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INFLUENCE OF THE COATING PARAMETERS ON THE T_c OF Nb_3Sn THIN FILMS ON COPPER DEPOSITED VIA DC MAGNETRON SPUTTERING*

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Abstract

The I.FAST collaboration aims at pushing the performance of particle accelerators by developing sustainable innovative technologies. Among its goals, the development of thin film-coated copper elliptical accelerating cavities covers both the optimization of the manufacturing of seamless substrates and the development of functional coatings able to conform to the 3D cavity geometry while delivering the needed performance. For the latter, the optimization of the deposition recipe is central to a successful outcome. The work presented here focuses on the deposition of Nb_3Sn films on flat, small copper samples. The films are deposited via DCMS from a planar stoichiometric Nb_3Sn commercial target. The results of the film characterization are presented here. The observed dependencies between the critical temperature T_c of the films and the deposition parameters are discussed and, in particular, $T_c^{90\%-10\%} = (17.9 \pm 0.1)$ K is reported for Nb_3Sn on sapphire and $T_c^{90\%-10\%} = (16.9 \pm 0.2)$ K for Nb_3Sn on copper with a 30 µm thick niobium buffer layer.

INTRODUCTION

In the pursuit for sustainability, one goal is to make possible the operation of the SRF cavities at 4.5 K to reduce the amount of cryopower needed for the operation [1]. To this aim, the A15 compound Nb_3Sn, with a higher critical temperature of 18.3 K compared to niobium's 9.2 K, has the potential to provide at 4.5 K quality factors comparable to the ones obtained with bulk niobium ($Q_0 \simeq 10^{10}$) at 2 K. Coating copper SRF cavities with a superconducting thin layer is a well-established technique, primarily using niobium coatings. However, the high temperatures required to form the A15 phase of Nb_3Sn ($\simeq 930$ °C) [2] introduce a major challenge when using copper as a substrate, so that optimizing the coating parameters becomes crucial to compensate for the limitations in reaching those high temperatures. Standardized procedures with high control and reproducibility are essential to identify factors influencing film quality and refine the R&D feedback process. The experimental methods adopted here, obtained results and future plans are detailed in the following sections.

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EXPERIMENTAL METHODS

Each Nb_3Sn film deposition process is performed on three different substrate types: sapphire, OFHC copper and OFHC copper pre-coated with a niobium buffer layer. Prior to the deposition of the Nb_3Sn film, the substrates are prepared according to a standard procedure: the sapphire substrates are cleaned with ethanol in an ultrasound bath; the copper substrates are first cleaned in a detergent solution (GP17.40) in an ultrasound bath, then treated with SUBU [3] for 2-3 minutes. A 1 µm thick niobium buffer layer is then deposited via DC Magnetron Sputtering (DCMS) on the prepared copper substrate. For the later tests presented in this study some copper substrates with a thicker 30 µm niobium buffer layer were also prepared and included in the deposition run, in addition to the ones prepared with the 1 µm niobium buffer layer. These were coated via DCMS as multi-layers [4]. The copper substrates pre-coated with the buffer layer are stored in vacuum until the Nb_3Sn coating process takes place, to minimize the chances of oxidation. In preparation to the coating, the substrates are then fixed on a custom plate, as shown in Fig. 1. Inside the ultra-high vacuum coating

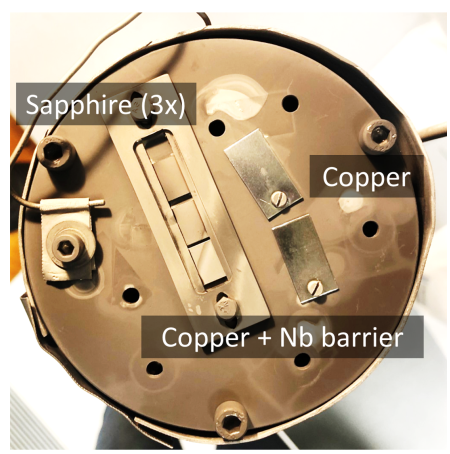

Figure 1: Flat samples right after the coating process, still mounted on the sample holder.

