

# SUPERCONDUCTING COATINGS SYNTHETIZED BY CVD/PECVD FOR SRF CAVITIES

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## Abstract

Bulk niobium cavities are widely employed in particle accelerators to create high accelerating gradient despite their high material and operation cost.

In order to reduce this cost, thin layer of niobium are deposited on a copper cavity, which has lower material cost with higher availability and more importantly higher thermal conductivity.

The coating of superconducting cavities currently is synthesized by physical vapour deposition (PVD) method which suffers from lack of conformity. By using chemical vapour deposition (CVD) and plasma enhanced chemical vapour deposition (PECVD) it is possible to deposit thin Nb layers uniformly with density very close to bulk material. This project explores the use of PECVD / CVD techniques to deposit metallic niobium on copper using NbCl<sub>5</sub> as precursor and hydrogen as a coreagent. The samples obtained were then characterized via SEM, TEM, SAD, XRD, XPS, and EDX as well as assessing their superconductivity characteristics (RRR and T<sub>c</sub>) All the samples deposited are superconductive and polycrystalline; the sample obtained with CVD measured RRR=31 and T<sub>c</sub>=7.9 K, while the sample obtained with PECVD exhibited RRR=9 and T<sub>c</sub>= 9.4 K. In both cases the films grew in a (100) preferred orientation.

## INTRODUCTION

Superconductive niobium cavities are widely used in particle accelerators to provide the gradient required to accelerate the desired particles via RF energy. Technology has caught up with the physical limits of bulk niobium, exploiting the material to its maximum potential [1]. Since the SRF requires less than 1 micron of material, it could be possible to obtain the same properties of niobium bulk with niobium thin films [2]. Thin films are a considerably cheaper option than bulk materials for the following reasons, firstly less material is being used and secondly, since SRF cavities operate at temperatures below 10K, the films can be deposited on high thermal conductivity materials such as copper making it easier to cool and maintain the low temperatures than with bulk superconductors [3]. Theoretical studies [4] have also suggested that the use of Superconductor / Insulator / Superconductor thin films (ISI) can lead to an increase in the accelerating gradient, surpassing the limits of bulk niobium. CVD [5] is a chemically driven technique that

allows the coating of large areas with great control over the produced film. This technique is therefore being explored to verify the feasibility of depositing metallic superconductive niobium with a homogeneous and uniform structure.

The purpose of the present study is to deposit thin films of Nb using CVD and plasma enhanced CVD (PECVD) techniques. The use of plasma enables reduction of the deposition temperature and allows coating of shapes with a high degree of complexity, such as cavities [6].

After microstructural evaluation, the films have then been assessed for their superconducting properties showing their suitability for use in SRF cavities.

## EXPERIMENTAL SETUP

A steel spherical chamber is kept at a base pressure of 10-6 mbar and constantly heated to 120 °C as shown in figure 1. The carrier gas (argon) and the reducing gas (hydrogen) are purified through a heated filters system to ensure that the presence of residual contaminants in the deposition chamber is kept as low as possible.

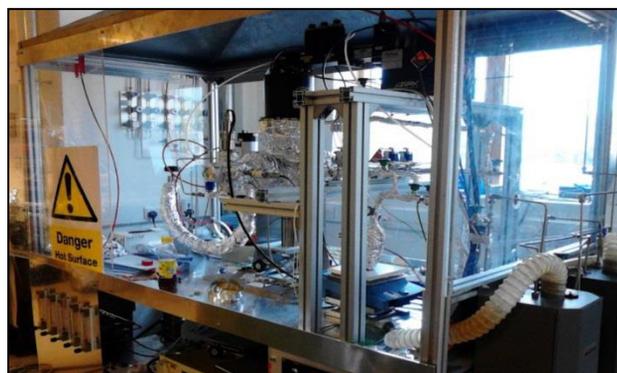


Figure 1: Deposition facility.

