Structural and RF properties of niobium films deposited onto annealed niobium resonators

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Introduction

Heat treatment at temperatures in excess of 1000°C is known to increase the residual resistivity ratio (RRR) of bulk niobium cavities and to reduce their residual surface resistance (R_{res}) accordingly. The low melting point of copper prevents the use of such a treatment in the case of niobium coated copper cavities (Nb/Cu). In order to overcome this problem, niobium coated niobium cavities (Nb/Nb) have been produced with the aim to study the evolution of the superconducting properties of the niobium film with increasing firing temperatures. In preparation for such a study, a number of preliminary steps have been taken and are reported in the present note. They include:

- the development of a firing procedure using titanium as solid state getter material, complemented with a Nb box to avoid Ti contamination of the Nb film,
- its use on bulk niobium cavities and the study of the resulting modification of their RF superconducting properties,
- the production of Nb/Nb cavities and the study of their RF superconducting properties prior to firing,
- studies performed on heat treated Nb/Nb samples, providing information on the evolution of the grain size with increasing firing temperature.

The study uses single cell resonators, operated at 1.5 GHz in the fundamental TM_{010} mode. Their surface resistance is parametrised in the usual form $R_s = R_{BCS} + R_{res} + R_{fl}$, where R_{BCS} is the BCS resistance and R_{fl} the resistance induced by the possible presence of trapped fluxons. The latter depends on the external magnetic field H_{ext}, applied along the cavity axis during cool down and on the RF field, H_{rf}. It is observed to take the form $R_{fl} = (R_{fl}^{0} + R_{fl}^{1}H_{rf})H_{ext}$ [1].

The cavities are produced from high purity niobium sheets (nominal RRR ~300) either by spinning or by electron welding of two deep drawn half cells. Buffered chemical polishing (BCP), using a standard solution of HF, HNO_3 and H_3PO_4, 1:1:1 in volume, is applied to remove at least 120 μm in order to suppress the damaged superficial layer. The niobium films, 1.5 μm thick, are grown on the inner walls of the resonators by sputtering in a cylindrical magnetron configuration at 150°C from a niobium cathode having a RRR of 300 [2]. Argon is used to establish the discharge at a pressure of 1.5×10^{-3} mbar and a current of 3 A. Before coating, the resonator and the cathode are baked out at 150°C for 20 hours. The ultimate pressure is typically 10^{-9} mbar, dominated by hydrogen.

In a few instances a different coating system, referred to as the double cathode system (DC) was used to suppress the thin oxide layer at the film - substrate interface. It includes a copper anode used to reverse sputter out from the cavity surface whatever impurities may be present over a depth of approximately 50 nm.

The firing procedure

Different cavities have been fired for 4 hours at 1000°C or 1100°C in a UHV furnace. The cavity to be treated is enclosed, together with test samples in a niobium box, itself enclosed in a titanium box, thus using titanium as getter material (Fig. 1). These two boxes are in turn enclosed in an external Nb box, ensuring an optimal gettering efficiency of the desorbed gases while protecting the cavity from the furnace residual pressure and preventing contamination of both the cavity and the furnace by titanium atoms. Secondary ion mass spectroscopy (SIMS) measurements of the titanium concentration at the surface of BCP polished samples, heat treated under identical conditions as the niobium cavities, indicate that the titanium contamination is below 100 ppm. After firing, the cavities are rinsed with ultra pure water at 100 bar for at least 60 min before being installed in a cryostat for RF measurement.

