

## Conference/Workshop Paper

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# DEVELOPMENT OF Nb<sub>3</sub>Sn COATINGS BY MAGNETRON SPUTTERING FOR SRF CAVITIES

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## Abstract

We present in this paper the first results obtained on DC magnetron sputtering of Nb<sub>3</sub>Sn thin films dedicated to superconducting radio frequency cavities (SRF). Nb/Sn ratio of 3.76 and 3.2 have been obtained for Ar coating pressures of respectively  $1.10^{-3}$  mbar and  $5.10^{-2}$  mbar. According to XRD analysis both coating pressures lead to amorphous Nb<sub>3</sub>Sn layers that are not superconducting. One coating has been annealed at 700°C, 750°C and 800°C under vacuum for 24h and exhibits for the three different temperatures the A15 cubic phase.

## INTRODUCTION

Very low losses SRF accelerating systems, together with high-efficiency cryogenics systems, have the potential of low running costs. Nb coated Cu cavity technology used for LEP [1], LHC [2] and HIE ISODLE [3] starts to show a limitation in terms of Q-slope and quench limit. The use of A15 materials is very promising as they exhibit higher T<sub>c</sub> value than pure Nb allowing thus to reach better quality factor for a given working temperature. Recent work [4] has demonstrated the possibility to use such materials in bulk Nb cavities showing good performances even at high accelerating gradients. We propose to combine the efficiency of such A15 materials with the good thermal performances of copper accelerating cavities.

The study considers the possibility to coat a copper resonator with an Nb<sub>3</sub>Sn layer by means of DC magnetron sputtering using an alloyed target. We present the influence of the process parameters on the as-deposited layer stoichiometry. The latter is in good agreement with previous results reported in the literature and can be fine-tuned by acting on the process gas pressure. The effect of post-coating annealing temperature on the morphology and crystallinity of the film is being also investigated.

## EXPERIMENTAL PROCEDURE

A 150 mm diameter magnetron sputtering source was used to deposit Nb<sub>3</sub>Sn from a 3mm thick stoichiometric Nb-Sn target. All samples were coated at room temperature using a cathode-substrate distance of 100mm with an average power of 200W and using Ar as the sputtering process gas for 60 min. Before coating a 135°C bakeout of the entire coating chamber was performed for at least 10

hours allowing reaching a base pressure of  $7.10^{-8}$  mbar at room temperature

The coating pressure was tuned by modifying the process gas flow through a mass flow controller.

The substrates used in this study were:

- High purity quartz with OH content lower than 1ppm dedicated to DC superconducting properties measurements
- OFE copper
- Standard borosilicate glass for thickness measurements

Film annealing was carried out in a separate furnace over 24h under vacuum with a constant ramp-up of 200°C/hour up to 3 different temperatures, 700°C, 750°C and 800°C, respectively. The furnace temperature was monitored by two thermocouples showing a maximum temperature deviation from target of 3°C over the entire process. The typical maximum pressure value reached during the annealing was around  $2.10^{-6}$  mbar.

Typical characterization were carried out using SEM for morphology, EDX for composition (at 10KeV incident electron beam), XRD ( $\theta$ -2 $\theta$ , fixed source and rotatable sample and detector, Cu anode) for crystal structure, and profilometry for thickness measurements. Superconducting properties were evaluated using a 4-probes setup cooled down to 4K using liquid He. The measures were performed at a constant current of 100 mA

## RESULTS

### *As deposited films*

It is well known that the critical temperature of the Nb<sub>3</sub>Sn phase is strongly coupled to the Sn content of the film [5]. Controlling such stoichiometry is thus compulsory in order to tune the film properties.

In order to modify the film stoichiometry we used two different coating pressures of  $1.10^{-3}$  mbar and  $5.10^{-2}$  mbar. The coatings thickness were 1.6  $\mu$ m and 2.1  $\mu$ m respectively

Figure 1 shows that the composition depends on process pressure and is in good agreement with the previous results reported by C.T. Wu et al [6] using the same coating method. This effect is directly linked to the modulation of the mean free path of sputtered atoms leading to a change in their transmission ratios.

