DEVELOPMENT OF Nb_3Sn COATINGS BY MAGNETRON SPUTTERING FOR SRF CAVITIES

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Abstract

This paper presents the first results obtained on DC magnetron sputtering of Nb_3Sn 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 analyses both coating pressures lead to amorphous Nb_3Sn layers that are not superconducting. Afterward, one coating has been annealed at 700°C, 750°C and 800°C under vacuum for 24h and exhibited 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 limited performances in terms of Q-slope and quench limit. The use of A15 materials is very promising as they exhibit higher T_c value than pure Nb, allowing 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. The present investigation proposes 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_3Sn layer by means of DC magnetron sputtering using an alloyed target. The influence of the process parameters on the as-deposited layer stoichiometry is presented. 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_3Sn from a 3 mm thick stoichiometric Nb-Sn target. All samples were coated at 150°C using a cathode-substrate distance of 100 mm with an average power of 200 W 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 h 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 24 h under vacuum with a constant ramp-up of 200°C/hour up to three different temperatures, 700°C, 750°C and 800°C. The furnace temperature was monitored by two thermocouples showing a maximum temperature deviation within the chamber of 3°C, over the entire process. The typical maximum pressure value reached during the annealing was around 2.10^{-6} mbar.

Characterization was carried out using SEM for morphology, EDX for composition (at 10 KeV incident electron beam), XRD (θ-2θ, fixed source and rotatable sample and detector, Cu anode) for crystal structure, and profilometry for thickness measurements. Superconducting DC (Tc, RRR) 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_3Sn phase is strongly coupled to the Sn content of the film [5]. Controlling such stoichiometry is thus compulsory to tune the film properties.

In order to modify the film stoichiometry, two different coating pressures were used, 1.10^{-3} mbar and 5.10^{-2} mbar. The resulting coating thickness were 1.6 µm and 2.1 µm 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.

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ISBN 978-3-95450-178-6

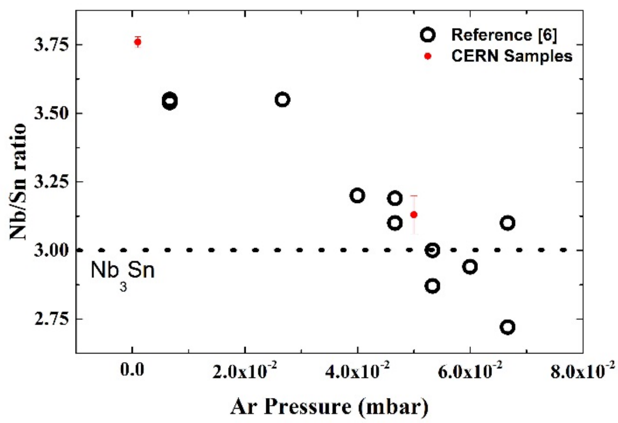

Figure 1: Nb/Sn ratio for as-deposited samples versus coating pressure compared with [6].

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

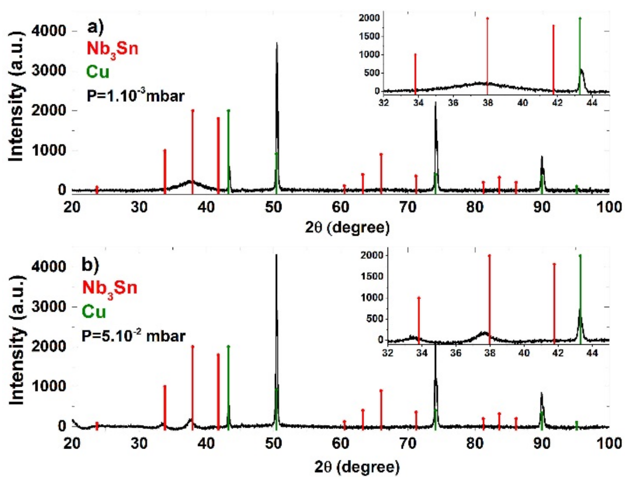

Figure 2: XRD diffractograms of as deposited Nb_3Sn thin films on copper substrate obtained for a process pressure of a) 1.10^{-3} mbar and b)5.10^{-2} mbar.

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, by extension of the Scherrer equation to anisotropic nano-crystallites, an assessment of the average crystallites size L can be proposed:

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

Where K is a shape factor taken as 0.89, λ is the XRD source wavelength, β is the full width at half maximum in radian of the considered peak and θ is the diffraction angle in radian. Using the [210] diffraction peak of Nb_3Sn the estimated average upper boundary crystallites size obtained at 1.10^{-3} mbar is around 10 nm. At 5.10^{-2} mbar, using the same calculation method, the crystallites are found to be at least twice bigger.