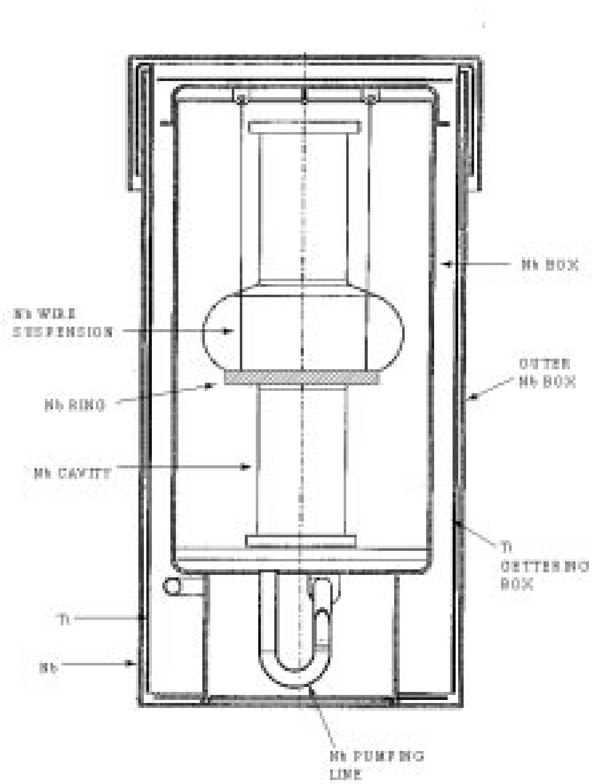
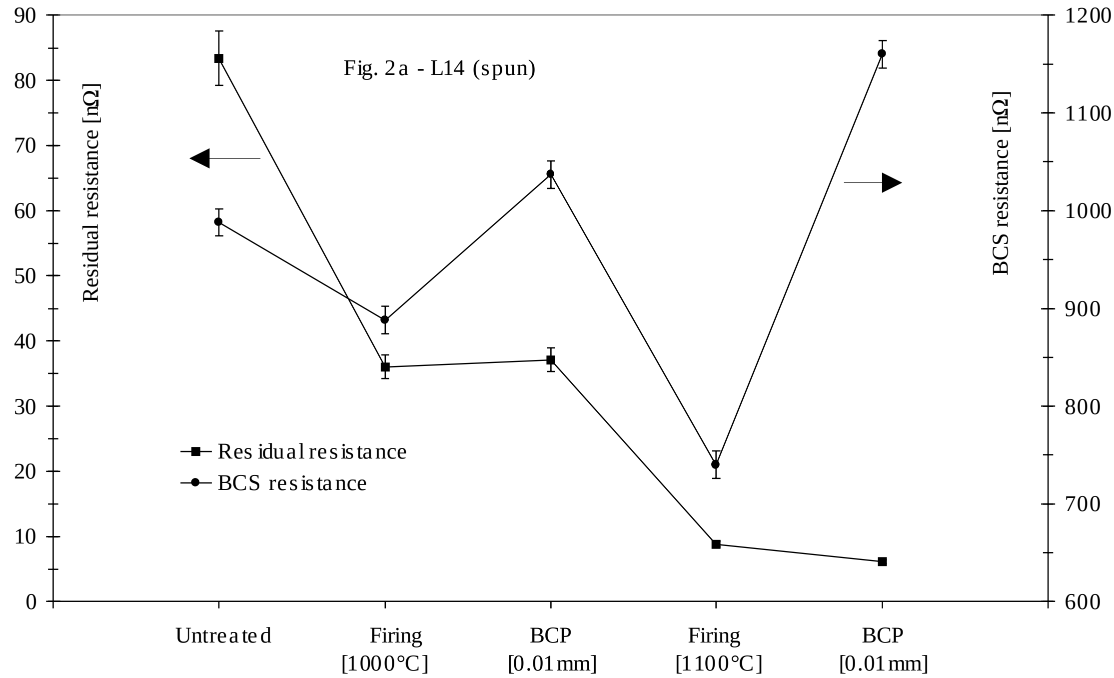


Figure 1: Schematic view of a cavity enclosed in a protection/gettering set-up

Effect of firing on the surface resistance of bulk niobium cavities

Four cavities have been studied, of which two (L14 and L15) were manufactured by lathe spinning at LNL [3], the other two (C1-1 and C1-2) being CEBAF cavities, made of two deep-drawn half-cells, electron-welded at the equator [4]. Figures 2a-d show the evolution of the residual and the BCS resistance with successive treatments. The observed decrease of the BCS resistance following heat treatment signals a corresponding decrease of the mean free path, suggesting a concentration of impurities (possibly titanium) in the thin superficial layer where the RF field penetrates. It is restored to a higher value after removal of a 5 to 10 μm layer by buffer chemical polishing (BCP). The residual resistance is not significantly affected by etching, suggesting that it is relatively insensitive to the presence of such impurities. The effect of firing on the residual resistance is important when its initial value is not too low (spun cavities), but no significant improvement could be achieved on the deep-drawn cavities which had a very small initial residual resistance.