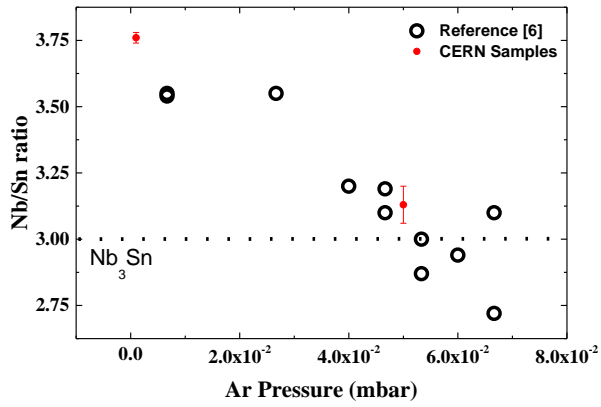


Figure 1: Nb/Sn ratio for as-deposited samples versus coating pressure compared with [6].

These two samples were also analysed by means of X-ray diffraction. The obtained spectra are presented in figure 2.

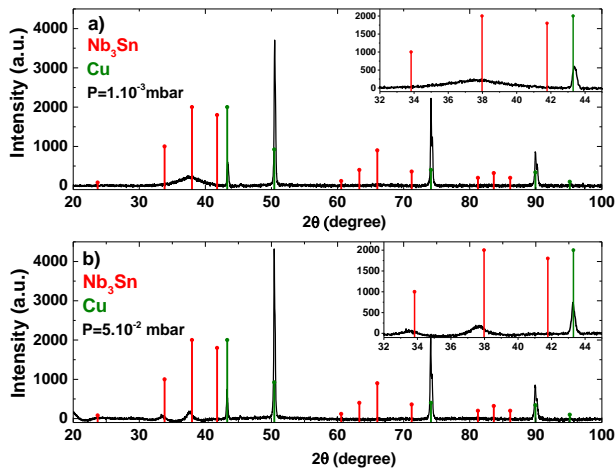


Figure 2: XRD spectra of as deposited  $Nb_3Sn$  thin films on copper substrate obtained for a process pressure of a)  $1.10^{-3}$  mbar and b)  $5.10^{-2}$  mbar

For a pressure of  $1.10^{-3}$  mbar the low angle diffraction peaks of  $Nb_3Sn$  are not resolved which suggest that the crystallites are even smaller than those obtained at  $5.10^{-2}$  mbar. Indeed according to the Scherrer equation the crystallites size  $L$  is defined as following:

$$L = \frac{K\lambda}{\beta \cos \theta}$$

Where  $K$  is a shape factor taken as 0.89,  $\lambda$  is the XRD source wavelength,  $\beta$  is the full width at half maximum in radian of the considered peak and  $\theta$  is the diffraction angle in radian. Using the [210] diffraction peak of  $Nb_3Sn$  one can estimate the crystallites size obtained at  $1.10^{-3}$  mbar to be at a maximum value of 10 nm. At  $5.10^{-2}$  mbar they are found to be at least twice bigger.

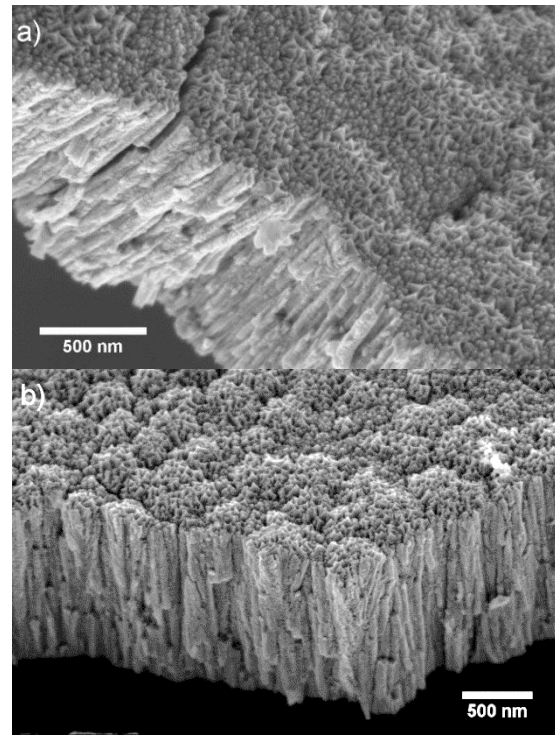


Figure 3: SEM pictures of  $Nb_3Sn$  thin films deposited on Cu samples at a)  $1.10^{-3}$  mbar and b)  $5.10^{-2}$  mbar.

