# UHV ARC FOR SUPERCONDUCTING NIOBIUM FILM DEPOSITION

R. Russo, A. Cianchi, R. Polini, S. Tazzari, University of Rome “Tor Vergata” and INFN-Roma2, Rome, Italy
L. Catani, INFN-Roma2, Rome, Italy
F.Tazzioli, Laboratori Nazionali di Frascati INFN, Frascati(RM),Italy
J. Langner, The Andrzej Soltan Institute for Nuclear Studies, Swierk, Poland
N.N.Koval, Institute of High Current Electronics, Tomsk 634055, Russian Federation

## *Abstract*

The coating of thin niobium film on copper RF cavities is a very interesting alternative to the bulk-Nb cavities, since copper is cheaper than niobium, has higher thermal conductivity and better mechanical stability. The technology of magnetron sputtering for coating copper cavities with superconducting thin film was successfully used for the production of 350 MHz LEP accelerating cavities at CERN. Unfortunately, the observed degradation of the quality factor with increasing cavity voltage restricts the use of sputtered accelerating cavities in future large accelerators working at gradients higher than ≈15 MV/m.

Vacuum arc coating is known to be a powerful technique to produce films on several materials [1]. Its main advantages compared to standard sputtering are the ionized state of the evaporated material, absence of gases to sustain the discharge, high energy of the atoms reaching the substrate surface and possibility to have very high deposition rates. For the arc ignition it is necessary to produce a small plasma plume of sufficient density to form a high-conductivity path between cathode and anode. We have used for ignition a Nd-YAG pulsed laser focused on the cathode surface, which provides a reliable system and allows eliminating all possible sources of contaminants. We have proven that the arc technique produces bulk-like films suitable for superconducting applications. Its main disadvantage is the production of macroparticles that deposit on the film, can increase its roughness and may induce field emission. We present our recent results on the characterization of niobium films produced by UHV cathodic arc under different conditions, focusing mainly on the study of the macroparticle phenomenology.

## INTRODUCTION

Super conducting (SC) cavities for particle acceleration are mainly based on Nb bulk technology. The Nb/Cu technology was proved a valid alternative, for relatively low accelerating fields (up to ≈ 10 MV/m), by the successful operation of the LEP II 200GeV acceleration system. With respect to bulk Nb, Nb coated Cu offers several advantages: better mechanical stability, lower cost, better thermal stability, easier conditioning on the machine, easier connection to the cryostat, less sensitivity to magnetic fields. So far though, the quality factor of magnetron sputtered cavities slopes down with increasing electric field [2], thereby limiting their usefulness to the new very high-energy, high field SC linear accelerators.

The reasons for the Q degradation being still far from completely understood, investigating alternative coating techniques, such as arc coating in ultra-high vacuum (UHV), that can lead to different film properties, is therefore interesting. The main advantages of arc deposition over sputtering are the highly ionized state of the evaporated material, the absence of gases to sustain the discharge and the high energy (about 50eV) of atoms reaching the substrate surface. Its main disadvantage is the production of macroparticles (also called microdroplets) of the cathode material that become embedded in the film. This paper presents our recent results on the superconducting properties of arc deposited film samples and on the droplet problem. Details on the deposition systems are presented in another paper [3]

## RESULTS AND DISCUSSION

Results on Niobium films presented in the following sections are obtained analysing samples deposited on copper and sapphire substrates, at room temperature and by the UHV planar arc system. Samples are characterized by measuring their Residual Resistivity Ratio (RRR) and superconducting critical temperature (Tc), and by X-ray diffraction and Atomic Force Microscope (AFM) observations. Macroparticles content is investigated using optical and electron microscopes and a Labview based software.

### *Transport Properties*

Samples have been obtained with RRR ranging from 10 to 80 even for film thicknesses of a few thousand Å only, with excellent reproducibility. Such RRR is higher by more than a factor of 2 than usually obtained under similar conditions (same thickness and coating temperature) by magnetron sputtering (RRR usually between 5 and 10).

Critical current density ($J_c$) and superconducting critical temperature ($T_c$) of the films are measured using an inductive method consisting in passing a 1KHz alternate current through a coil placed on the sample surface. The coil serves both as exciter and as pick-up: a lock-in amplifier in fact records the third harmonic signal generated during the transition between the normal and the superconducting phase. Typical results, in good agreement with Nb bulk (Tc = 9.26K ΔTc ~ 0.01K) data, are shown in Fig.1.

