

SUPERCONDUCTING COATINGS SYNTHETIZED BY CVD/PECVD FOR SRF CAVITIES

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Abstract

Finding a way to overcome the acceleration gradient limits that bulk niobium cavities can provide is a major challenge, fundamental to allow the accelerator science field to progress.

In order to overcome the accelerating gradient limits of bulk niobium and reduce manufacturing and operation costs, the idea of using thin layers of niobium deposited on a copper cavity is being explored. This approach has lower material cost with higher availability and more importantly higher thermal conductivity.

Physical vapour deposition (PVD) method is currently the preferred method to coat superconducting cavities, but its lack of conformity renders complicated shapes such as crab cavities very difficult to coat. 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_5 as precursor and hydrogen as a coreagent. The samples obtained were then characterized via SEM, XRD, and EDX as well as assessing their superconductivity characteristics (RRR and T_c). The samples deposited are superconductive and polycrystalline; the sample obtained with PECVD exhibited RRR=9 and $T_c=9.4$ K. The film grew in a (100) preferred orientation.

A successful preliminary deposition of NbN has also been performed and analyzed via EDX.

INTRODUCTION

SRF niobium cavities are used in particle accelerators to provide accelerating gradients with low power dissipation than traditional copper cavities. Bulk niobium has been exploited to its technological limit [1], leading the research to explore alternative ways to progress. 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 10 K, 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. These superconductor thin films could then be chosen from a range of alloys with a higher H_c and T_c , such as NbN, Nb_3Sn or MgB_2 . 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 niobium nitrate 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, and to deposit thin films of NbN using plasma enhanced CVD (PECVD) techniques. The use of plasma enables the creation of N radicals, the 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. 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.

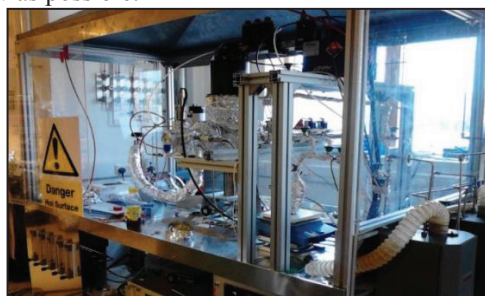


Figure 1: Deposition facility.

The chemical precursor (NbCl_5) 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_5 precursor is heated up until a vapor pressure suitable for the deposition is reached [7] (130 °C). The substrate is heated to 650 °C and the deposition starts once the sample stops degassing. 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. Two samples were prepared: S1 deposited at 5mbar with PECVD, and one preliminary sample, S2, to verify the possibility of obtaining niobium nitride using N as a coreagent: nitrogen and hydrogen gasses in ratio 1:3 were bled from the top of the ECR, with NbCl_5 introduced close to the sample. The deposition parameters such as pressure, plasma power and gas flows were kept the same for both the samples.

Microstructural analyses were performed by SEM and EDX to determine the thickness of the films, their uniformity and their chemical composition. Four point probe measurements were conducted on sample S1 to verify its superconductive properties.

RESULTS AND DISCUSSION

The composition and purity from contaminants of the thin films deposited with CVD can be a concern. The presence of oxygen or other unwanted elements can seriously reduce and hinder the superconductive properties of the deposited material [8].

The desire to lower the contamination of the deposited film drove the design of our system, which operates in high vacuum and at high temperatures. The EDX results show that the deposited 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 was measured at < 1 weight %, which is important for superconductivity properties.

The uniformity of the deposited film is another CVD issue. The morphological characteristics of the deposited film can be influenced by such factors as gasses flow speed and directions, sample tilting and orientation in relation to the gasses inlets.

The high magnification SEM (fig. 2 and 3) 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.

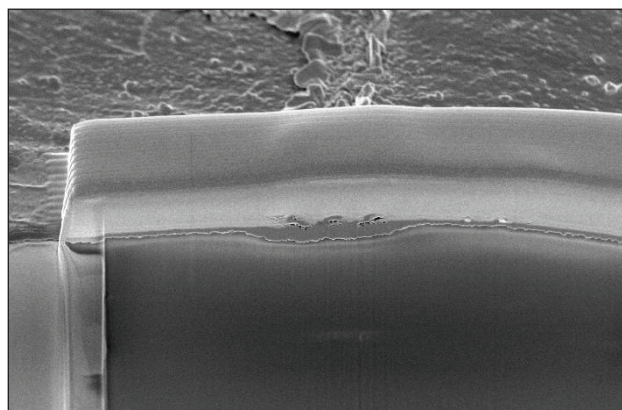


Figure 2: High magnification SEM of section of thin Nb film (sample S3).

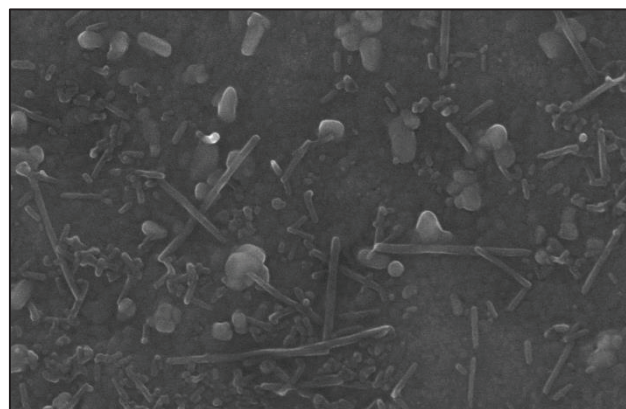


Figure 3: High magnification SEM of surface of thin Nb film (sample S1).

