Study of the Residual Resistance of Superconducting Niobium Films at 1.5 GHz

C.Benvenuti, S.Calatroni, P.Darriulat, M.A. Peck, A.-M. Valente[#] and C.A. Van 't Hof,
CERN, EST/SM, CH-1211 Geneva 23, Switzerland

Abstract

A study of the main potential contributions to the residual surface resistance of niobium-coated cavities is reviewed. They are the formation of hydride precipitates, the contamination by sputter gas atoms and the presence of macroscopic defects in the film, induced by defects in the substrate. It will be shown that residual resistances as low as those obtained for bulk niobium can be achieved, together with a very small dependence on the amplitude of the RF field.

1 INTRODUCTION

The presence in a superconductor of non-magnetic impurities and defects with dimensions exceeding the coherence length induces the loss of the overall coherence of the superconducting state. When sensed by microwaves such impurities and defects may cause additional losses, which persist at zero temperature. The generated residual resistance term will depend on their geometry and location and on the density of free electrons which they contain (normal conductors and insulators are expected to behave in different ways).

The aim of the present work is the study of the residual resistance of niobium films coated on the inner walls of 1.5GHz resonators of design similar to that used at DESY or CEBAF. In a previous publication [1], in the framework of a general study of the surface resistance, no direct correlation has been found between the residual resistance and the other superconducting parameters. Arguments were also presented against the hypothesis [2-4] that most of the residual resistance of niobium films should be blamed on their small grain size, at variance with the bulk case. This statement, inspired from results obtained on granular high T_c superconductors [5], postulates the presence of “weak links” at grain boundaries and implies the existence of a fundamental limitation to the production of low residual resistance niobium films. The results presented here provide additional experimental evidence against such hypothesis.

This study explores various mechanisms, which are potential contributors to the generation of a non-zero residual resistance. These include defects on the substrate prior to coating, the oxidation of the film surface, or the presence in the film of impurities such as hydrogen or atoms of the noble gas used for the sputtering process.

Particular attention is given to the study and understanding of the dependence of the residual resistance on the amplitude of the microwave. In practical applications as particle accelerators this may be an important limitation to the performance at high fields. The value of the residual resistance is obtained from the measurement of the surface resistance at 1.7K with no trapped field with the BCS contribution subtracted. In most cases the residual resistance is well described by a linear dependence on the magnetic component H_{RF} of the microwave of the form $R_{res} = R_{res}^{0} + R_{res}^{1} H_{RF}$. Beyond a certain threshold H_{fe}, field emission may occur, signalled by X-rays and electron emission. Its effect implies the addition of a nearly exponential term to the above expression of the residual resistance [1,6,7]. This is illustrated in Fig. 1 where typical data are shown together with the result of the fit restricted to an interval $H_{low} \le H_{RF} \le H_{fe}$. Here H_{low} expresses occasional deviations from the linear fit in the low H_{RF} range which however, most of the time, are small enough to be ignored. As field emission is usually due to the accidental presence inside the cavity of small foreign particles, the present work is restricted to the interval $0 \le H_{RF} \le H_{fe}$ where field emission does not occur.

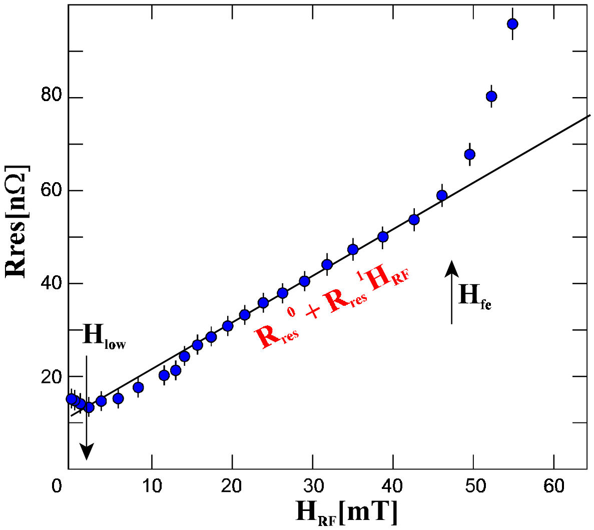

Figure 1: Dependence of the residual resistance on H_{RF} for a typical film. The solid line represents the fit used to define R_{res}^{0} and R_{res}^{1}.