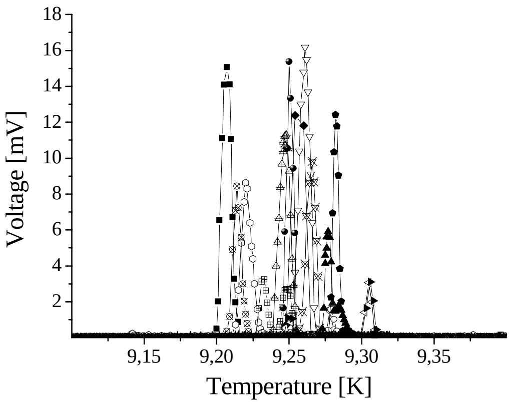

Figure 1: Transition curve of several Niobium films deposited on copper and sapphire. Tc is 9.25K within 0.05K and transition widths are smaller than 0.02K.

The critical temperature Tc is here defined as the temperature at which the transition starts, while the transition width is defined as the temperature variation to complete the transition. In our samples the width is very narrow (0.01 – 0.02K), a strong indication that the films are very homogeneous.

In some cases it was possible to observe a curve with two transitions at slightly different temperatures as shown in fig. 2. Two transitions are observed only in samples where a large number of macroparticles (covering more than ≈ 10% of the surface) is present and are interpreted as one corresponding to the macroparticles transition at the Tc of the bulk (macroparticles are molten pieces of the pure niobium cathode) and the second as that of the niobium film, occurring at a slightly lower temperature as a consequence of stress present in the film [4]. As earlier said, the difference in critical temperature with respect to bulk is much less than in the magnetron sputtering case (Tc about 9.5K) [5], indicating a less stressed structure, as also confirmed by X-ray diffraction measurements (see next section).

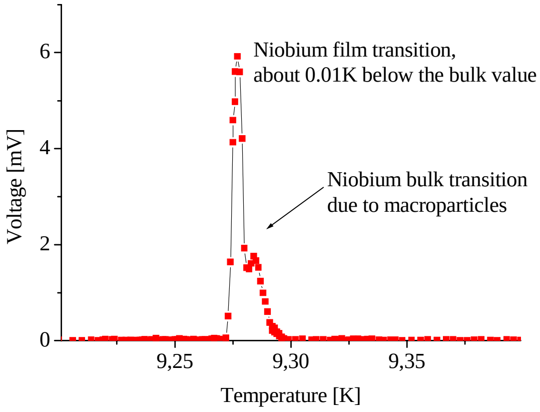

Figure 2: Detail of the superconductive transition for a sample with a large number of microdroplets measured inductively. Both transitions are very narrow and the overall transition is 0.02K.

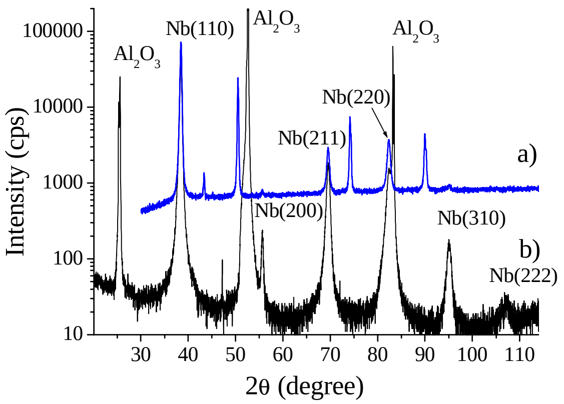

Figure 3: X-ray spectra of Niobium films on copper a) and on sapphire b). In the case of Cu substrate the high background signal is due to fluorescence. Films are in both cases preferentially growing along the (110) plane, but other orientations are also present.

## *Structural Analysis*

The Nb film structure has been analysed using X-ray diffraction and atomic force microscopy. X-ray diffraction spectra (see Fig. 3) are collected using Cu–$K_\alpha$ radiation with filtered $K_\beta$ line, in the θ-2θ configuration. Due to the relatively small film thickness X-rays are not all stopped in the Niobium film but also penetrate the substrate. When the substrate is copper they produce fluorescence, thus increasing the background noise. Since the main difference in X-ray spectra collected on Nb grown on the two different substrates is the background level of noise, we have so far concentrated on niobium on sapphire. A number of copper grown Nb films have nevertheless been produced for comparison purposes.

