Niobium Coating Techniques

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Abstract. We will give a historical overview of the niobium on copper sputtering technology for RF cavities and discuss the main advantages and disadvantages with respect to bulk niobium cavities. Some highlights of the present understanding will be given and some recent developments in the coating technology will be discussed.

1. The LEP cavities

The development of the deposition of Nb thin films onto Cu cavities with the sputtering technique has started at CERN in 1980 [1], the target application being of course the LEP collider, operating at 352 MHz. At that time, the main reasons for undertaking such an approach were: a) a better thermal stability of the cavity (resistance to “quench”) thanks to the much higher thermal conductivity of the OFE copper substrate compared to the superconducting niobium; b) greatly reduced material cost for the fabrication; c) safer handling for the chemical surface treatments; d) possibility of foreseeing higher Tc coatings than Nb (NbTiN, V3Si, Nb3Sn; HTS not being known at that time).

After first studies using the diode sputtering technique at 3 GHz and 500 MHz, the magnetron sputtering technique was adopted in 1985 (figure 1). This technique, where a magnetic field is superposed (crossed) to the electric field thus increasing the ionization rate, allows much lower sputter gas pressure and cathode voltage compared to diode sputtering [2] and is highly beneficial in terms of film purity, structure and overall quality. This allowed a significant breakthrough in performance with film cavities showing a higher Q at low field than bulk ones. The Q factor decreased then more strongly with field compared to bulk Nb cavities, finally attaining comparable values at typical maximum field levels. In those days, accuracy and cleanliness of surface preparation were not as accurate as can be done today, and it was rare that the accelerating field reached a value higher than 8 MV/m. It should be noted that Nb bulk cavities were produced in the eighties starting from raw material having RRR of 40, resulting in a very limited thermal conductivity at cryogenic temperatures. Nb bulk cavities were thus usually limited by quenching even at those rather low field levels, phenomenon which was instead completely avoided in Nb/Cu cavities. Films showed also an unexpected advantage, in that their surface resistance is almost insensitive to the Earth’s magnetic field. As an order of magnitude the effect is 100 nΩ/Gauss of external magnetic field for bulk Nb, and only 1 nΩ/Gauss for films. This allows for the fabrication of much simpler and cheaper cryostats without the need of complex magnetic shielding of the cavities.

The establishment of an adequate chemical polishing procedure [3] was a further important development in order to improve the copper surface smoothness and promote film adhesion, compared to the simple acid etching used previously. This was first tested with 500 MHz cavities and then
chosen for the production of the 352 MHz prototype cavities for LEP. The results showed a performance even better than Nb bulk at the field level specified for LEP (figure 2). Eight pre-series 4-cell cavities for LEP were built at CERN, the remaining 264 were made by three European industrial suppliers.

Figure 1. First magnetron coating system for 500 MHz cavities. (From [2]).

LEP was operated at 4.5 K and 352 MHz, where the BCS surface resistance and the residual surface resistance have roughly the same magnitude, i.e. about 20 nΩ at zero field. Approaching 0 K the BCS term vanishes exponentially while the residual term being constant with temperature remains dominant. The residual resistance happens to increase with field much strongly for films than for bulk, translating into a “slope” in Q(E) plots. This “slope” has become the most known feature of films, hindering sometimes the acknowledgement of several other important features and advantages.
2. R&D after LEP

The same Nb/Cu magnetron sputtering technology has been applied for the LHC cavities. Sixteen cavities (single cell, 400 MHz) are installed in the LHC. No particular developments were done for this project, apart from the obvious adaptation of the technique to a different geometry. Nevertheless, the progress in surface preparation, final water rinsing and the overall improvement in cleanliness allowed exceeding the specification values, reaching routinely fields in excess of 10 MV/m. It was rather clear from this experience that the electron-field-emission limitation to the maximum achievable accelerating field was not intrinsic. This was in line with what observed in parallel by the bulk-Nb community [5].