The experimental procedures for the production of 1.5µm niobium films grown on the inner wall of copper cavities, the measurement of their superconducting properties, in particular of their surface resistance, using

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[#] E-mail: Anne-Marie.Valente@cern.ch

1.5 GHz microwaves and their characterisation in terms of a limited number of variables are described in Ref. [1] and summarised in the present Proceedings [8].

2 MACROSCOPIC FILM DEFECTS

2.1 Substrate surface quality

The roughness of the substrate is known to enhance the residual resistance of the films grown on the inner wall of radiofrequency cavities [1]. Films grown on spun copper seem to provide on average better results than films grown on hydroformed copper both in terms of residual resistance at zero field and its dependence on RF field (the improvement is about 13 nΩ for R_{res}^0 and 1.2 nΩ/mT for R_{res}^1). The main difference is the average roughness of the substrate, respectively 0.2µm and 0.8µm for spun and hydroformed cavities.

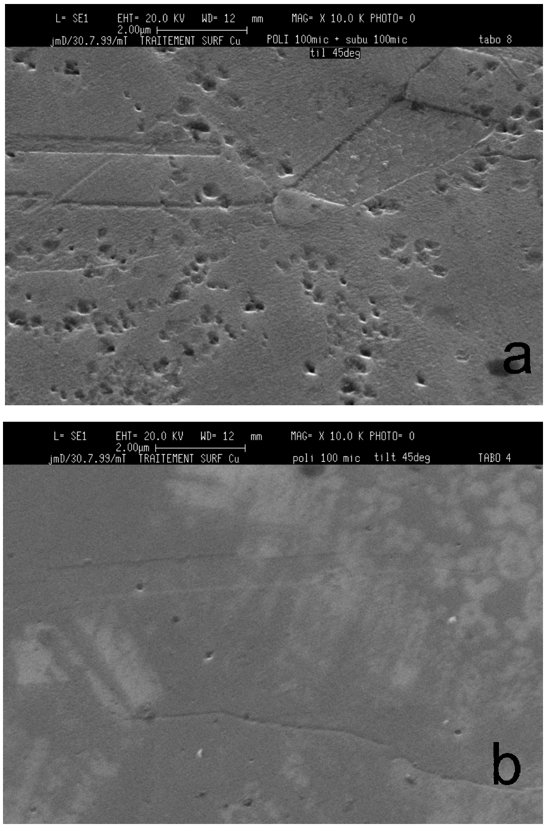

Figure 2: SEM micrographs for (a) chemically polished and (b) electropolished copper substrates

As the average incidence angle of sputtered niobium is about 15°, reaching up to 50° near the iris, irregularities of the substrate induce film inhomogeneities in their shadow [9]. The influence of the incidence angle θ_{sp} on the film quality has been investigated by measuring the He leak rate through bare niobium films. This experiment has shown a significant increase of the film porosity with the incidence angle. The fraction of the "leaky" film surface increases from ≅ 4.4 ppm at the equator ($\theta_{sp} \cong 9°$) to ≅ 25 ppm at the iris ($\theta_{sp} \cong 50°$).

In addition to the intrinsic roughness of the substrate, accidental damages caused by defective polishing can play an important role. The standard method of chemical polishing described in Ref. [1] is known to produce pinholes, typically 0.3 µm in diameter. This is clearly illustrated on Fig. 2a, which shows micrographs of samples that have undergone chemical polishing. In order to improve the surface quality of the substrate, chemical polishing has been replaced by electropolishing. The main advantage of this method is that it does not produce macroscopic defects as it can be observed on Fig. 2b. This has led to a significant improvement in residual resistances (Fig. 3). The values obtained this way are comparable to those obtained with bulk niobium.