The position of the maximum in the diffraction peak is

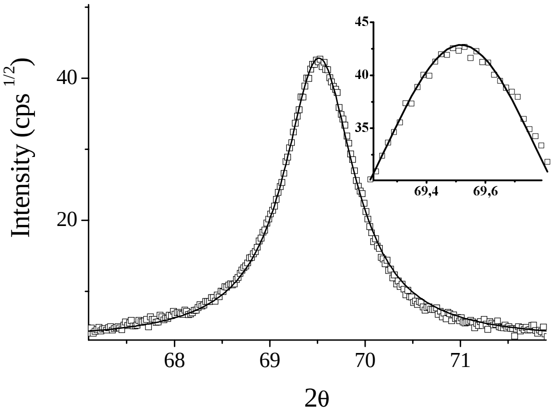

Figure 4: Example of measured niobium peak (211). Full line is the fit, the inset showing a detail of the region around the maximum. The maximum θ value is obtained from the fit.

obtained by fitting the curve with an appropriate function (see Fig.4). The lattice parameter is then obtained by extrapolating the lattice parameter values obtained at various different angles to $2\theta = \pi$ . The results range from 0.3306 to 0.3318nm, indicating a much lower stress than observed in Niobium deposited by magnetron sputtering on copper substrates [5-7], a result in agreement with $T_c$ measurements. Also the width of the diffraction peak is slightly narrower than in the sputtering case, an indication of larger and/or more ordered grains. This result is also confirmed by Atomic Force Microscope pictures showing an average niobium grain size of 200nm (see Fig. 5). The roughness of Nb samples deposited on copper is comparable to that of the copper substrate itself while that of films on sapphire is much smaller. This clearly indicates that, on copper, film roughness is determined by the substrate. When using a magnetic filter against microdroplets (see below), the roughness of arc deposited Nb samples on sapphire is observed to be of the order of few tenth of a nm, comparable to that of niobium sputtered films deposited on the same substrate

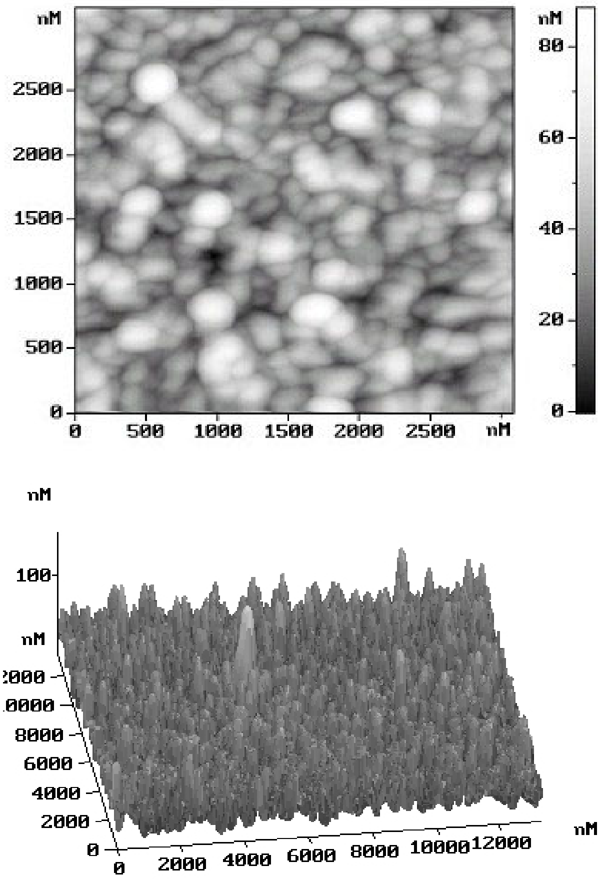

Figure 5: AFM picture of a niobium film deposited on sapphire. A) Nb grains are visible (average dimension 200nm). B) tridimensional view of the surface: a small microdroplets is visible near the center of the picture.

## *Macroparticle Analysis*

The main disadvantage of arc coating is, as above mentioned, the production of microdroplets of high purity molten Nb that become embedded in the growing film. While not contaminating the film, the droplets increase its surface roughness and become possible sources of field emission. The number and size distribution of micro droplets in our films has been studied, in both unfiltered and filtered planar arc setups, by optical and electron microscopy and by roughness measurements.

