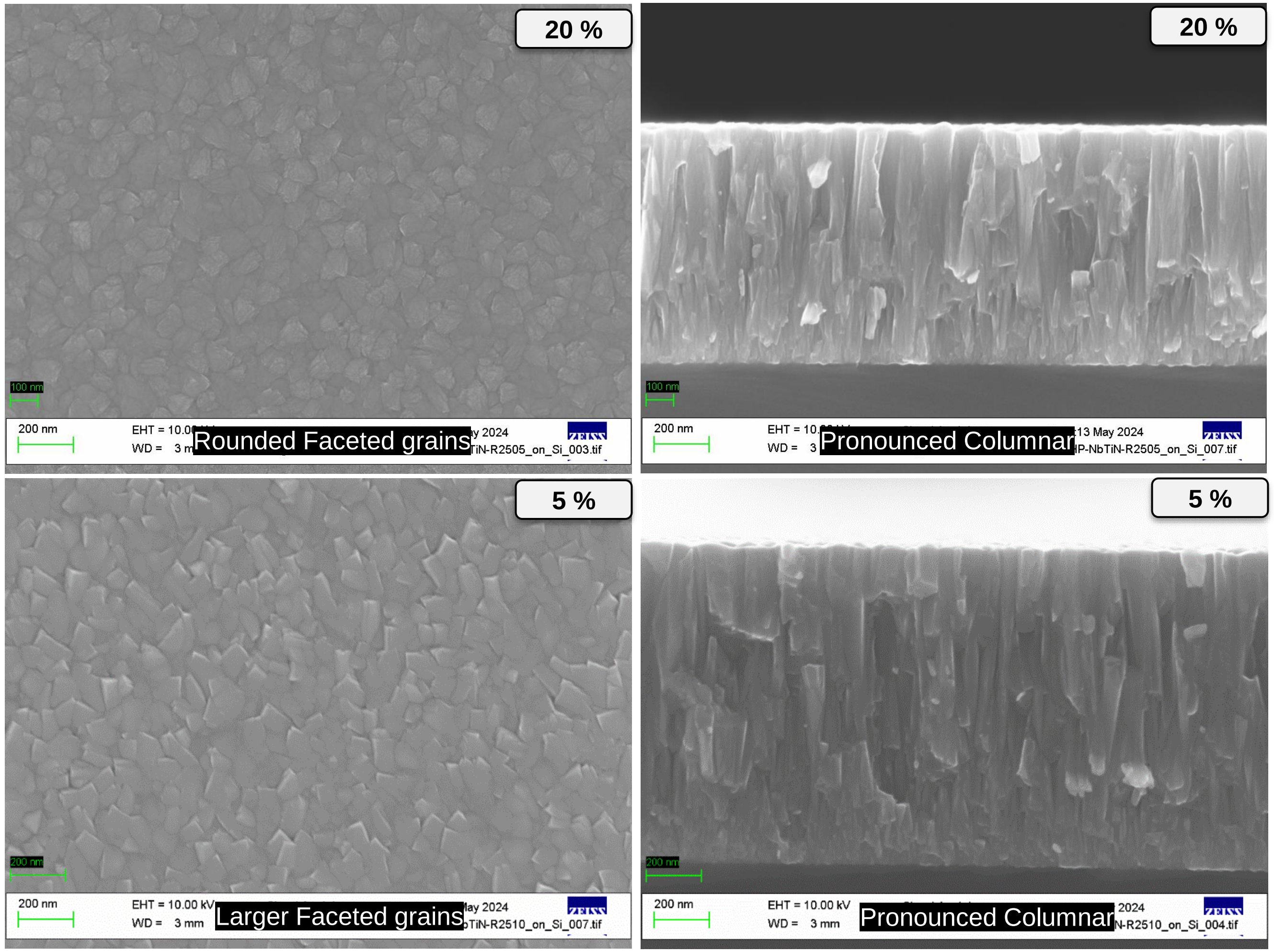
Film with a **higher NbTiN (111)** texture relative to the (200) texture

Cathode power = 400 W,
Substrate bias = 50 V,
N2% = 10,
Duty cycle = 20 %
Substrate temperature = 150 ℃