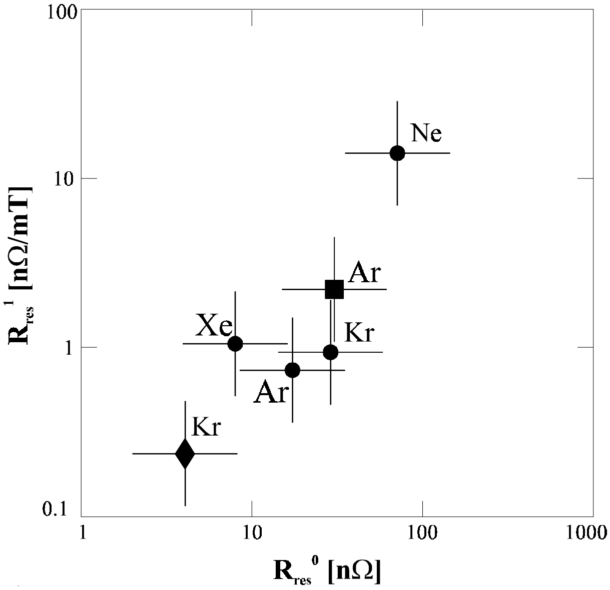

Figure 3: Each point represents the average of the residual resistance for films coated with various gases, as indicated by the label. Shown are films grown on hydroformed chemically polished copper (squares), spun chemically polished copper (circles) and spun electropolished copper (diamond)

2.2 Water rinsing

In order to reach high gradients it is essential to work in dust free conditions. Before and after coating, cavities undergo a high-pressure water rinsing. Different tests have been performed on several cavities without rinsing or with rinsing performed on systems having different cleanliness. In some cases, by an adequate rinsing in dust free conditions, the maximum reachable accelerating field was improved by a factor of 2 and the residual resistance could be lowered by up to a factor of 3. In order to provide the best conditions during the rinsing process, the usual system has thus been upgraded for closed cycle operation and a particle counter at the output of the cavity monitors the cleanliness.

3 OXIDES, HYDROGEN, NOBLE GASES

The presence in a film of impurities such as oxygen or hydrogen can be expected to be a source of residual resistance. Their effect has been studied extensively in the case of bulk niobium. Another source, specific of films, is noble gases implanted during the sputtering process.

3.1 Oxide on niobium surface

In practical applications such as RF cavities it would be technically difficult to prevent the formation of a thin oxide (Nb_2O_5) layer on the film surface. In the case of bulk niobium, such a layer is harmless and does not produce more than 1.5 nΩ of residual resistance [10,11]. The cavities considered in the present study are vented to dry air after sputtering as soon as they have cooled down to room temperature. The thickness of the resulting oxide layer measured by Auger electron spectroscopy has been found not to exceed 6 nm. Moreover, values of R_{res}^0 and R_{res}^1 lower than 1 nΩ and 0.2 nΩ/mT respectively have been obtained without taking particular precaution to prevent oxidation after coating. No significant effect has been observed when a thin protective alumina overlayer has been grown on top of a Nb film to prevent the formation of an oxide layer. This gives evidence that the oxide layer on the Nb surface is not more harmful than in the bulk niobium case.

3.2 Oxide at the Nb/Cu interface

In the standard LEP2 procedure, the substrate surface is passivated at the end of the chemical preparation, and then exposed to (dust-free) air. An oxide layer is therefore present at the interface between the niobium coating and the copper substrate. A special two-cathode coating system has been used to coat several cavities. It allows either in-situ sputter etching of the oxide or coating of a copper underlayer. Oxide removal from the substrate is considered good practice in thin film technology. When applied to standard niobium films coated on copper by keeping all other conditions unchanged, higher RRR (about 30) have been obtained compared to standard films with an oxide layer (about 10). Analyses by X-ray diffraction and transmission electron microscopy (TEM) have revealed a totally different structure for the two types of films. A complete analysis of stress and texture by X-ray diffraction will be given in a further publication. It has shown that standard films have a typical fibre texture while the texture of films grown on oxide-free copper is influenced by heteroepitaxy. TEM observations in plane view have also revealed that whereas films grown on oxidised copper show small crystallite size (in the range of 100 nm), films grown on oxide-free copper (sputter-etched substrate or copper intermediate coating) provide grains in the range of some microns (Fig. 4).