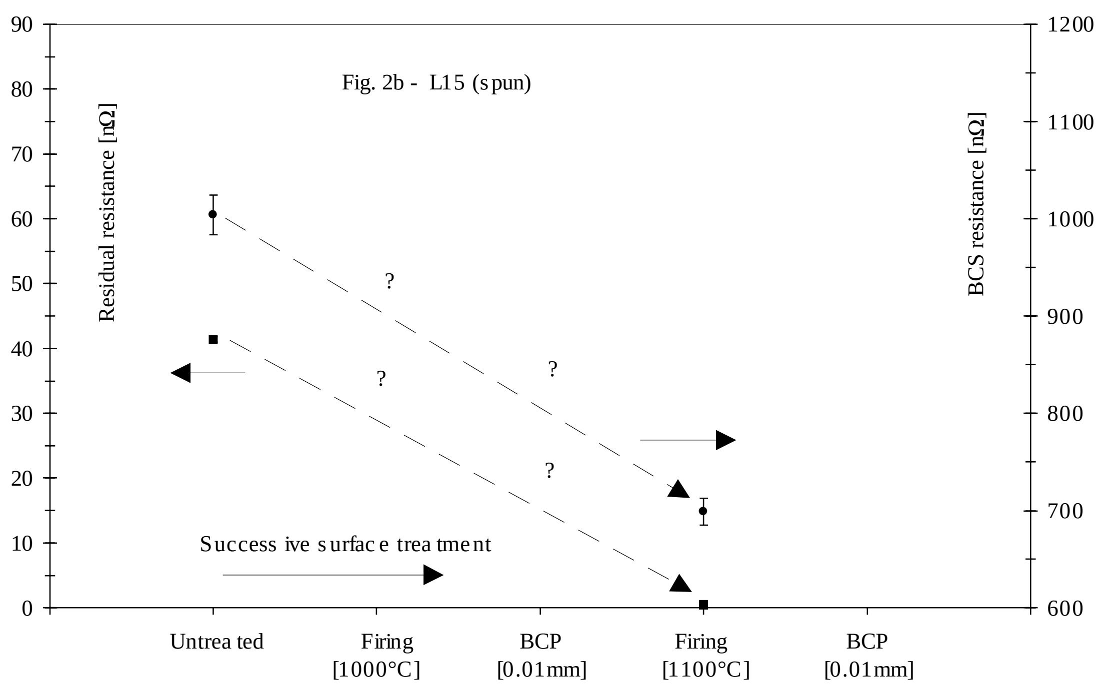
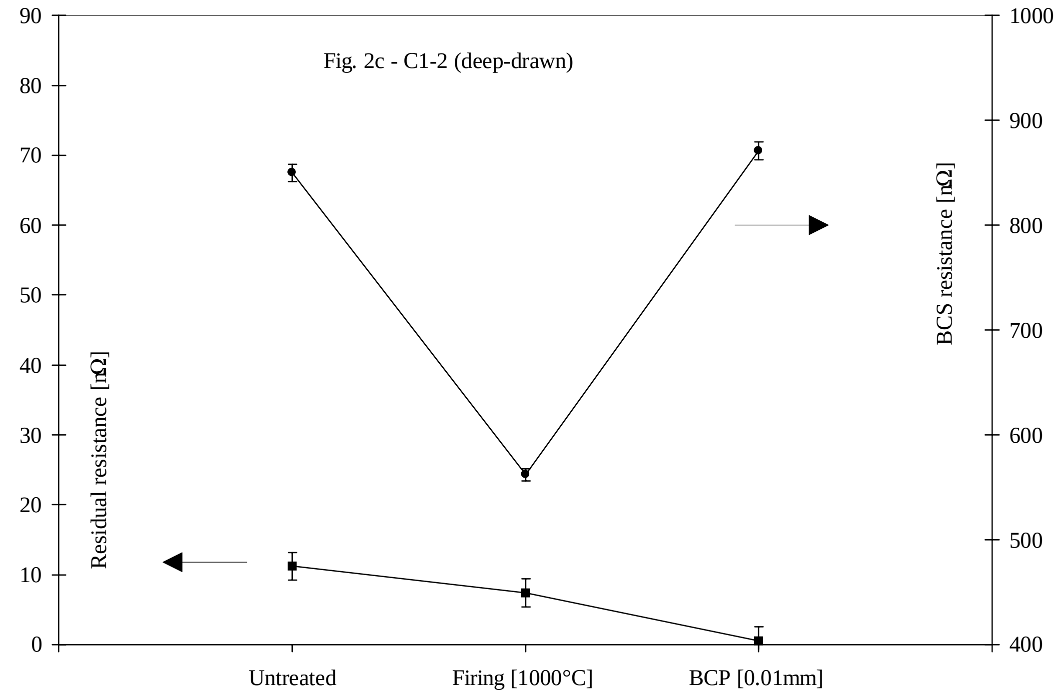


Fig. 2a-b : Effect of surface treatments of spun cavities on R_{res} and R_{BCS}. Cavity L15 (bottom) has been fired only at 1100°C, therefore no data at 1000°C are available. Small errors are included in the data points.