T_c= 14.03 K

Film with a **higher NbTiN (200)** texture relative to the (111) texture

T_c Variation with Nitrogen Flow



T_c= 12.63 K

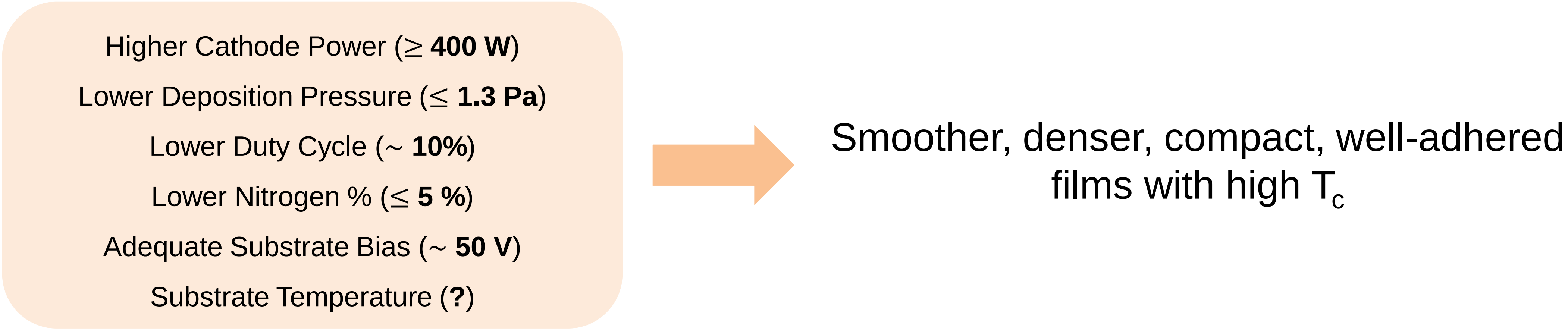
Film with a **higher NbTiN (111)** texture relative to the (200) texture

Cathode Power = 400 W,
Substrate bias = 50 V,
Deposition pressure = 1.6 Pa
Duty cycle = 20 %
Substrate temperature = 150 ℃

T_c= 14.80 K

Film with a **higher NbTiN (200)** texture relative to the (111) texture

Conclusion and Future Work



Higher Cathode Power (≥ **400 W**)

Lower Deposition Pressure (≤ **1.3 Pa**)

Lower Duty Cycle (~ **10%**)

Lower Nitrogen % (≤ **5 %**)

Adequate Substrate Bias (~ **50 V**)

Substrate Temperature (**?**)

Smoother, denser, compact, well-adhered films with high T_c

- Additional depositions to further investigate the influence of process parameters on thin film quality.
- Use of additional characterisation techniques (AFM, XPS, EBSD, TEM, etc).
- Optimization of insulating (AlN or Al_2O_3) thin films.
- Fabrication of SIS multilayered structures using optimised recipes.

Thank you for your attention!

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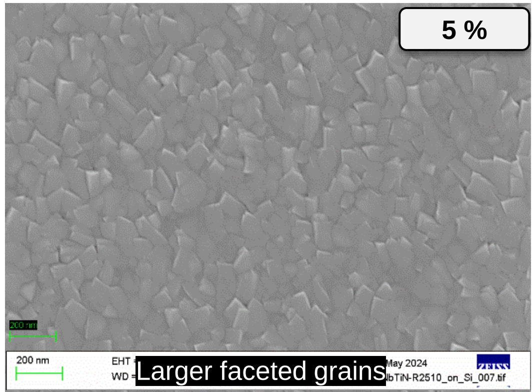
Backup slides

Nitrogen Flow Study

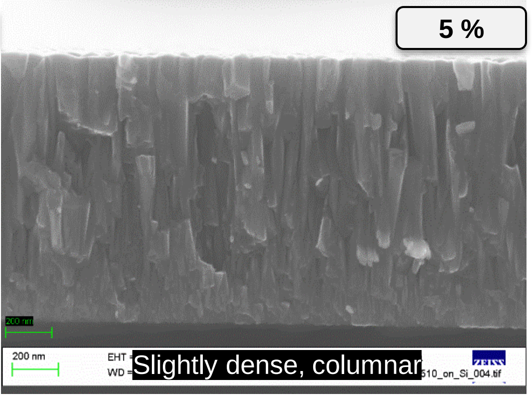
Cathode power = 400W, Sub. Bias = 50 V, Deposition pressure = 1.3 Pa, Duty cycle = 20 % Substrate temperature = 150 ℃