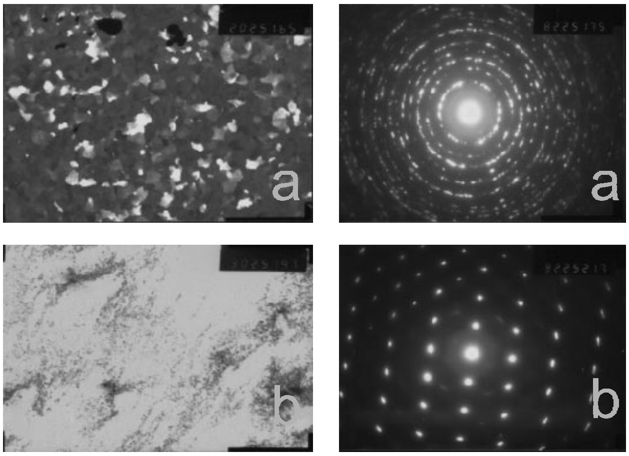

Figure 4: TEM micrographs (×20000) and their corresponding diffraction pattern for (a) standard and (b) oxide-free films

It has been found that, on average, films coated on oxide-free copper do not display smaller residual resistances than films on oxidised substrate [12,13]. This is additional very strong evidence against the conjecture that small grain sizes generate high values of R_{res}^0 and R_{res}^1. In Fig. 5 are shown values of R_{res}^0 and R_{res}^1 for spun cavities coated using argon, krypton or xenon as sputtering gases.

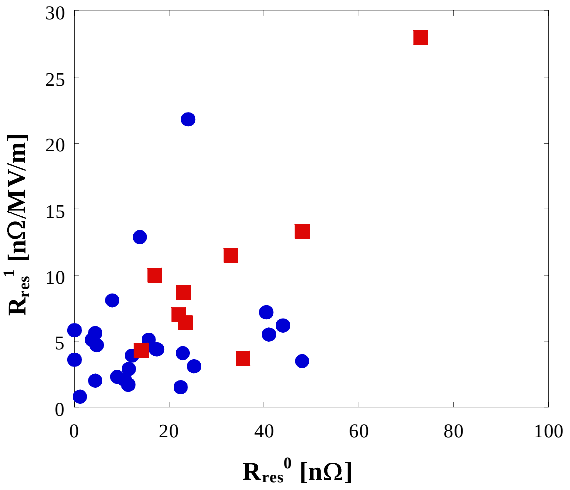

Figure 5: Residual resistance R_{res}^0 and its RF field dependence R_{res}^1 for standard cavities (dots) and for cavities without oxide interface (squares).

3.3 Hydrogen

Hydrogen is known to be a potentially important cause of residual resistance for bulk niobium cavities [14-16]. This effect is understood to result from the formation of hydride precipitates with typical dimensions ranging between several hundred Å and a few µm, and occurring during cavity cooling between ≅ 70 K and ≅ 170 K. This effect can be prevented by rapid cool-down, crossing the range of dangerous temperatures fast enough to make sure that the dissolved hydrogen atoms have no time to be expelled from the niobium grains before their mobility is reduced to negligibly small values. Using niobium of low residual resistivity ratio, where impurities and defects act as trapping centres for hydrogen, can also reduce this effect. Different experiments are being carried out in order to study the effect of hydrogen in the niobium film case. Some preliminary results are quoted here. The outgassing at 350°C of niobium films has revealed that hydrogen is naturally present in a comparable amount both in oxidised and oxide-free films (0.07±0.01at.% and 0.09±0.01at.% respectively). However, the two films might have a different efficiency for hydrogen trapping. A few films of the two types were loaded with hydrogen and their residual resistances have shown a different degradation, reaching at times several µΩ . The different behaviour may be related to different trapping mechanisms for hydrogen (dislocations, impurities such as oxygen…). The niobium cathode can be an important source of hydrogen in the coating process. Niobium cathode outgassing prior to the coating has at times provided an improvement in residual resistance. Further studies are however needed to obtain a complete description of the behaviour of hydrogen in our niobium films.