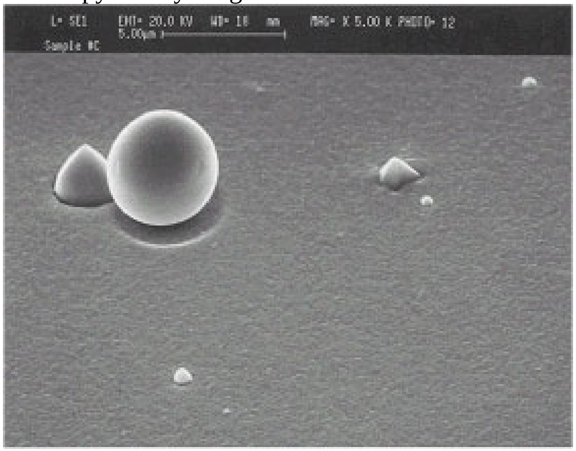

Figure 6: SEM picture in tilted view for a non filtered niobium film deposited on sapphire.

In particular, we have investigated the droplet distribution dependence from discharge parameters, at constant film thickness. On each produced sample between 5 and 10 pictures have been taken at different locations, using a 500X magnification optical microscope. Photographs are analyzed using a LabView computer code. The number of observed droplets is thus in fact a measure of the droplet surface density. Two situations, the normal one (Fig. 8) and one when thermal conductivity between Nb cathode and copper support was much reduced, leading to a much higher average cathode operation temperature (Fig. 9), have also been compared.

Figure 7: An optical image of the niobium film surface used for microdroplet analysis. Microdroplets show up as dark spots.

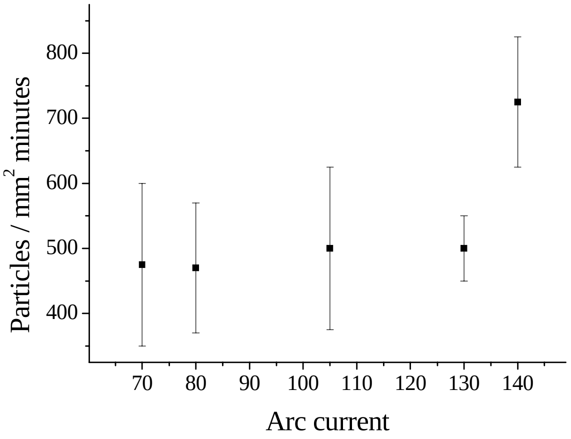

Figure 8: Number of droplets vs. current when cathode and coolant are in good thermal contact: up to 130 A there is no evidence of current dependence.

An interesting correlation emerges between number of droplets and average cathode temperature. A summary of results is shown in Fig.8. For arc currents IA ≤ ≈130 A there is no evidence of correlation between IA and the number of droplets per unit deposition time. Since the deposition rate increases linearly with IA and, as earlier said, all compared samples have the same thickness, the data indicate that, under normal cathode temperature conditions, the minimum number of micro droplets for a given thickness is reached at around 130 A. A rough estimate of the steady state cathode surface temperature T(C), obtained by comparing the computed power dissipation by thermal radiation and by cooling to that deposited on the cathode by the arc current, gives T(C) ≈ 2000K at an arc current of 140 A. On the other hand when the thermal contact between the cathode and the cooling system is poor, irradiation is the main source of dissipation and the cathode surface can reach much higher high temperatures, close to the Nb melting point.

In such a case one sees a clear dependence of the number of micro droplets on IA. Also note that the number of droplets at 60 A in Fig. 9 is comparable to the 140 A value in Fig. 8, an indication of a higher cathode temperature. Moreover, in the case of Fig.9, assuming power is mainly dissipated by radiation, one estimates a cathode temperature at IA=60 A of ≈ 2000 K, in remarkable agreement with the 140 A value obtained from the data of Fig. 8. The present interpretation is that the number of micro droplets emitted per unit time is independent from the arc current as long as the cathode surface temperature stays below ≈2000 K, while it increases fast with increasing IA for higher cathode temperatures.

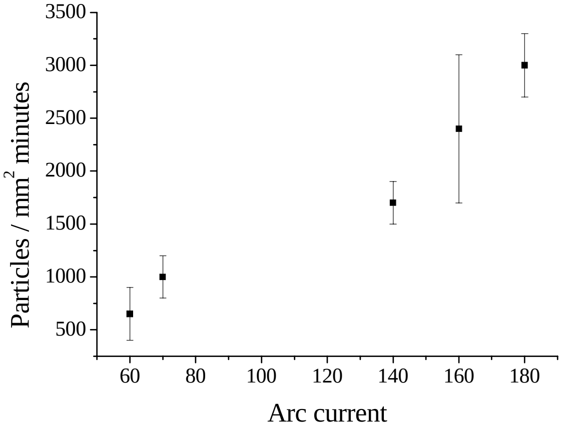

Figure 9: In the case when the thermal contact is bad so that the cathode heats up rapidly, the number of droplets increases rapidly with increasing current.