chamber, the plate is placed in front of a commercial 4" planar stoichiometric Nb_3Sn target. The distance between the target and the samples is 9 cm. The process takes place in three main phases: the baking, the coating phase and the annealing. The baking of the vacuum system is performed by heating the system for 48 h while pumping the vacuum. After the baking, the coating of the Nb_3Sn films

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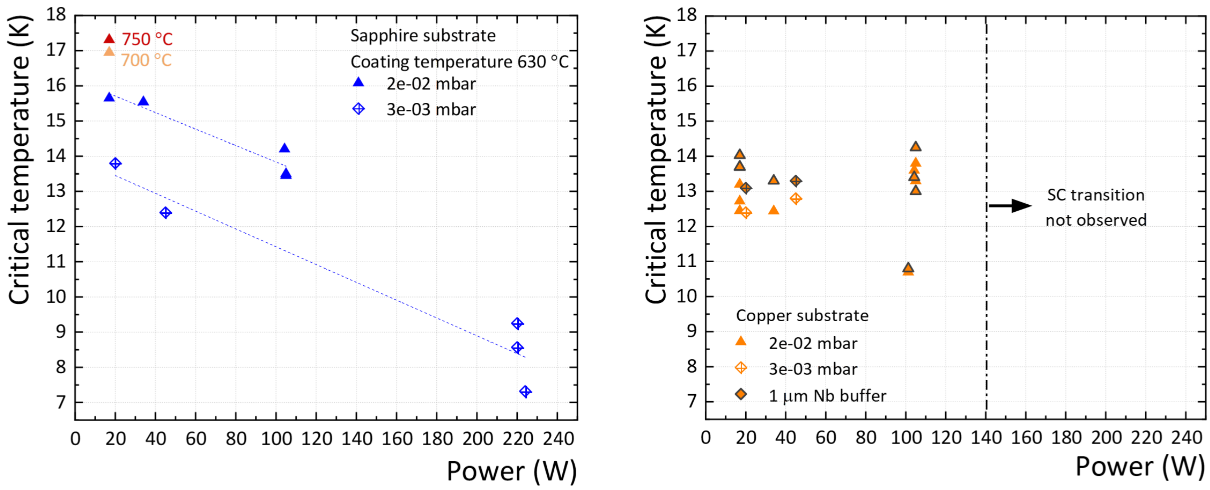

Figure 2: Dependence of the critical temperature of the Nb_3Sn films on the cathode power for the films on sapphire substrate (left) and on copper and copper with 1 µm niobium buffer layer (right).

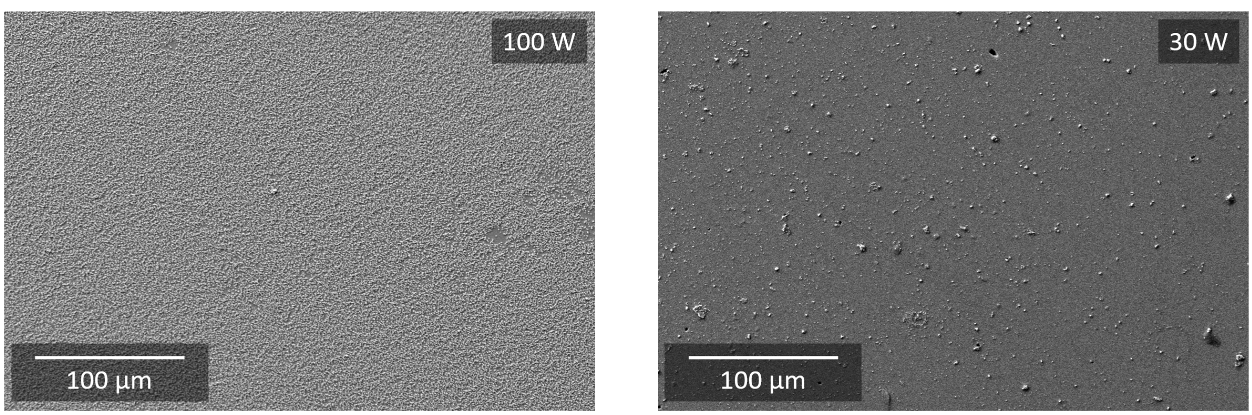

Figure 3: SEM images of the surface of a Nb_3Sn film on sapphire, deposited at high (left) and low (right) cathode power. The mitigation of surface tin clustering with decreasing power becomes visible in the sample deposited at low power.