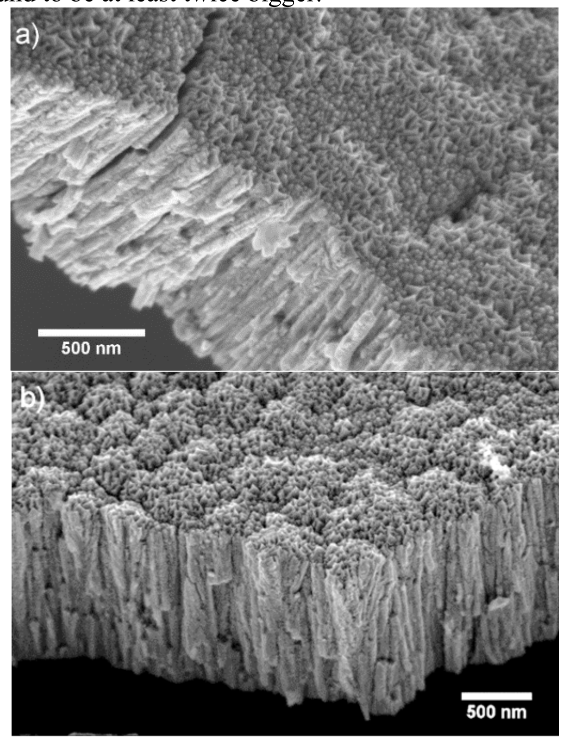

Figure 3: SEM pictures of Nb3Sn thin films deposited on Cu samples at a) 1.10^{-3} mbar and b) 5.10^{-2} mbar.

Both observations are consistent with the fact that the coating is performed at a low temperature, estimated to be around 150°C. The ad-atom thermal energy is too low to allow them forming the A15 phase once on the substrate. SEM pictures on Fig. 3 confirm this hypothesis. 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 (~300 V) in the magnetron discharge. From the SEM image of the cross section, no indication of a change in the grain size along the film thickness was observed.

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

The 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, the A15 cubic phase still need to be formed. In order to achieve this, the samples elaborated at 1.10^{-3} mbar, were 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°C, 750°C and 800°C. Further studies will be carried out later on to repeat this on the samples elaborated at higher pressure.

The XRD diffractograms of the reference as-deposited film and annealed films are compared in the Fig. 4.

The formation of the expected A15 cubic Nb_3Sn phase is visible for the three different temperatures. Because the furnace was cooled down using its own thermal inertia (time constant of hours) it can be concluded 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 diffractograms and the powder diffraction reference patterns confirm the random orientation of the crystals Nb_3Sn.

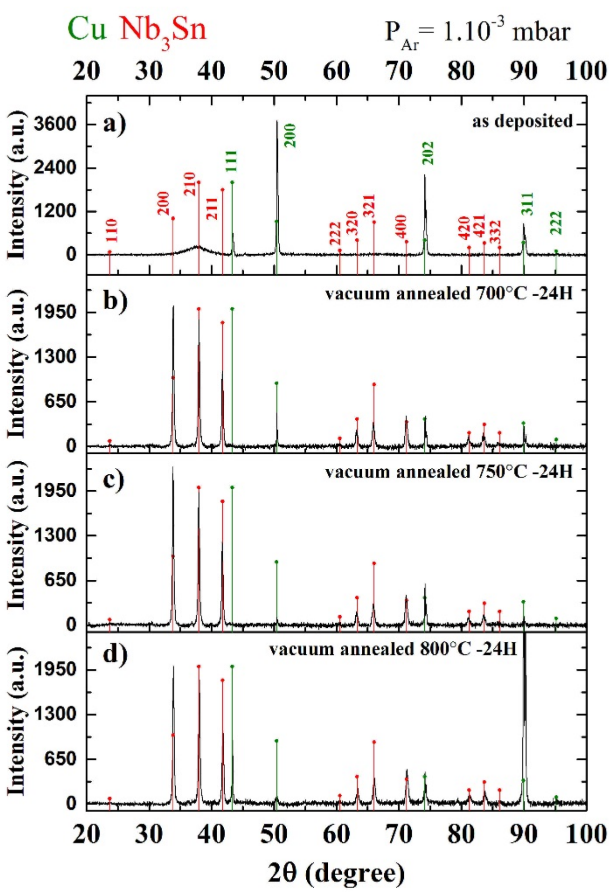

Figure 4: XRD diffractograms of Nb3Sn thin films deposited on copper substrate at 1.10^{-3} mbar Ar pressure a) as deposited b) annealed at 700°C c) annealed at 750°C and d) annealed at 800°C.

The samples were also observed under SEM in order to evaluate the surface grain size and surface quality. The pictures are shown in the Fig. 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 smoothened compared to the as-deposited material thanks to the higher atoms mobility at high temperature.