After the development of the LHC cavities, two main lines of research have been pursued at CERN starting from 1995. The first one was devoted to applying the magnetron sputtering technology to elliptical accelerating cavities for particles of $\beta<1$ (for proton linear accelerators). In this effort CERN complemented the successful activity pursued in particular by LNL on quarter wave resonators (QWR) [6]. The second line was devoted to studying the ultimate performance that can be reached with the magnetron technology in terms of Q and accelerating field at 1.7 K in cavities for electron linear accelerators.

2.1. Low-$\beta$ cavities

Several elliptical cavities of 352 MHz frequency and with $\beta$ values ranging from 0.48 up to 0.8 have been manufactured and coated, by applying with minor modifications the usual magnetron sputtering technique [7]. A summary of the results is illustrated in figure 3.

The increase in “slope” with decreasing $\beta$ is essentially due to the residual resistance, which in turn was proven in this case to be related to the average and peak incidence angle of the niobium atoms impinging on the substrate during film growth, the more grazing the worse. The effect happens also to be strongly influenced by the roughness of the substrate and results in a very poor granularity and an enhanced roughness of the film. This phenomenon

![Figure 2. Performance of prototype LEP 352 MHz cavities, bulk Nb and Nb/Cu. (From [4]).](image)
triggered several investigations at CERN and at LNL [8], and although a general consensus is not yet established it seems that there is a threshold value for the angle of incidence, above which the RF performance starts to be degraded. The phenomenon should thus have only a marginal impact on the residual resistance of standard $\beta=1$ cavities. This is confirmed by RF tests and temperature-mapping of Nb-coated Nb cavities, which showed that the dissipation is uniform over the entire cavity surface [9], thus supporting the threshold-effect model for the angle of incidence.

![Figure 3](image.png)

**Figure 3.** Best results at 4.2 K with low-$\beta$ cavities, all limited by amplifier power (adapted from [7]).

As already mentioned Nb films have also been applied to lower-$\beta$ quarter wave resonators, essentially by LNL-INFN. The problems encountered in coatings are of the same type as mentioned above, albeit in a completely different geometry, and could be minimised by using a biased diode sputtering approach. This allows a good quality of the coating even in recessed areas. After successful prototyping, a first series of eight resonators for the high-$\beta$ section of the ALPI accelerator was constructed and operated in the machine for several years at nominal performance and without major failures. This led to the decision of upgrading the 46 resonators of the medium-$\beta$ section, which had been electrolitically Pb-plated, by stripping the previous film and depositing a new sputtered Nb film [10]. Since 2003 the accelerator runs exclusively with Nb/Cu cavities. Figure 4 illustrates the gain in accelerating field due to the Nb coating of the medium $\beta$ resonators compared to the previous Pb coating, for the same total power dissipation of 7 W.

It should be mentioned that QWR Nb/Cu resonators present a distinctive advantage over their bulk Nb counterparts, since by using very thick Cu material they can be made extremely stiff and thus much less sensitive to microphonic effects which are quite difficult to compensate in this geometry, and thereby allowing significant cost savings compared to hypothetical similar bulk-Nb resonators.
2.2. **Ultimate performance in $\beta=1$ cavities**

The search for ultimate performance was carried out on single-cell 1.5 GHz resonators after first encouraging results obtained by a CERN-CEA/Saclay collaboration [11], and was essentially focussed until 1999 in identifying whether the standard superconducting quantities have any influence on the residual resistance. More than 200 test coatings have been carried out using adapted magnetron coating technology, and completely characterized in RF. It turns out that the residual surface resistance is not at all correlated with the measurable superconducting quantities [12]. This result was supported by a large wealth of material studies carried out on samples, such as SEM, TEM, XRD and composition analyses, as well as the classical superconductivity characterizations.

Comforted from this result, the work was focussed from 1999 onwards into improving the quality of the copper surface preparation, by pioneering the electropolishing of the full cavity, in order to have the smoothest possible surface. Previous results with chemical polishing and different techniques for cavity manufacturing (hydroforming, half-cell welding, full-cavity spinning, electroforming) already gave indications that this was the right road for the improvement of the surface resistance [12, 13]. In parallel, the high-pressure water rinsing facility at CERN was improved and optimised for the treatment of these cavities. It should be mentioned that the large world community working on Nb-bulk cavities was proving at the same time that the maximum field, below the superconductor critical field, is function only of surface cleanliness and freedom from defects (incidentally, the same HPWR facility developed for Nb/Cu cavities has been used for the first European high-quality fully electropolished TESLA-type Nb-bulk cavities [14]). The outcome of these efforts proved to be fruitful [15] as illustrated in figure 5.