takes place via DCMS using argon as sputtering gas. The process parameters such as the gas pressure, cathode power and substrate temperature are custom set for each coating run and are object of this study. Generally, the duration of the sputtering phase ranges between 6 h and 10 h. When the Nb_3Sn film deposition is over, the annealing of the samples is made by keeping them at the temperature at which the coating was made for a duration of 24 h, although shorter annealing times have been investigated whose results are presented later on.

The first quantity checked on the Nb_3Sn films is the critical temperature T_c. It is measured inductively via a double coil system, and extracted from the superconducting transition curve via the 90%-10% method, as it provides more conservative values compared to the curve onset value. The surface morphology and atomic composition of the films are assessed via scanning electron microscope (SEM) imaging and energy dispersive X-ray spectroscopy (EDS).

RESULTS AND DISCUSSION

The quality of the film is affected by the substrate type, so that in the same coating the Nb_3Sn films deposited on sapphire will exhibit different properties from the ones deposited on copper or copper with niobium buffer layer. Generally, due to the mechanical stability of the substrate and to the accordance of the crystalline lattice, the films grown on sapphire exhibit higher T_c, better texture and Nb-Sn ratio than the ones deposited on copper. For this reason, the films deposited on sapphire are taken as a reference for the effectiveness of the chosen coating parameters, especially in the early stages of the study when the results obtained from the films on copper do not show any strong dependence from the coating parameters.

The dependence of the critical temperature on the power applied to the target is shown in Fig. 2 (left) for the samples deposited on sapphire. It is likely that high cathode power induces tin evaporation resulting in final tin excess on the

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films which is likely to cause the observed degradation of the critical temperature with increasing power. This is supported by the SEM images of the film surface at different cathode power shown in Fig. 3, where tin clustering is observed to take place at high power (100 W) and decrease for lower power (30 W). Therefore lower power is preferred as sputtering condition. From Fig. 2 (left) it is also possible to see, on one hand, how the processes performed at higher pressure (2×10^{-2} mbar) exhibit a T_c which is systematically higher than the ones deposited at lower pressure (3×10^{-3} mbar) but same process temperature (630 °C), and on the other hand, how for the same pressure and power, increasing the coating temperature enhances the critical temperature. For this reason high pressure is also preferred as sputtering parameter, while higher temperatures cannot be pursued, despite leading to higher T_c, for reasons related to mechanical properties of copper as discussed below. No clear trend of this kind was observed on copper substrate, as visible in Fig. 2 (right). The diffusion of copper from the substrate into the Nb_3Sn film and the migration of tin from the film into the copper substrate are likely to have a major effect on the poorer quality of the films on copper substrate, at least partly justifying the observed lower T_c

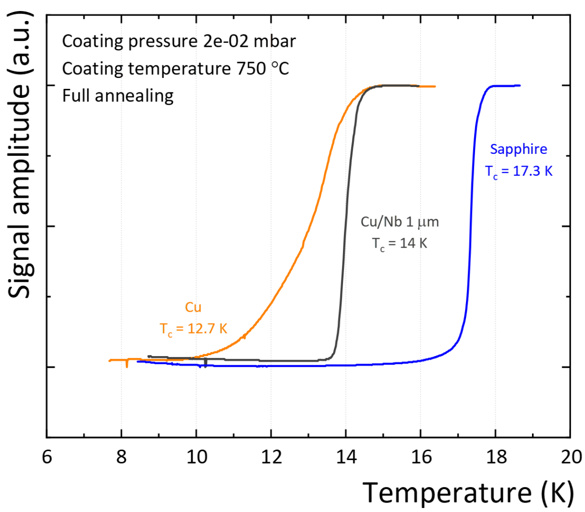

Figure 4: Superconducting transition curves of samples deposited at 750 °C for which full annealing was done.