N_2 % = *5, 10, 15, and 20*



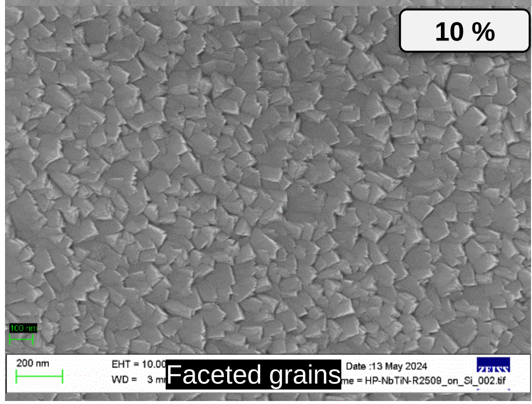
5 %

Larger faceted grains



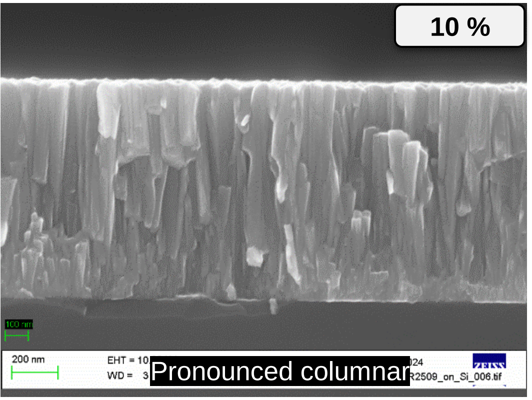
5 %

Slightly dense, columnar



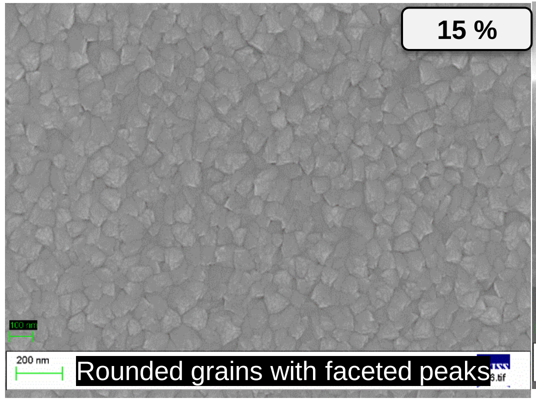
10 %

Faceted grains



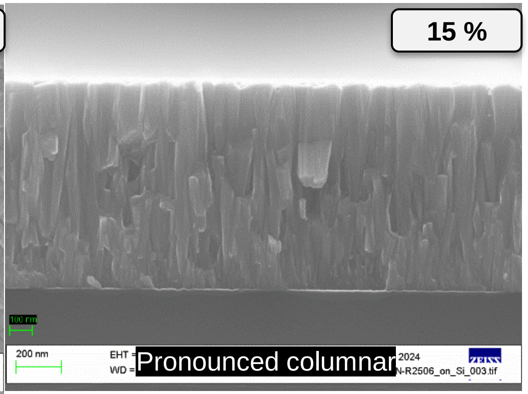
10 %

Pronounced columnar



