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**20 YEARS OF EXPERIENCE WITH THE Nb/Cu TECHNOLOGY FOR
SUPERCONDUCTING CAVITIES AND PERSPECTIVES FOR FUTURE
DEVELOPMENTS**

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The first niobium-coated copper cavities were produced at CERN in the early eighties. The sputter technology was chosen, first in the pure diode configuration and subsequently in the magnetron configuration, which was adopted for the successful series production of the LEP and LHC cavities. In parallel, an intensive R&D effort was undertaken at CERN and other Laboratories in order to understand the advantages and limitations of this technique. Some highlights of the present understanding will be given. Several new developments in the coating technique are being pursued around the world, which will be discussed together with their motivations.

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Abstract

The first niobium-coated copper cavities were produced at CERN in the early eighties. The sputter technology was chosen, first in the pure diode configuration and subsequently in the magnetron configuration, which was adopted for the successful series production of the LEP and LHC cavities. In parallel, an intensive R&D effort was undertaken at CERN and other Laboratories in order to understand the advantages and limitations of this technique. Some highlights of the present understanding will be given. Several new developments in the coating technique are being pursued around the world, which will be discussed together with their motivations.

THE LEP ERA

The development of the sputtering technique for the deposition of thin Nb films onto Cu cavities 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 the following: a) Better thermal stability (resistance to “quench”) thanks to the much higher thermal conductivity of the OFE copper substrate compared to the superconducting niobium; b) Reduced material cost; c) Possibility of applying high T_c coatings (NbTiN, V3Si, Nb3Sn, HTS...).

First developments started at the frequencies of 3 GHz and then 500 MHz. The coating technology employed was the bias diode sputtering configuration. The diode technique, working at a very high sputter gas pressure and high (a few kV) voltage, produced films that were not very compact and were strongly columnar in grain shape. The film structure could nevertheless be somewhat improved by negatively (~ 100 V) biasing the substrate using a third electrode, resulting in an ion bombardment of the growing film.

It should be noted that Nb bulk cavities were produced in the eighties with sheets having RRR of 40, resulting in a very limited thermal conductivity at cryogenic temperatures. Typical performance of bulk cavities at 500 MHz and 4.2 K was $Q \approx 2.5 \times 10^9$ (≈ 100 n Ω) at low field, approximately decreasing by a factor 2 at fields of the order of 10 MV/m, where quenching usually happened. Film cavities immediately showed the important advantage of a higher Q at low field ($\sim 3.5 \times 10^9$) than bulk ones (Fig. 1). This is due to a lower BCS surface resistance, in turn related to a normal state

Figure 1: Typical $Q(E_a)$ curves of niobium sheet metal (line) and niobium sputter coated copper cavities (hatched) in the 80's. (From [2]).

electrical resistivity close to the theoretical optimum (Fig. 2). The Q factor decreased more strongly with field compared to bulk Nb values due to the residual component, but remained comparable to the bulk Nb at field levels of interest for LEP. 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 values higher than 8 MV/m.

A new development was started in 1985 with the magnetron sputtering technique [4], which allows operating at a much lower sputter gas pressure and cathode voltage by increasing the ionization rate, thanks to the addition of a magnetic field crossed with the electric field (see Fig. 3). A further important development was the establishment of an adequate chemical polishing solution in order to improve the

Figure 2: The BCS surface resistance at 1.5 GHz as a function of niobium purity. The abscissa is equal to 1 in the limit of electron mean free path $l \rightarrow \infty$. (From [3]).

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Figure 3: Sketch of sputtering cathodes for the diode configuration (a) and for the magnetron configuration (b). (From [5]).

copper surface smoothness and promote film adhesion, compared to the simple acid etching used previously. Both developments were first applied to 500 MHz cavities and then chosen for the production of the prototypes of the 352 MHz cavities for LEP. The results showed an even better performance compared to Nb bulk (Fig. 4).

Only operation at 4.2 K was relevant for LEP. At that temperature, the BCS component and the residual component of the surface resistance have roughly the same magnitude at 352 MHz, about 20 nΩ. At 1.7 K the BCS component of the surface resistance reduces exponentially and the residual term remains dominant. Although of comparable order of magnitude between

Figure 4: Performance of prototype LEP 352 MHz cavities, bulk Nb and Nb/Cu. (From [6]).