and the absence of trends, observed instead on sapphire. The positive influence of the 1 μm niobium buffer layer can be appreciated in this context by noticing that the T_c is always slightly increased for the films deposited on it with respect to the ones deposited on copper in the same run. The fact that the buffer layer partly mitigates the Cu-Sn interdiffusion is also supported by the shape of the superconducting transition, as shown in Fig. 4. Although it was observed that the T_c of the films is increased by increasing the temperature of the coating process, this result was only partly satisfying, because it did not reproduce on copper, and neither would be applicable to large copper structures (such as cavities) if it did, as copper is known to start weakening at temperatures

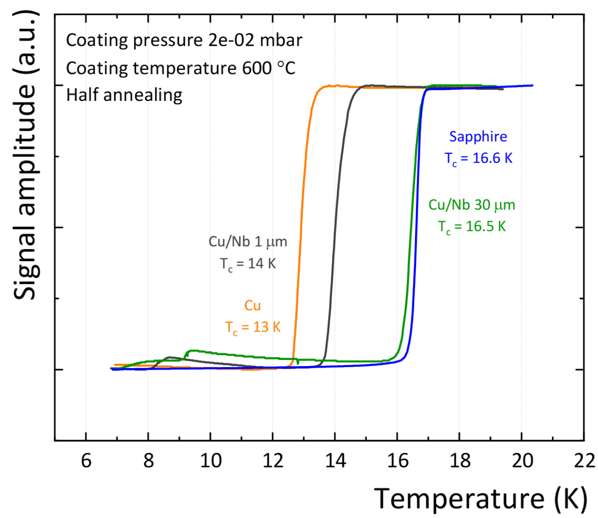

Figure 5: Superconducting transition curves of samples deposited at 600 °C for which the annealing time was reduced.

around 600 °C. Therefore, to improve the results on copper, two new aspects are currently being investigated: the effect of a thicker (30 μm) niobium buffer layer for a more effective prevention of the interdiffusion phenomenon, and the effect of shorter annealing times. The large improvement introduced by both the thicker buffer layer and shorter annealing can be observed in Fig. 5. The critical temperature values obtained for all different substrates are shown for a run performed at 600 °C and 2×10^{-2} mbar. One can see that, despite the lower coating temperature than the one for the samples shown in Fig. 4, which were coated at 750 °C, not only the obtained T_c with shorter annealing are all comparable (for the sapphire, copper and copper with 1 μm buffer layer substrates) but also the thicker buffer layer introduces a major improvement with respect to the thin one, with a final T_c comparable to the one obtained on sapphire. In Fig. 6, the T_c obtained for the coatings for which a full and short annealing have both been done are shown as a function of the annealing temperature, where the coatings shown in Fig. 4 are the ones corresponding to 750 °C annealing temperature and the ones shown in Fig. 5 correspond to 600 °C. The samples deposited on copper and on copper with 1 μm buffer layer do not exhibit any trend with the annealing temperature and duration. The T_c of the films on sapphire improves with the annealing temperature for the fully annealed samples, and the trend becomes even more obvious for the half annealed samples, remarkably at lower temperatures, for which $T_c^{90\%-10\%} = 17.9$ K and $T_c^{onset} = 18.1$ K were observed. The same trend is observed for the samples deposited on copper with 30 μm buffer layer and a major improvement in the value of T_c is obtained by the introduction of the thick buffer layer, resulting in $T_c^{90\%-10\%} = 17$ K and $T_c^{onset} = 17.7$ K. Further understanding is provided by Fig. 7, where the T_c distribution of the samples as a function of the tin atomic content is shown together with the literature data by Godeke [2]. The annealing temperature is also indicated (in degrees Celsius) next to the data points to show how the tin content

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seems to decrease with increasing temperature for all substrates, with the exception of the data points at 600 °C and 630 °C for copper and copper with 1 µm niobium buffer layer respectively. This temperature dependence is still under assessment, although the distribution of the data about the literature curve is encouraging.