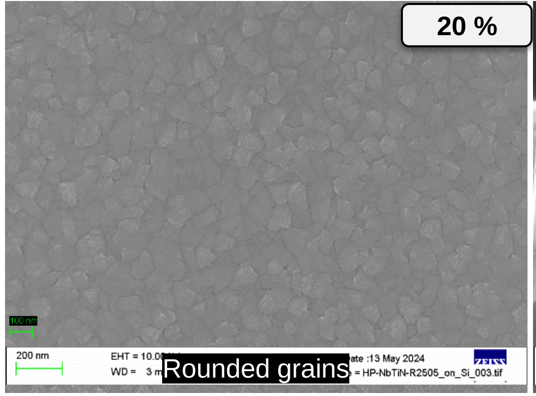
15 %

Rounded grains with faceted peaks



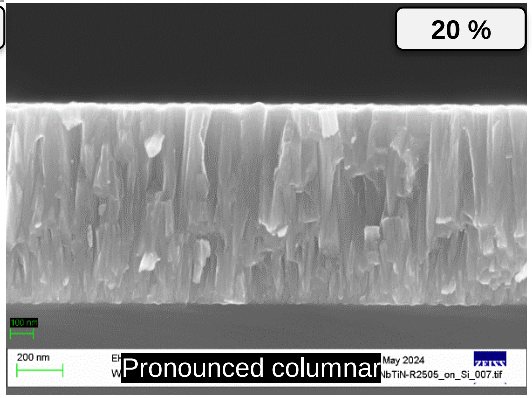
15 %

Pronounced columnar



20 %

Rounded grains

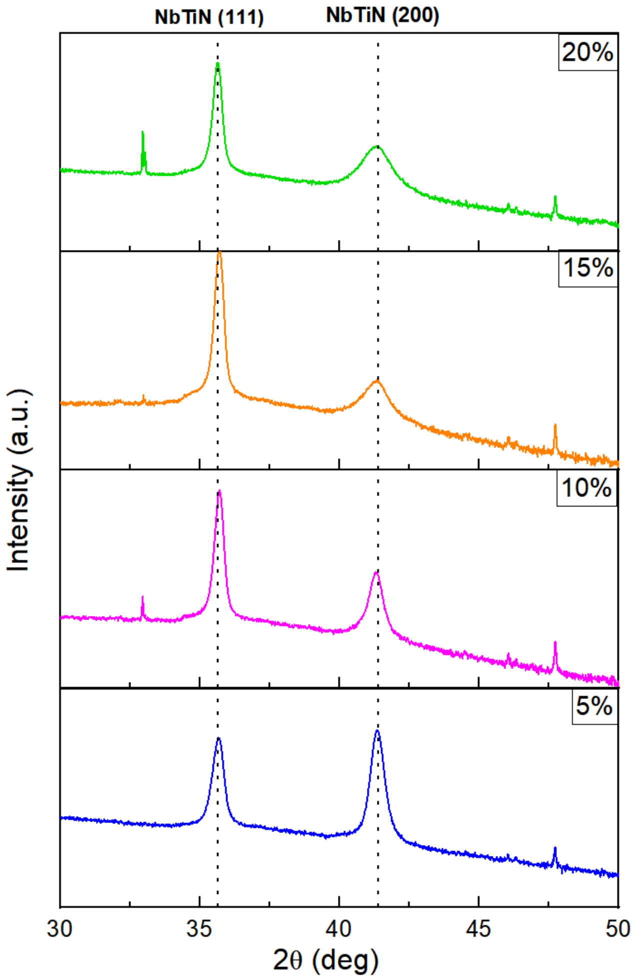


20 %

Pronounced columnar

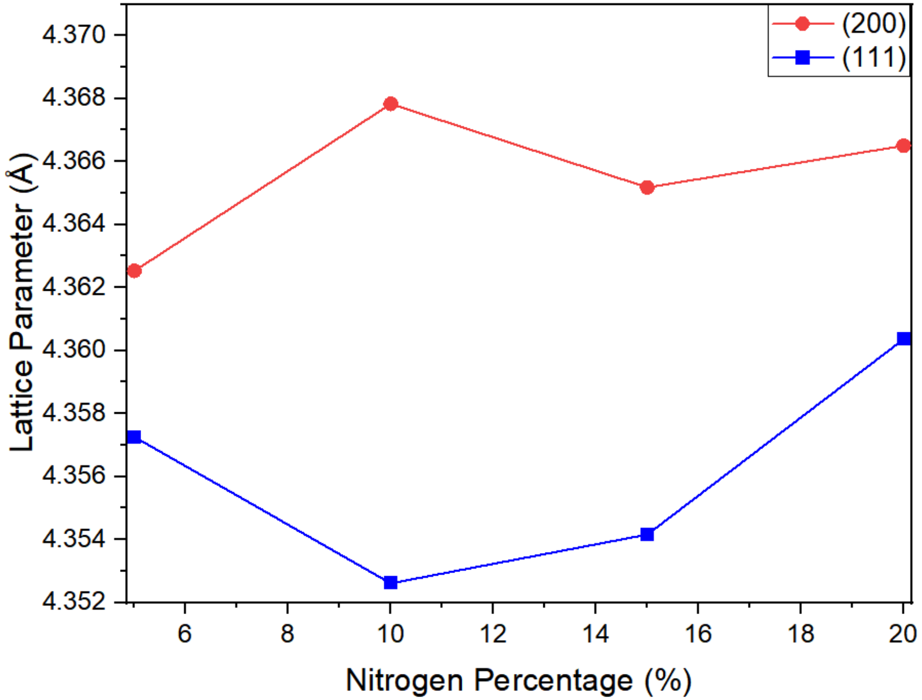
N_2%↑ argon ions % ↓ **sputtering yield** ↓