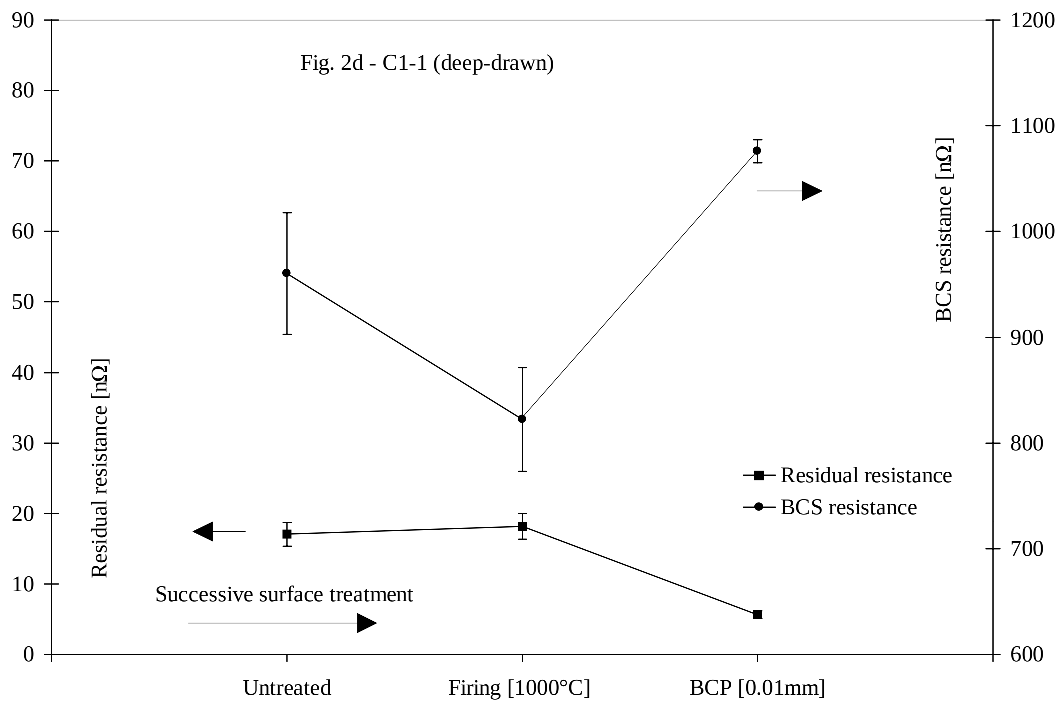


Fig. 2c-d : Effect of surface treatments of deep-drawn cavities on R_{res} and R_{BCS}.

A distinctive feature is the alteration of the fluxon trapping efficiency following firing. It is measured with Hall probes, distributed on a meridian of the outer cavity wall according to a procedure described in Ref. 1. Whereas the trapping efficiency for cavities before firing is always 100%, incomplete trapping is observed for fired cavities. More precisely, different trapping efficiencies are observed for different Hall probes, spanning the whole range between 0 and 100% and going hand in hand with a lack of reproducibility of the R_{fl}^{0} and R_{fl}^{1} measurements in successive trapping cycles.

Superconducting RF-properties of Nb/Nb cavities

Two cavities (L14.1, L15.1) have been coated using the standard system (SC) and two others (L14.2 and C1-2.1) using the double cathode system (DC). Table 1 compares their RF properties with those of bulk niobium and Nb/Cu cavities.