However, even if the performance was greatly improved from the LEP-era values, the “slope” of the residual resistance was still present. This was a limit to the maximum achievable field because of the high RF power dissipation, contrary to what happens for Nb-bulk cavities due to their much less pronounced “slope”. A maximum accelerating field of 28 MV/m could nevertheless be attained in a few ad-hoc experiments with large power.
amplifiers and excess cryogenic capacity. In general phenomena like quenching or field emission never occurred on properly treated Nb/Cu cavities.

2.3. Search for the origin of the residual resistance

The CERN studies have next focussed in finding the possible causes of the “slope”. One should underline first that some models predict that such a “slope” is inherent in films because of the limited electron mean free path compared to bulk. This should manifest either in a reduction of $H_{c1}$ and thus nucleation of (Abrikosov) fluxons [16] in a rather low RF field, effect possibly enhanced by demagnetization due to surface roughness. Or it could manifest itself in a depression of the superconducting gap due to a reduction of the critical superfluid velocity [17], this transforming directly into an increase of the BCS surface resistance. Both phenomena do clearly happen in films, however it is difficult to estimate a priori their importance.

Nb/Cu cavities develop thermoelectric currents if a temperature gradient is present over the cavity body when becoming superconducting. These currents can be quite large, inducing large flux densities which are then trapped in fluxons, which in turn contribute to the residual resistance [18]. Careful estimates of this phenomenon had been carried out in the past but never published before. It resulted that in typical 1.5 GHz cavities a temperature gradient of 1 K over its length is equivalent to a trapped magnetic flux of 0.6 Gauss. The residual resistance of Nb/Cu cavities is known to be less sensitive to trapped magnetic fields than for bulk cavities, but in the latter thermoelectric currents do not develop at all. Careful controlled cooling is of course the cure to the problem, and it is easily achievable on bath cooled cavities. However this problem has never been addressed for QWR resonators, which are cooled partly by conduction, and might by a serious extrinsic performance limit.

Much effort has been devoted to identifying whether the hydrogen trapped in the film was a possible cause of the “slope”, since this has always been a primary source of losses in bulk Nb cavities. The quantity of hydrogen contained in the films, depending on the coating procedure, has been measured accurately, as well as its binding state. The largest possible sources, i.e. the Nb cathode and the copper substrate, have also been characterised fully and suitable means to reduce their hydrogen content have been found [19]. Further ways of reducing the hydrogen content of films by means of NEGs have been devised. Unfortunately hydrogen reduction was not effective [20] for reducing the “slope”.

Further efforts have been devoted in determining whether the Nb/Cu interface introduces a thermal

Figure 5. State-of-the art performance of Nb/Cu cavities at 1.7 K and 1500 MHz (adapted from [15]).
barrier, such that the “slope” would be produced by a thermal runaway effect [21]. Accurate measurements on samples showed that Nb coated specimens have the same thermal conductivity (in the direction normal to the surface) at 1.7 K as the naked substrate, be it Cu or Nb [20].

A third line of thought lies in further optimising the roughness and the structure of the film, having in mind the flux penetration mechanism mentioned before. Copper electropolishing was put under firm control by elaborated numerical simulations and chemical analyses, and it is not believed that this could be optimised any further [20]. The roughness of the substrate has strong influence on the roughness of the film, and self-shadowing effects during film growth may lead to poorly connected Nb film grains, possibly enhanced by a non-normal angle of incidence. Granularity effects have always been seen as a major source of trouble in literature, either because of possible losses in weak-links [22], or because of easier penetration of (Josephson) fluxons [16].

This leads naturally to the idea of introducing important changes to the coating technique, with the aim of optimising the smoothness of the films at the crystal grain scale and minimising the density of defects. Several developments are being pursued at present in various Laboratories.