Nitrogen Flow Study



XRD Spectra

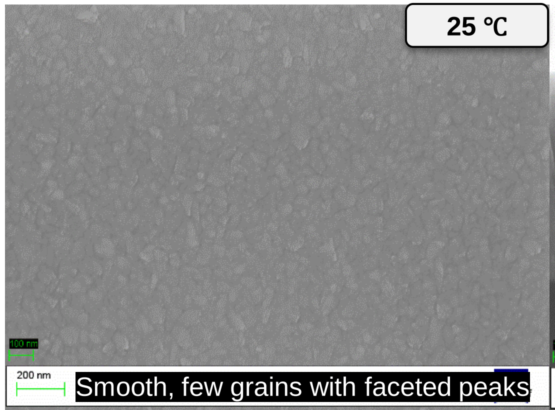
- N_2% ↑ → transition from NbTiN (200) to (111)
- N_2% ↑ → peak broadening for (200) → a decrease in the crystallite size



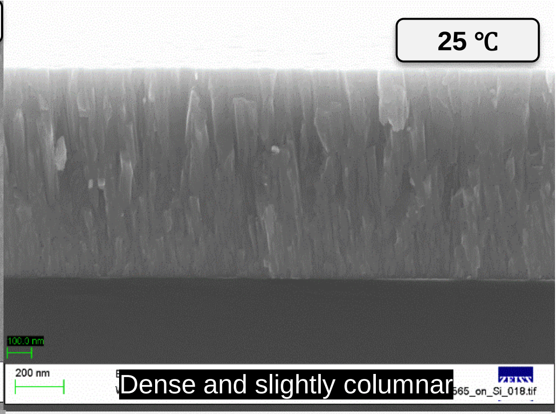
Substrate Temperature Study

Cathode power = 400 W, N_2% = 10, Substrate bias = 50 V, Deposition pressure = 1.3 Pa, Duty cycle = 10 %

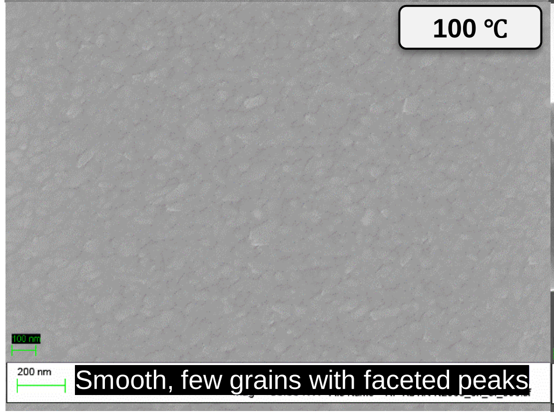
Substrate temperature = 25, 100, 150, and 250 ℃