3.4 Noble gases

Sputtered films are known to contain atoms of the noble gas used in the sputtering discharge. The issue was amply discussed in Reference [1] and the study of the dependence of the noble gas concentration on its atomic mass and on the stresses present in the film (i.e. in practice on the nature of the substrate) will be published elsewhere. Whereas the other superconducting properties take defined values for a defined gas, the influence on the residual resistance of noble gas is difficult to evaluate. The residual resistances do not significantly differ from one gas to the other, except for neon, as shown in Fig. 3. However, large spreads are observed between films grown with same sputtering gas and prevent the detection of a small difference between two different types. Therefore, small differences between xenon, krypton and argon data cannot be excluded. In the case of neon, the residual resistance increases significantly due to the large concentration of gas present in the film, in the range of a few atomic percents instead of a few ppm for Kr and Xe or a few hundreds ppm for Ar. However, noble gases implanted in metals are known to precipitate into clusters. As further, the pinning properties of the film [12] suggest that at least part of the trapped gas tends to cluster in solid form. It would be natural to expect that the trapped noble gas atoms contribute also to the generation of a non-zero residual resistance term. However, the solid "bubbles" have small dimensions, typically 3 nm, that is to say one tenth of a coherence length and they are not expected to contain free electrons.

4 DISCUSSION

As a result of the present study films have been produced with very low residual resistances maintained over a broad range of RF field amplitudes, comparable to what is obtained with clean bulk niobium. These reached accelerating gradient in excess of 20 MV/m, with quality factors exceeding 10^{10} at 10 MV/m and of the order of 5.10^{9} at 20 MV/m (Fig. 6).

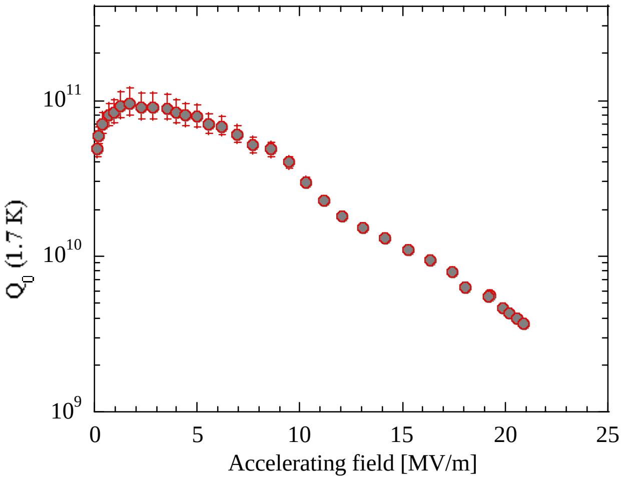

Figure 6: Quality factor of high performance cavity as a function of the accelerating gradient. The coating was performed with krypton as sputtering gas

The main factor of residual resistance improvement is the smoothness of the substrate and absence of defects. Rather than weak links at grain boundaries, the main cause for the significantly larger residual resistances which were commonly obtained in the past is now identified as being the presence within the film of islands of non superconducting defects. Of course a contribution of grain boundary weak links at the level of a few nΩ for R_{res}^{0} and a few tenths of nΩ/mT for R_{res}^{1} cannot be excluded. Nevertheless, above such a level the small grain size characteristic of sputtered films cannot be blamed for the occasional generation of important residual resistances. One should remark that only a fraction of the data displayed in Fig. 4 was obtained with the upgraded rinsing installation. The data collected earlier were usually limited by field emission to accelerating gradients ranging between 10 and 15 MV/m. This situation is very reminiscent of that of bulk niobium data [17,18]. There is thus no reason to suspect the existence of any fundamental limitation that could prevent reaching even higher gradients if yet cleaner conditions were achieved in the production process.

5 CONCLUSION

The main results of the present study are the production of niobium films having very low residual resistances over a broad range of microwave amplitudes and the identification of the smoothness of the substrate as the main factor of this achievement. These results have important implications for the large-scale production of RF cavities for particle accelerators. The present study provides ample evidence that film technology suffers no limitation compared to the bulk technology. However, more R&D efforts are required to optimise the substrate surface quality by investigating other manufacturing procedures than spinning, which is really damaging for copper. One of these procedures could be the electroforming. The use of other materials as substrate, like aluminium, should also be considered.

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