The thickness of the film is about 200 nm, 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.

Glancing angle X-ray Diffraction (GI-XRD) was performed on the sample to verify the crystalline structure.

Sample S3 film was analysed without removing it from the copper layer, and the spectrum is shown in figure 4.

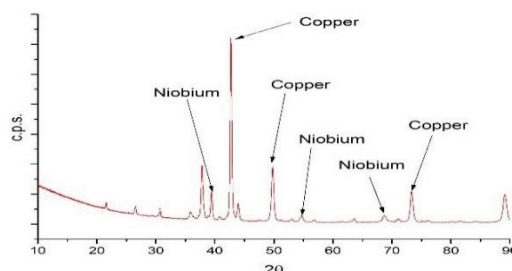


Figure 4: XRD of Niobium on Copper substrate (sample S3).

The spectra clearly shows the diffraction peaks of niobium are identifiable (JCPDS card no. 035-0789),

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 $RRR=9$ and $T_c=9.4$ K for the PECVD sample S1. Sample S1 shows the same transition temperature as bulk niobium, but shows a low RRR, the reasons why are being investigated.

High magnification SEM analysis was conducted on sample S2, to investigate the morphology of the deposited film.

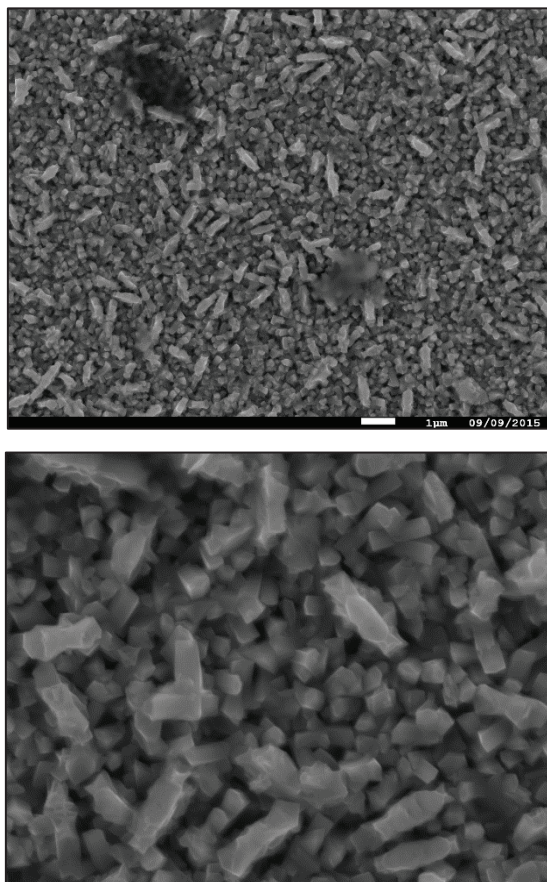


Figure 5: High magnification SEM of surface of thin NbN film (sample S2).

Figure 5 shows that the structure of the film appears to be crystalline, although not further comments can be made without more in depth analyses.

EDX elemental analysis was performed also on sample S2, to verify its composition. The results show the presence of niobium and nitrogen in comparable quantities (Nb: 44%, N: 48%). The sample also shows oxygen (5%) which could be attributed to exposure to atmosphere.

Titanium was also detected in small quantities on the sample, probably leftover by a previous deposition run.

Table 1: EDX Results Comparison

| SAMPLE NAME | NIObIUM (% WEIGHT) | COPPER (% WEIGHT) | OXYGEN (% WEIGHT) | NITROGE N (% WEIGHT) |
|-------------|--------------------|-------------------|-------------------|----------------------|
| S1 | 98% | <1% | <1% | / |
| S2 | 46% | <1% | 4.5% | 46% |

These quantitative results, although promising, cannot be considered conclusive: the difference in energy between the nitrogen K line (0.39) and the titanium L line (0.45) is too little for the analysis software to successfully deconvolute the two elements. This added to the high correction factor applied to lighter elements can lead to mistakes in the quantification of the elements present on the samples. To resolve this issue electron energy loss spectroscopy (EELS) and Rutherford backscattering (RBS) will be performed on the sample to confirm and quantify the presence of nitrogen.

CONCLUSIONS

We have deposited niobium thin films by using CVD and PECVD and their superconductivity and microstructural properties have been investigated. The low presence of oxygen and the lack of contaminants proves that the deposition system is effective in maintaining the contamination level low. The high deposition temperature allows the complete dissociation of the $NbCl_5$ 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.

We have also successfully deposited niobium nitride thin films by using PECVD and their microstructural properties have been preliminarily investigated. Future studies will explore the superconductive properties of the deposited films.

Atomic layer deposition, which offers lower deposition temperatures and high conformity, is proposed as an alternative to CVD, for the deposition of thin films for accelerator cavities.

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