Figure 5: Typical performance of industry-produced LHC 400 MHz cavities. Specification values are $Q=2 \times 10^9$ at $E_{acc}=5.5$ MV/m and 4.5 K.

sputtered film and bulk at zero-field, the residual resistance had a stronger increase with field in the case of films, thus showing a “slope” in Q .

Films showed a further 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 cavities.

Eight pre-series 4-cell cavities for LEP were built at CERN, the remaining 264 were made by three European companies.

THE POST-LEP ERA

The same technology has been applied for LHC cavities. Sixteen cavities (single cell, 400 MHz) will be 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 and the overall improvement in cleanliness allowed exceeding the specification values, and reaching routinely fields in excess of 10 MV/m (see Fig. 5). It was rather clear from this experience that the electron-field-emission limitation to the maximum achievable accelerating field was a problem of cleanliness and accuracy of the final water rinsing, and no intrinsic limitation was inherent to the films. This was in line with what observed in parallel by the bulk-Nb community [7].

After the developments for 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 cavities of rather low frequency for accelerating particles of $\beta < 1$ (for proton linear accelerators). The second line was devoted to studying the ultimate performance that can be reached with the magnetron technology in terms of Q and maximum accelerating field at 1.7 K (for electron linear accelerators).

Figure 6: Best results at 4.2 K with low- β cavities, all limited by amplifier power (adapted from [8]).

Low- β cavities

Several tests have been done on elliptical cavities of 352 MHz frequency and with β values ranging from 0.48 up to 0.8, by applying with minor modifications the usual magnetron sputtering technique [8]. A summary of the results is indicated in Fig. 6.

The increase in “slope” with decreasing β is essentially due to the residual resistance, which in turn was proven 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 triggered several investigations at CERN and in other Laboratories [9], 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 standard $\beta=1$ cavities. In parallel, RF tests and temperature-mapping on Nb-coated Nb cavities led to the conclusion that the source of the dissipation is uniform over the film surface [10], thus supporting the threshold-effect for the angle of incidence.

It should be underlined that Nb films have also been successfully applied to lower- β quarter wave resonators, exceeding the required performance and with good reliability [11]. 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.

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 [12], and was essentially focused until 1999 in identifying whether the standard superconducting quantities have any influence on the residual resistance. A number of test coatings in excess of 200 have been carried out using a magnetron coating system and completely characterized in RF. It turns out that the residual surface resistance is

not at all correlated with measurable superconducting quantities [3]. 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 first 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 improvement of surface resistance [13, 14]. In parallel, the high-pressure water rinsing facility at CERN has been improved and optimised for the treatment of these small cavities (the same facility has been used a few years later for the first European high-quality fully electropolished TESLA-type Nb-bulk cavities). The outcome of these efforts proved to be fruitful [15] as illustrated in Fig. 7.

However, even if the performance was greatly improved from LEP-era values, the “slope” of the residual resistance was still present. This was limiting the achievable maximum field because of the high power dissipation, resulting in helium boil-off, or simply saturating the available power amplifiers. A maximum accelerating field of 28 MV/m could nevertheless be attained in an ad-hoc experiment in a large volume cryostat. Phenomena like quenching or field emission never occurred on properly treated Nb/Cu cavities. Moreover, the large world community working on Nb-bulk cavities was proving at the same time that the maximum field is function only of the cleanliness of surface preparation, and the performance could be

Figure 7: State-of-the art performance of Nb/Cu cavities at 1.7 K and 1500 MHz (adapted from [15]).

extended to unprecedented high fields thanks to the fact that bulk Nb does not show a similar “slope” [16].

Search for the origin of the residual resistance

The activity has next been 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 [17] 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 [18], this transforming directly in an increase of the BCS surface resistance. Both phenomena do clearly happen in films, however their importance is difficult to estimate *a priori*. It is then important to study in depth the film material parameters and all possible methods to alter them in a way that can be correlated to changes to the “slope”.

Much effort has thus 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 NEG's have been devised. Unfortunately hydrogen reduction was not effective [10].

Further efforts have been devoted in determining whether the Nb/Cu interface introduces a thermal barrier, such that the “slope” would be produced by a thermal runaway effect [20]. 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 [10].

A third line of thought lies in further optimising the roughness and the structure of the film. 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 [10]. 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 [21], or because of easier penetration of (Josephson) fluxons [17].