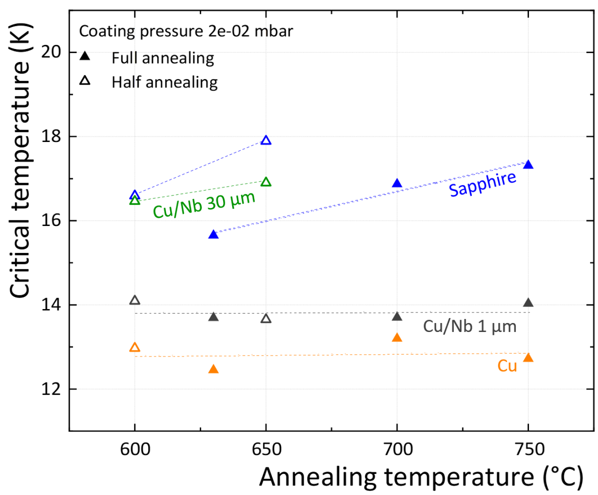

Figure 6: Effect of the annealing temperature and duration on the critical temperature of the Nb_3Sn samples on all substrate types.

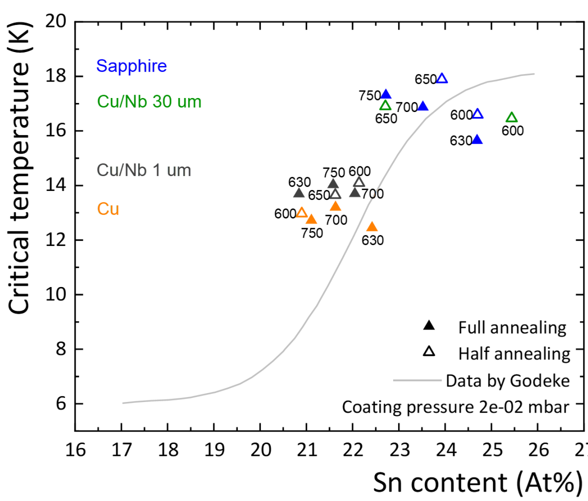

Figure 7: Distribution of the critical temperature of the Nb_3Sn samples as a function of the tin atomic content for all substrate types and both the fully annealed and half annealed samples. The annealing temperature is indicated (in degrees Celsius) next to the data points.

CONCLUSION

The R&D chain for the production of Nb_3Sn films deposited via DCMS is based on a continuous "characterization results - choice of next coating parameters" feedback loop, which allows for the optimisation of the coating recipe. According to the methods presented above, the aim is to produce a large data set including the coating parameters and the film characterisation results in order to study the dependencies and advance with the production of Nb_3Sn films on copper suitable for the end-use as SRF cavity material. The results obtained so far with the Nb_3Sn DCMS coatings from a planar, stoichiometric, commercial target performed at INFN-LNL, suggest high coating pressure (2×10^{-2} mbar in argon atmosphere) and low cathode power as starting point for quality coatings, sporting good T_c and stoichiometry on sapphire substrate. The expected improvement of T_c with increasing coating temperature is also observed on sapphire substrate, although this route cannot be pursued to obtain high quality coatings on copper as it would not be applicable to large structures such as elliptical cavities. To improve the quality of the coatings on copper and copper with niobium buffer layer two new routes are currently being investigated: the effect of a thick (30 µm) niobium buffer layer, and the effect of shortening the annealing times. Both these methods have proven successful at increasing the T_c of the films at the first attempts: not only the best results obtained on sapphire at 750 °C with full annealing were reproduced at 600 °C by shortening the annealing, but also, at the same temperature, the same results of sapphire were obtained on copper with niobium buffer layer by increasing the layer thickness from 1 µm to 30 µm. The best T_c results for the Nb_3Sn films obtained so far by investigating this route are $T_c^{90\%-10\%} = 17.9$ K on sapphire substrate and $T_c^{90\%-10\%} = 17$ K on copper substrate with thick buffer layer. These are preliminary results and cannot be taken as conclusive. The study is ongoing and the temperature dependencies are still under investigation, although the results can be considered encouraging towards the production of functional Nb_3Sn on copper exhibiting good superconducting properties. The next steps will include investigating the RF compatibility of the film surface.

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