Both observations are consistent with the fact that the coating is performed at a low temperature, estimated to be around  $150^\circ C$ . The ad-atom thermal energy is too low to allow them forming the A15 phase once on the substrate. SEM pictures on Figure 3 confirm this. One can clearly see a cauliflower structure on the surface. The film is columnar, rough and porous resulting from low temperature coating and weak ion bombardment assistance due to the low voltage drop ( $\sim 300V$ ) in the magnetron discharge. From the SEM image of the cross section there is no indication of a change in the grain size along the film thickness.

Contrary to XRD analysis the SEM pictures do not reveal any visible morphology impact of the coating pressure.

This highly disordered structure has also an effect on the conductivity properties of the films. Indeed no superconducting transition is found.

Even though the right composition has been obtained one still needs to obtain the A15 cubic phase. In order to achieve this the samples elaborated at  $1.10^{-3}$  mbar have been annealed at three different temperatures under vacuum.

### Annealed films

$Nb_3Sn$  as-deposited thin films on copper substrate prepared at  $1.10^{-3}$  mbar have been annealed at  $700^\circ C$ ,  $750^\circ C$  and  $800^\circ C$ . Further studies will be carried out later on to repeat this on the samples elaborated at higher pressure.

The XRD spectra of the reference as-deposited film and annealed films are compared in the figure 4. The formation of the expected A15 cubic Nb<sub>3</sub>Sn phase is visible for the three different temperatures. Because the furnace was cooled down using its own thermal inertia (time constant of hours) we can conclude that the phase remains stable even at room temperature. One can also notice the modification of the Cu substrate which shows a clear [311] preferential orientation becoming dominant in volume and thus giving a high peak intensity when annealed at 800°C. The good match between the spectra and the powder diffraction reference patterns confirm the random orientation of the crystals Nb<sub>3</sub>Sn.

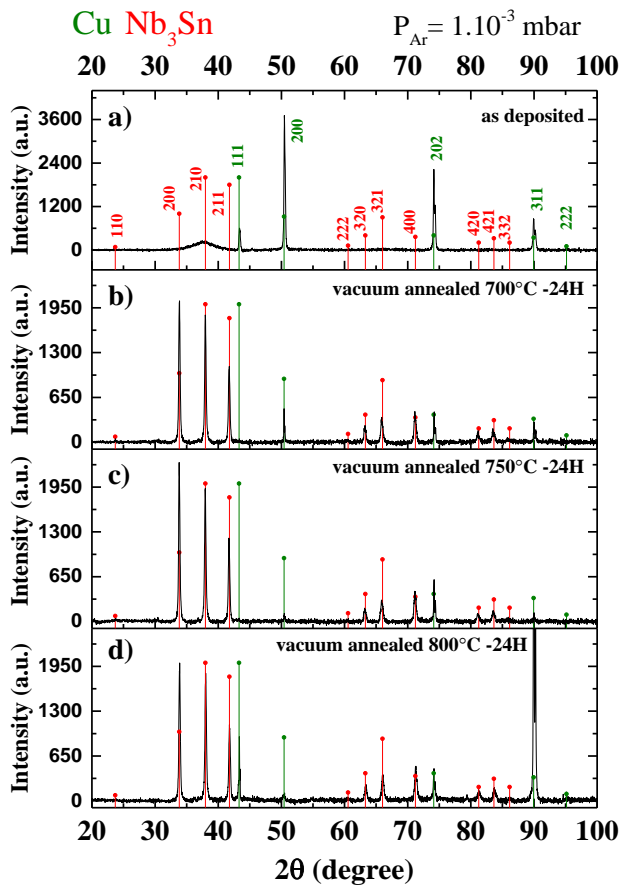


Figure 4: XRD spectra of Nb<sub>3</sub>Sn thin films deposited on copper substrate at  $1.10^{-3}$  mbar Ar pressure a) as deposited b) annealed at 700°C c) annealed at 750°C and d) annealed at 800°C.

The samples were also observed under SEM in order to evaluate grain size and surface quality. The pictures are shown in the figure 5. The pictures highlight a clear surface morphology difference between the films annealed at different temperature. As expected, the higher the annealing temperature the bigger the grains. The surface has been smoothed compared to the as-deposited material thanks to the higher atoms mobility at high temperature.