The chemical precursor (NbCl<sub>5</sub>) is placed under controlled atmosphere in a two-legged steel bubbler. The bubbler is then connected to the deposition rig and maintained at the chamber base pressure. Oxygen free copper substrates are cleaned via in-house process (acetone – IPA- distilled water) to further reduce the quantity of contaminants. A substrate is introduced in the chamber and kept at the chamber base pressure for 24 hours prior deposition. The NbCl<sub>5</sub> precursor is heated up until a vapor pressure suitable for the deposition is

reached [7] (130 °C). The substrate is heated to 650 °C. The carrier gas is then bled into the bubbler where it forces the evaporated precursor in the reaction chamber, where the reducing gas is also introduced. A flow of 100 sccm of Ar and H grants a sufficient transport of precursor and a sufficient amount of reducing gas in the chamber. Once reached the desired deposition time, the coated substrate (sample) is cooled down to room temperature while kept at the chamber base pressure. Once the sample has cooled down, it is then extracted and analyzed. Three samples were prepared: S1 deposited at 1 mbar, S2 at 5 mbar and S3 with PECVD with the remaining deposition parameters kept the same for all the samples.

Microstructural analyses were performed by SEM, TEM, XRD and EDAX. SEM and EDAX analyses were performed to determine the thickness of the films, their uniformity and their chemical composition. TEM was performed to study the samples section and in conjunction with XRD they were used to verify the crystalline structure of the films.

## RESULTS AND DISCUSSION

One of the main concerns for CVD materials regards the quantity of contaminants that remain trapped in the deposited layer. These contaminants are not desired, especially for superconductive materials, since they will hinder and reduce the desired properties of the obtained film [8].

This consideration led us to choose to deposit in high vacuum and at high temperatures, to ensure the lowest possible presence of contaminants in the proximity of the sample. The EDX results show that the films have a very low percentage of contaminants, especially chlorine that is undetectable which is attributed to the deposition temperatures in excess of 400 °C. Oxygen and carbon were measured at < 1 weight %, which is important for superconductivity properties.

Thickness uniformity is another issue for films deposited using these techniques. The orientation of the sample in relation to the precursors gas inlets, the gasses flow speed and path are all factors that influence the morphological characteristics of the obtained film.

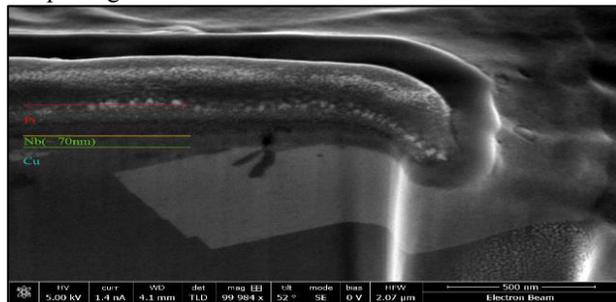


Figure 2: High magnification SEM of section of thin Nb film (sample S1).

The high magnification SEM shown in figure 2 of sample S1 shows a clear distinction between the substrate (copper) and the film, however no information on the

structure of the deposited film is evident as shown in figure 3.

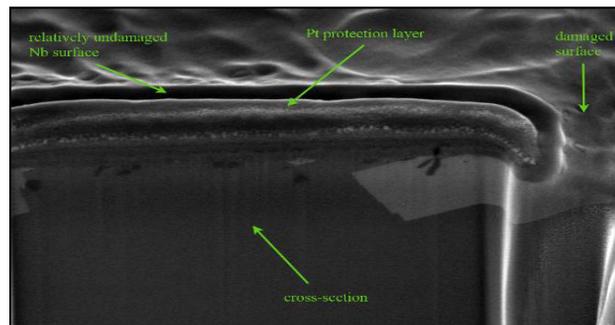


Figure 3: High magnification SEM of section of thin Nb film (sample S1).

To explore the film microstructure, TEM analysis was conducted on a side section of sample S2, to investigate the thickness and the composition of the film. Figure 4 shows the film on the copper substrate.

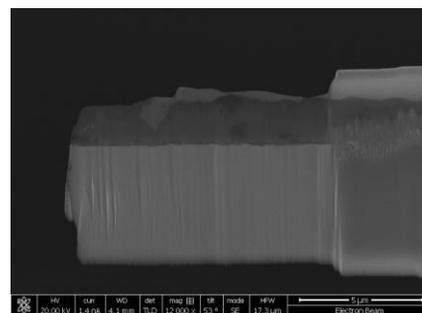


Figure 4: TEM of section of thin Nb film (sample S2).