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

FUTURE RESEARCH AND DEVELOPMENT

A first simple step towards improving film quality is adding a bias to the classical magnetron configuration for having an ion bombardment during film growth. This should produce smoother films and is being tested at CERN. First results did not show however significant changes in RF performance.

A further possibility is to create the film using Nb ions, instead of neutrals such as in sputtering, attracted to the substrate by a bias, thereby allowing conformal deposition with a normal angle of incidence everywhere and thus suppress self-shadowing. The most promising techniques have been selected by different Laboratories and are under development or are being tested.

High Power Pulsed Magnetron Sputtering (HPPMS)

HPPMS is an evolution of the magnetron technique which relies on $\sim 100 \mu s$ high-voltage pulses of the order of ~ 1 kV compared to the ~ 300 V of the standard DC magnetron process [22]. During the pulse a huge power density is deposited onto the target, of the order of a few kW/cm^2 compared to a few W/cm^2 of the standard DC process, producing a highly dense plasma in which also the Nb atoms are partially ionised. These can in turn be attracted to the substrate with a suitable bias. A further advantage of the technique lies in the fact that no hardware changes are required compared to a standard DC biased magnetron system, except for the obvious replacement of the power supply. Experiments are under way at CERN in a classical planar magnetron system using a low repetition rate power supply, and indications whether the process should be implemented for cavity coating will be available before the end of the year 2005.

Cathodic arc

Another promising approach is the one of UHV cathodic arc [23], pursued by INFN. In this coating technique an electric arc is established over the cathode's surface by a suitable trigger (high voltage or laser pulse).

Figure 8: UHV arc coating set-up with 90° magnetic filter for 3-cell cavities (courtesy R. Russo – INFN).

The arc is sustained by an adequate power supply, with the plasma plume containing Nb atoms at such a high density that they are fully ionised. Discharge gas is not needed in this case, contrary to classical sputtering, resulting in no trapped impurities. The technique provides a high coating rate, and of course the coating flux can be attracted at the desired energy towards the substrate. A strong disadvantage is however the formation of macroparticles because of the explosive nature of the process. Magnetic filtering and steering of the ion flux is then needed, in order to remove macroparticles from the flux and to obtain a defect-free coating, adding complexity to the coating system.

Nb coatings have already been performed on Cu samples which have been fully characterized. An important difference of “energetic” coatings compared to sputtered films is the absence of preferential orientation of the crystallites, whose growth in the case of sputtering is instead dominated by the lattice free energy resulting in a (110) texture. Moreover the grains are several microns in size, compared to a few hundred nm in the case of sputtering, and completely free from defects and microstrain, which are a major source of electron scattering in sputtered films.

Work is progressing towards the construction of a filtered UHV arc coating system for RF cavities, either with a planar arc as a source (see Fig. 8), or using a cylindrical arc which is somewhat similar in geometry to the standard DC magnetron system. Prototype cavity coatings are expected by the end of 2005.

ECR post-ionization of evaporated Nb

A different approach, pursued at JLAB, consists in first creating a flux of Nb atoms by e-beam evaporation, and then ionise it by an ECR process. The ions can then be steered to the suitably biased substrate by magnetic guidance [24]. The advantage compared to arc deposition is the total absence of macroparticles, albeit at the expense of a much lower coating rate which calls for an extremely clean XHV environment. The vacuum conditions, and in particular the hydrogen partial pressure during the process, must anyway be kept under tight control in “energetic” coatings. The virtually defect-free films produced by these techniques will in fact result in a much higher mobility of hydrogen that may reflect in a degradation of RF performance.

Encouraging results in terms of film quality have been obtained on small samples by this technique, and work is progressing towards the coating of a full cavity (see Fig. 9), expected in the course of 2006.

CONCLUSIONS AND OUTLOOK

Superconducting cavities produced by the magnetron sputtering technology have been successfully used at CERN, and are also employed in several other present or future accelerator facilities, such as ALPI or SOLEIL for acceleration, or ELETTRA and SLS for 3rd harmonic bunch lengthening. There are clearly defined sets of

Figure 9: Schematic set-ups of evaporation + ECR coating system for small samples and for cavities (courtesy G. Wu – JLAB).

accelerator machine parameters where films show a clear advantage compared to bulk niobium, in particular for low frequencies or for operation at 4.2 K. Niobium films have however not yet achieved their possible ultimate performance, contrary to what has 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 coating technology are however under study which, on the grounds of the present understanding, may produce an important leap forward.

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