Cavity	**T_c [K]**	**R_{BCS} [nΩ]**	**R_{fl}^{0} [nΩ/G]**	**R_{res} [nΩ]**
L14.1 (SC)	---	571 ± 35	1.9 ± 0.2	10
L14.2 (DC)	9.33 ± 0.03	468 ± 11	64 ± 8	17
L15.1 (SC)	---	514 ± 10	7.4 ± 0.2	23
C1-2.1 (DC)	9.29 ± 0.03	510 ± 9	123 ± 4	129
Bulk Nb(fired)	9.28 ± 0.08	981 ± 13	≥ 100	0-80
Nb/Cu (SC)	9.54 ± 0.06	401 ± 1	4.8 ± 0.1	0-100
Nb/Cu (DC)	9.47 ± 0.08	466 ± 8	54 ± 4	20-100

Table 1 : Comparison among relevant parameters for bulk Nb, Nb/Nb and Nb/Cu cavities, coated in the single and double cathode system. Bulk niobium and Nb/Cu data represent an average of at least four different cavities.

The Nb/Nb BCS resistance at 4.2K is much closer to that of Nb/Cu, than to that of bulk cavities, indicating the importance of the intrinsic film properties in comparison with the nature of the substrate. The residual resistance is similar to those usually obtained for Nb/Cu spun cavities. Whereas R_{BCS}, R_{fl} and T_c have nearly identical values for films prepared in the same way, R_{res} display instead large differences. For this reason, the quoted R_{res} values are only indicative. The similarity between Nb/Cu and Nb/Nb films is also visible in their response to trapped fluxons with low R_{fl}^{0} values for films grown on the standard coating system and larger R_{fl}^{0} values for films grown on the double cathode system.

Annealing of niobium films - Sample characterisation

Samples have been prepared by sputter coating a 1.5µm niobium film on small rectangular niobium substrates (RRR~300). XRD spectra have been recorded in the θ-2θ mode to evaluate the lattice parameter of the Nb film. The obtained value of a(z) = 3.32214 ± 0.001Å (compared to a_{ref} = 3.30322Å for bulk Nb) indicates an expansion of the film in the z-direction caused by the sputtering process itself [5]. The observed increase of T_c in the films (Table 1) can be explained by the resulting residual stresses [6]. In the Nb/Cu case T_c is even higher by 0.21 K because of an additional thermal contribution (different expansion coefficients for film and substrate) to the lattice distortion.

The coated samples were heat treated in steps, ranging between 700 and 1400°C, and analysed in order to estimate the impurity content of the film and to obtain information about the lattice parameter and the evolution of the grain size with temperature. The analytical methods which have been used are glow discharge optical emission spectroscopy (GDOS), X-ray diffraction (XRD) and scanning electron microscopy (SEM). The average grain size, estimated from image analysis of scanning electron micrographs is of the order of 200 nm for films and 50 µm for bulk. Anomalous grain growth (formation of grains with preferred orientation at the expense of adjoining, smaller grains) starts at 1100°C for the film and at 1300°C for bulk niobium. The final grain size at 1400°C is >2mm for bulk and ~20µm for film niobium (Fig. 3). SE-micrographs (Fig. 4) show the structure of the film at 1000°C (top) and when anomalous grain growth appears (bottom). GDOS analysis of the Nb/Nb interface indicate that the anomalous grain growth is accompanied by the disappearance of the oxide layer at the film/bulk interface.