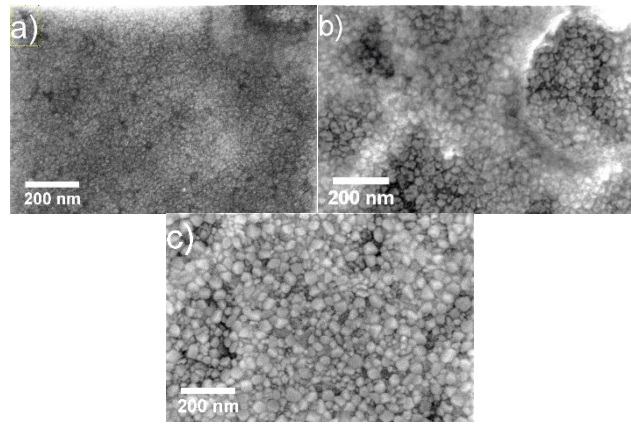


Figure 5 : SEM pictures of Nb<sub>3</sub>Sn thin films annealed at a) 700°C, b) 750°C and c) 800°C

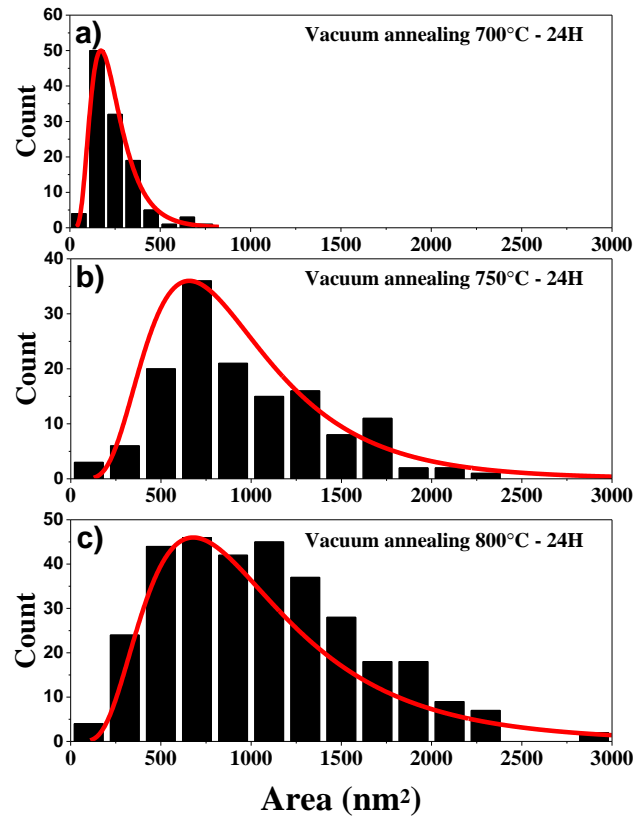


Figure 6: Grains size distributions from image analysis on the SEM data, for Nb<sub>3</sub>Sn films annealed for 24 hours at a) 700°C, b) 750°C and c) 800°C. The red curves correspond to a lognormal fit of the experimental data [7].

The figure 6 shows the effect of the annealing temperature on the grain size. While an annealing at 700°C leads to small grains the annealing at 750°C and 800°C shift this distribution toward higher values. However one can still notice that values we obtain here remain much smaller than those reported by Becker *et al* [8] that are in the range of  $\mu\text{m}^2$ . This can be directly linked to the much lower process temperature used for this study compared to the 1000°C used in the so-called Wuppertal process.

## CONCLUSION

We have demonstrated the feasibility of coating copper with Nb<sub>3</sub>Sn by sputtering an alloyed target of stoichiometric Nb-Sn. A thermal treatment compatible with the copper mechanical properties has been set up and led to the formation of the cubic A15 Nb<sub>3</sub>Sn phase. As expected the grain size can be tuned by using different annealing temperature.

Next steps of this study will consist of evaluating the superconducting performances of the annealed films as well as comparing them to films directly deposited at high temperature. Annealing the films at temperature lower than 700°C will also be studied as well as improving the vacuum base pressure and quality in the furnace. Finally RF superconducting properties will have to be assessed using a quadrupole resonator [9].

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