3. Future research and development

A first simple step towards improving the film quality is by adding a bias to the classical magnetron configuration, for having a Kr ion bombardment during film growth. This should produce smoother films and has been tested at CERN. First results did not show however significant changes in RF performance. A confirmation comes from similar tests on samples carried out at LNL [23].

A further possibility is to create the film using Nb ions, instead of neutrals such as in sputtering, directed to the substrate by a bias thereby allowing conformal deposition with a normal angle of incidence everywhere and thus suppressing self-shadowing. Several promising techniques have been selected by different Laboratories and are under development or being tested. Cathodic arc is being developed by INFN-Roma2 [24] and ECR post-ionisation of evaporated Nb is being pursued by TJNAF [25]. CERN developments are concentrated on High Power Pulsed Magnetron Sputtering (HPPMS).

3.1. High Power Pulsed Magnetron Sputtering (HPPMS)

HPPMS is an evolution of the magnetron technique which relies on ~100 µs high-voltage pulses of the order of ~1 kV, compared to the ~350 V of the standard DC magnetron process [26]. During the pulse a very large power density is deposited onto the target, of the order of a few kW/cm² compared to a few 10s of W/cm² of the standard DC process, producing a highly dense plasma in which a fraction of the Nb atoms is also ionised, attaining values close to 100% in the best cases [27]. These ions can in turn be attracted to the substrate with a suitable bias to produce the coating. The repetition rate should be of the order of a few 100s Hz in order to keep the same average power and coating rate as in the equivalent DC sputtering process. A further advantage of the technique is the fact that no hardware modifications are required to a standard DC biased magnetron system, except for the obvious replacement of the power supply.

First experiments at CERN have been carried out in a classical planar magnetron system using a 1 Hz repetition rate power supply (surplus from old pulsed LINAC magnets). The implementation of the technique is fairly smooth and coatings can be obtained from the first run. Figure 6 illustrates typical pulse values obtained in a test run. It should be underlined that the results depend greatly on the power supply adopted and that the one used was clearly not optimal and its specifications underrated. The sputtering parameters are however in the good ballpark, and the films obtained have RRR and Tc similar to films produced with the same coating system using DC sputtering [28]. There were however no significant changes in the film morphology, probably because of an insufficient ion fraction in the coating, estimated at 1% from the ratio of sputter and bias current, assuming a sputtering yield of 1.

A drawback encountered in the process was the generation of arcs, which were not quenched by
adequate circuitry within the power supply and even resulted in cathode damage. This appears to be the major obstacle in the correct implementation of the HPPMS coating technique [29]. A suitable power supply, with higher power, faster repetition rate and a sophisticated arc-suppression circuit is being studied at CERN in collaboration with HES-SO (Yverdon, Switzerland) and the design is scheduled for completion by end 2007. A decision on its construction will then be taken depending on market availability of similar items, on the latest literature results and of course on CERN strategic interest and availability of funding.

4. Conclusions and outlook

In the opinion of many authors, niobium films have not yet achieved their possible ultimate performance, contrary to what has now been obtained with niobium sheet cavities, and this hinders at present their use for electron linacs although their cost is far inferior. Several novel developments in the coating technique are under study which, on the grounds of the present understanding, may produce an important leap forward both scientifically and technologically.

Films are however mature for all applications wherever ultimate performance is not the goal, typically operated at 4.2 K, allowing significant cost reduction, hardware simplifications, and more stable operation. This includes high energy accelerators like the LHC at CERN, synchrotron light sources like SOLEIL (CEA, France), and low energy ion accelerators like ALPI at INFN-LNL.

CERN has recently approved the HIE-ISOLDE project, in order to extend the energy reach of the RIA beams for the ISOLDE facility up to 10 MeV/u. QWR Nb/Cu accelerating cavities have been chosen for this machine for their better projected operational flexibility, and optimum cost-performance balance. The coating technique will probably rely on the LNL biased diode technology, but a development of the magnetron technology to this geometry is also foreseen in parallel. Prototyping is scheduled to be completed by end 2009, followed in the next two years by the first phase of construction of the machine, with 15 accelerating cavities.

Figure 6. Sputter voltage (top line), bias current (middle line) and sputter current (bottom line) during a typical HPPMS pulse.
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