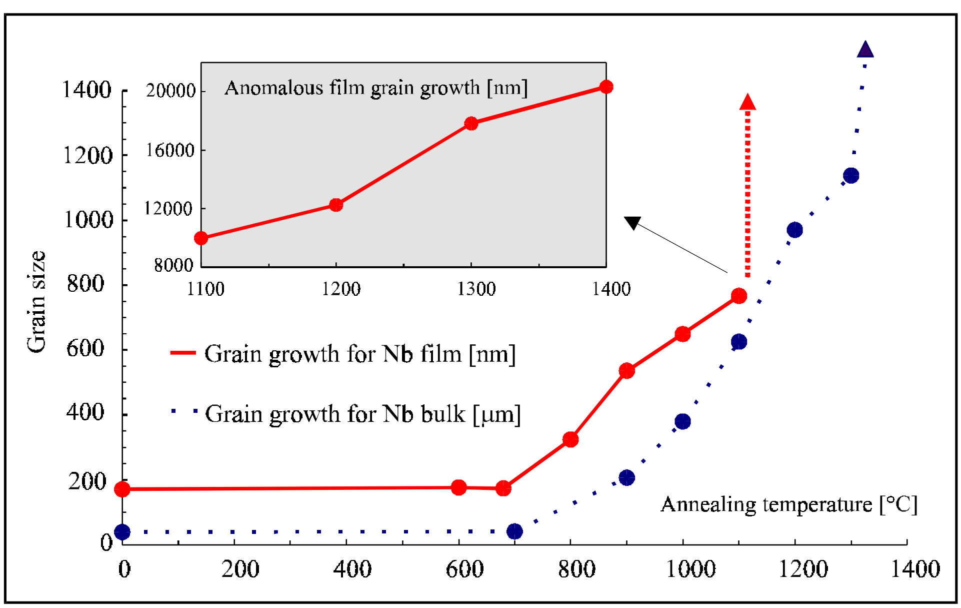


Fig 3.: Evolution of the grain size of the substrate and of the film (1.5µm) as a function of the temperature of thermal treatment.

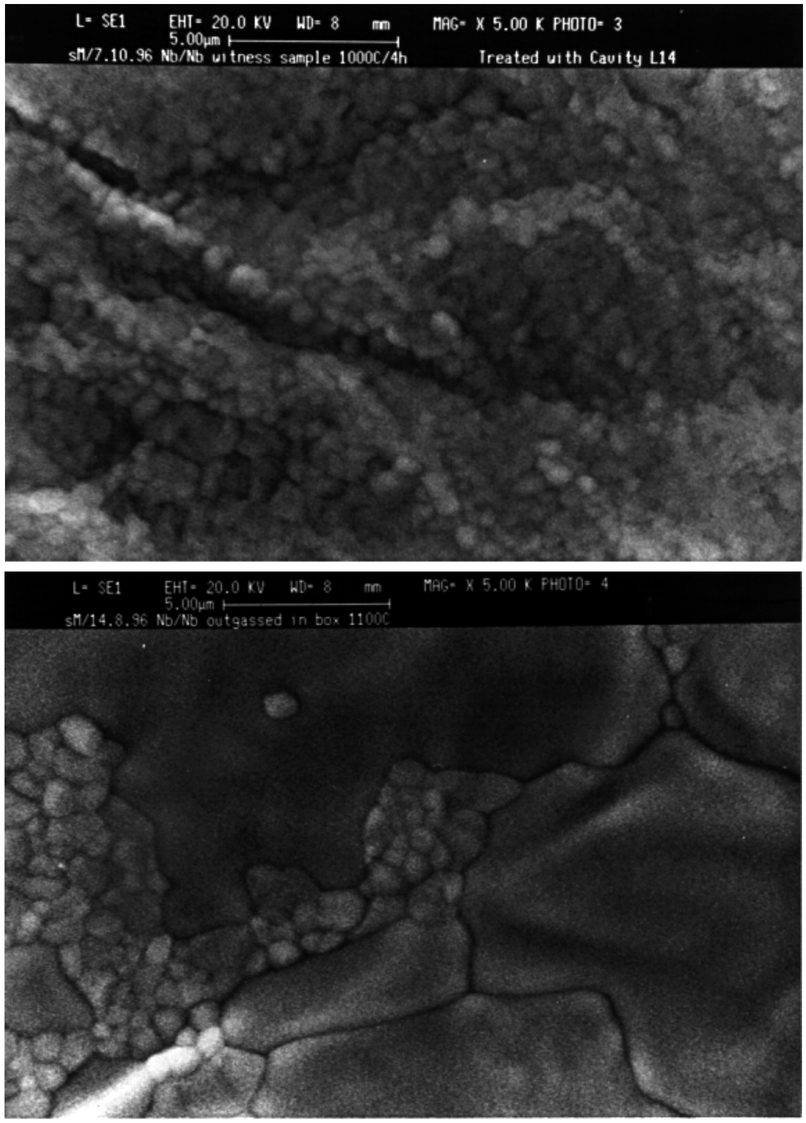


Fig 4. : SE-micrographs showing the structure of the film at 1000°C (top) and when anomalous grain growth appears (bottom).

Conclusion

Furnace treatment of Nb bulk resonators at about 1000°C in a Nb/Ti/Nb getter box, followed by a light chemical polishing, results in a significant change of their RF properties, revealing the increase of the RRR value obtained after this process. Niobium films deposited on fired bulk niobium resonators show properties which are similar to those of niobium films deposited on copper. Analysis of heat treated Nb/Nb samples show similar recrystallization behaviour for the film and for the bulk. Ongoing RF tests on Nb/Nb resonators will soon be able to ascertain if the RF and magnetic properties of the film also merge with those of the bulk in the same temperature range.

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