25 ℃

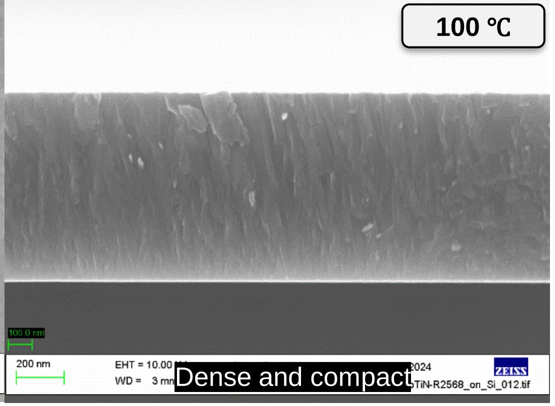
Smooth, few grains with faceted peaks


25 ℃

Dense and slightly columnar


100 ℃

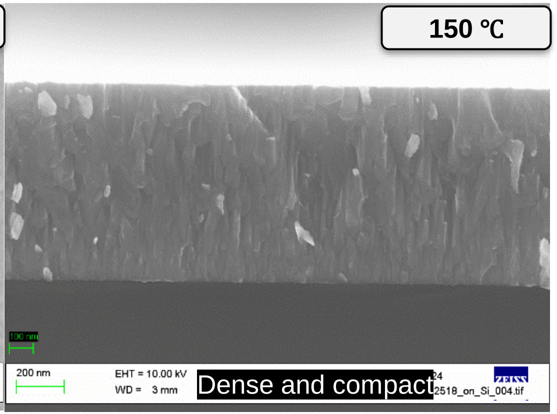
Smooth, few grains with faceted peaks


100 ℃

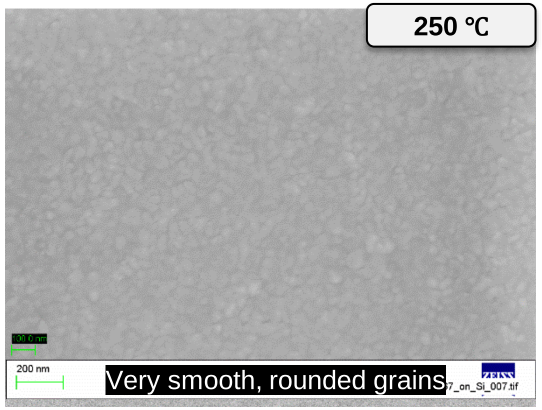
Dense and compact


150 ℃

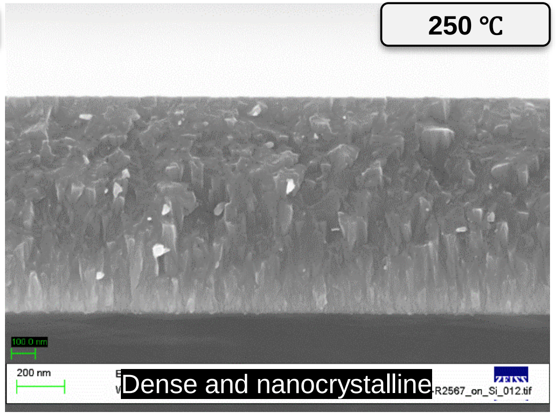
Very smooth, rounded grains


150 ℃

Dense and compact


250 ℃

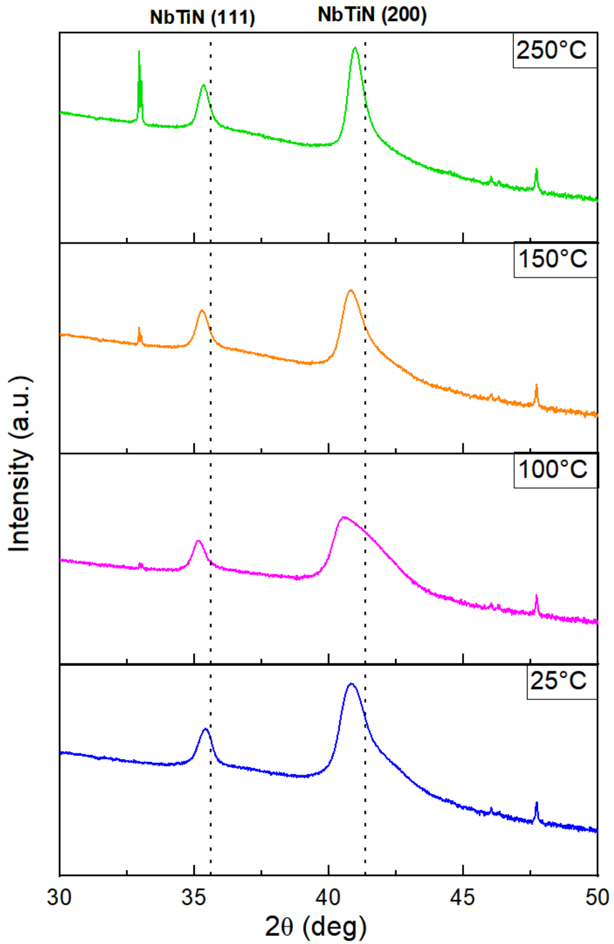
Very smooth, rounded grains


250 ℃

Dense and nanocrystalline

Substrate Temperature ↑ growing film temperature ↑ surface mobility and diffusion ↑ **film density** ↑

Substrate Temperature Study



XRD Spectra