The thickness of the film is about 2 μm, and it is uniform over the substrate surface. EDX elemental analysis performed on the sample confirmed its composition, with 98% in weight being niobium and traces (less than 1% in weight) of oxygen and carbon.

The sample crystalline structure was also studied via selected area electron diffraction (SAED) of the TEM images.

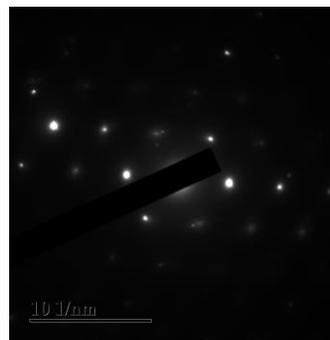


Figure 5: SAED of thin Nb film (sample S2).

The diffraction pattern in figure 5 shows that the obtained film is polycrystalline body-centered cubic (bcc) Nb.

Glancing angle X-ray Diffraction (GI-XRD) was also performed on the sample to further confirm the crystalline

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structure. The CVD deposited layer was removed from the substrate via chemical etching of the copper substrate and subsequently analysed. The spectrum is reported in figure 6. The pattern matches the standard XRD pattern of niobium bulk metal (JCPDS card no. 035-0789), which allows the identification of the preferred lattice orientation. This corresponds to the (100) orientation.

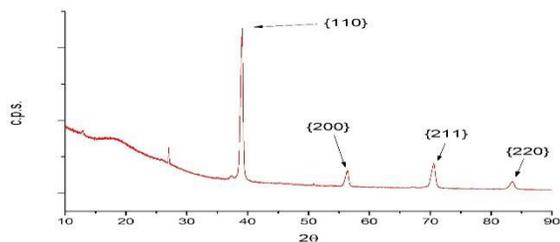


Figure 6: XRD of Niobium on Kapton tape (sample S2).

Sample S3 film was analysed without removing it from the copper layer, and the spectrum is shown in figure 7.

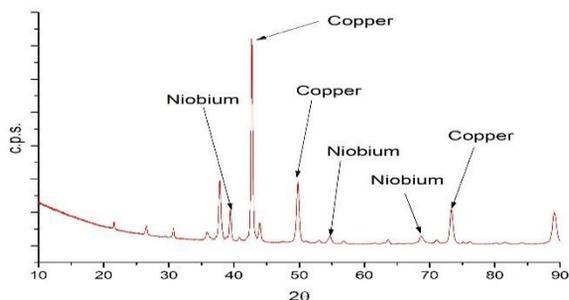


Figure 7: XRD of Niobium on Copper substrate (sample S3).

The spectra clearly show the diffraction peaks of niobium are identifiable, amongst the diffraction features coming from the underlying copper substrate.

Residual resistivity ratio measurements were then made on the samples. This was done by chemically etching the copper substrates and transferring the Nb film to Kapton tape as a support.

The results obtained were RRR=31 and  $T_c=7.9$  K for the CVD obtained sample, while RRR=9 and  $T_c=9.4$  K for the PECVD sample. In the CVD film, the critical temperature is lower than the equivalent for bulk niobium, and the reason why this happens is being investigated. Sample S3 shows the same transition temperature as bulk niobium, but shows a lower RRR, and this is also being investigated.

## CONCLUSIONS

We have deposited niobium thin films by using CVD and PECVD and their superconductivity and

microstructural properties have been investigated. Performing the depositions in high vacuum has considerably reduced the contaminants trapped in the films. The high deposition temperature allows the complete dissociation of the NbCl<sub>5</sub> precursor and the elimination of residual chlorine, but it presents some engineering challenges in the event of coating large copper structures.

Further investigation is required to understand the relationship between the RRR measurements and the deposition methods and parameters. An alternative to CVD, offering lower deposition temperatures is atomic layer deposition [9], which we propose to exploit for the deposition of thin films for accelerator cavities.

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