Last, samples produced with the filtered arc apparatus show a dramatically reduced number of droplets, for any given condition, at the expense of the deposition rate being reduced by a factor of ≈ 5 with respect to that of the unfiltered arc. A quantitative datum has not been obtained so far because the number of droplets in

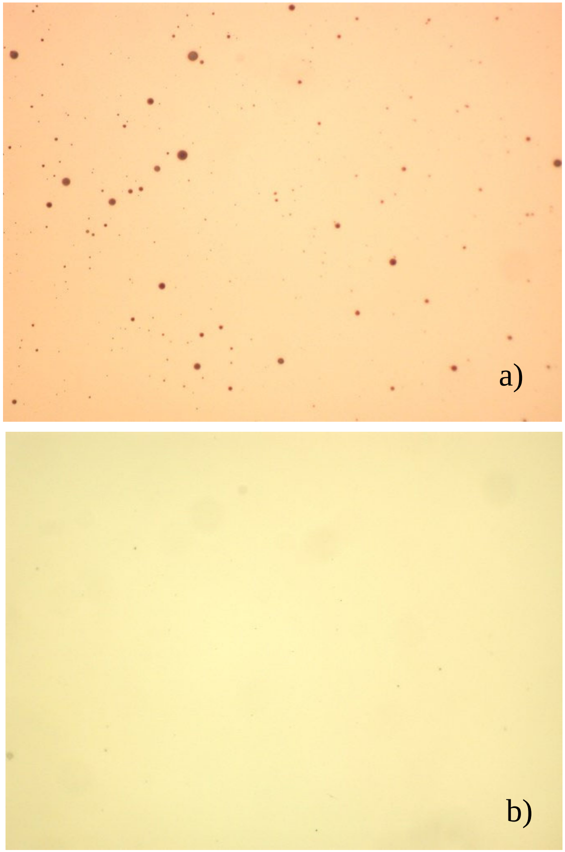

Figure 10: Optical image of the sample surface: a) planar configuration without a filter, b) planar arc source with filter for microdroplets.

the observed field is often below the detection threshold, as shown in fig. 10.

## CONCLUSIONS AND FUTURE PLANS

We designed and built a planar arc source working in UHV conditions to study the deposition of super conducting Nb films. The results obtained with sample Nb films are promising in that films with "bulk-like" properties were produced.

Niobium films produced has higher RRR, and larger grain size compared to sputtered niobium films deposited at the same temperature. Also, films produced by UHV arc discharge are less stressed and more randomly oriented compared to the sputtered one. These differences are interpreted as resulting from the higher energy of the niobium ions impacting on the growing film that favours a more regular arrangement on the film surface.

Study of the surface density of micro droplets as a function of arc currents shows that under normal operating conditions the droplet surface density does not depend on arc current up to ≈130 A. A filtered arc was used to produce quasi microdroplet-free samples: the number of droplets mostly falls below the instrumental detection limit.

A linear cylindrical arc is under development and filtering system for it is also being developed.

Possible effects of droplets on field emission, as well as procedures to remove them (high pressure water rinsing and others) after deposition, will be the object of future study. Pulsed rather than dc operation is also to be investigated.

## REFERENCES

[1] D.M.Sanders, J. Vac. Sci. Technol. A 7, (1989) 2339

[2] C. Benvenuti, S. Calatroni, P.Darriulat, M.A.Peck, A.-M.Valente, C.A.Van't Hof, *Physica C*, **351** (2001) 421-428

[3] S.Tazzari et al., Proc. XI Workshop on RF Superconductivy, Set 2003, Travemünde, in print

[4] G. Heim and E. Kay, J. Appl. Phys, 46 (1975) 4006

[5] R. Russo and S. Sgobba, Particle accelerator 60 (1999) 135-145

[6] C. Benvenuti, S. Calatroni, I.E. Campisi, P. Darriulat, M.A. Peck, R. Russo and A.-M. Valente, *Physica C,* **316** (1999) 153-188

[7] C. Benvenuti, S. Calatroni, P.Darriulat, M.A.Peck, A.-M.Valente, C.A.Van't Hof, "*Influence of the nature of the substrate on the growth of superconducting niobium films*" internal note CERN–EST 2000-5 (SM)