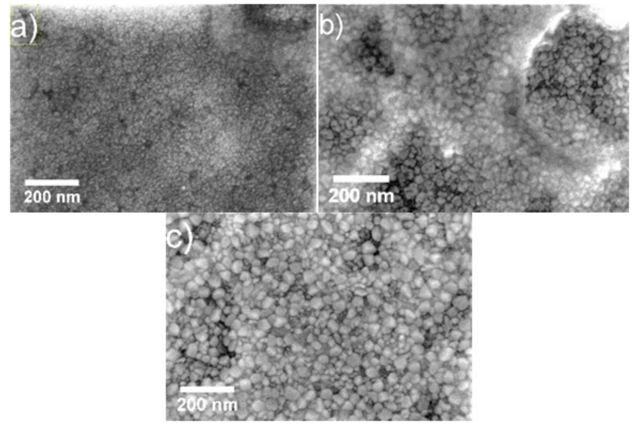

Figure 5: SEM pictures of Nb3Sn thin films annealed at a) 700°C, b) 750°C and c) 800°C.

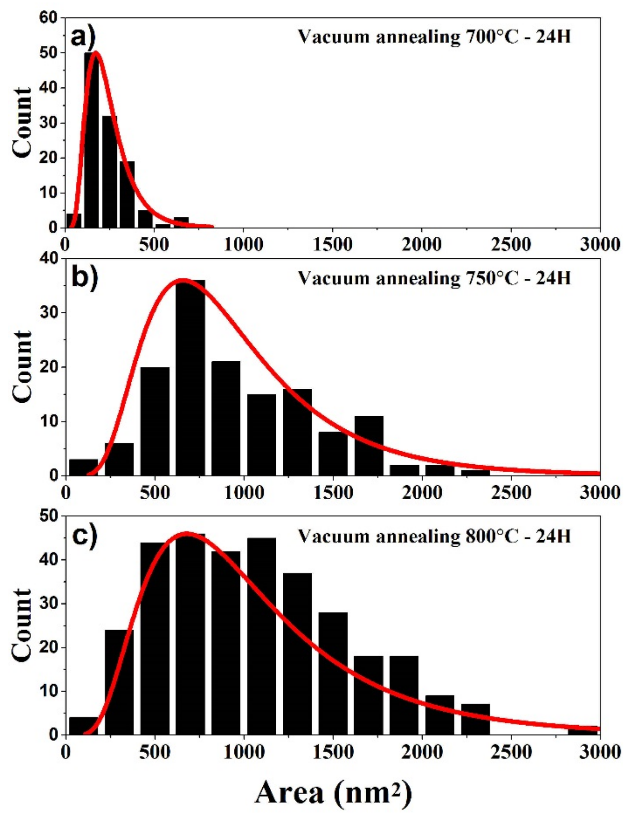

Figure 6: Surface grain size distributions from image analysis on the SEM data, for Nb_3Sn films annealed for 24 h at a) 700°C, b) 750°C and c) 800°C. The red curves correspond to a lognormal fit of the experimental data [7].

Figure 6 shows the effect of the annealing temperature on the surface 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 obtained here remain much smaller than those reported by Becker *et al* [8] that are in the range of μm^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.

ISBN 978-3-95450-178-6

CONCLUSION

The feasibility of coating copper with Nb_3Sn by sputtering an alloyed target of stoichiometric Nb-Sn has been demonstrated. A thermal treatment compatible with the copper mechanical properties has been set up and led to the formation of the cubic A15 Nb_3Sn phase. As expected, the grain size can be tuned by using different annealing temperature.

Next steps of this study will consist of evaluating the DC superconducting properties of the annealed films and comparing them to those of 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 CERN's Quadrupole Resonator [9].

ACKNOWLEDGMENT

The research leading to these results has received funding from the European Commission under the FP7 Research Infrastructures project EuCARD-2, grant agreement no.312453.

The authors would like to thank the CERN's chemistry workshop and cryolab for their support for sample preparation and characterization.

REFERENCES

[1] E. Chiaveri *et al*, Industrial production of superconducting niobium sputter coated copper cavities for LEP, SRF93, p.746 (1993).
[2] E. Chiaveri, The CERN Nb/Cu Programme for the LHC and reduced-b superconducting cavities, SRF99, CERN SL-99-076 CT (1999).
[3] A. Sublet *et al*, Nb coated HIE-ISOLDE QWR superconducting accelerating cavities: from process development to series production, IPAC2014, p.2571 (2014).
[4] S. Posen *et al*, Proof-of-principle demonstration of Nb_3Sn superconducting radiofrequency cavities for high Q_0 applications, Applied Physics Letters 106, 082601 (2015).
[5] A. Godeke, A review of the properties of Nb_3Sn and their variation with A15 composition, morphology and strain state, Supercond. Sci. Technol. 19, R68-R80 (2006).
[6] C. T. Wu *et al*, Highrate magnetron sputtering of high Tc Nb_3Sn films, Journal of Vacuum Science & Technology 14, 134 (1977).
[7] R. B. Bergmann *et al*, On the origin of logarithmic-normal distributions: An analytical derivation, and its application to nucleation and growth processes, Journal of Crystal Growth 310, 3135-3138 (2008).
[8] C. Becker *et al*, Analysis of Nb_3Sn surface layers for superconducting radio frequency cavity applications, Applied Physics Letters 106, 082602 (2015).
[9] T. Junginger et al, Extension of the measurement capabilities of the quadrupole resonator, Rev. Sci. Instrum. 83, 063902 (2012).